

**Oklo: Des reacteurs nucleaires fossiles  
(Oklo: The fossil nuclear reactors).  
Physics study (R Naudet, CEA)  
- Translation of chapter 6, 13, and  
conclusions**

V O Oversby

VMO Konsult

September 1996

**SVENSK KÄRNBRÄNSLEHANTERING AB**

*SWEDISH NUCLEAR FUEL AND WASTE MANAGEMENT CO*

BOX 5864 S-102 40 STOCKHOLM SWEDEN

PHONE +46 8 665 28 00

FAX +46 8 661 57 19

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FOSSILES (OKLO: THE FOSSIL  
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PHYSICS STUDY (R NAUDET, CEA)**

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# **Oklo: Des Reacteurs Nucleaires Fossiles (Oklo: The Fossil Nuclear Reactors)**

**Physics study**

*Roger Naudet*

**Commissariat à l'Énergie Atomique**

## **Translation of chapters 6, 13, and Conclusions**

*V. O. Oversby*

**VMO Konsult**

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## CHAPTER 6 - STUDY OF CRITICALITY

The study of neutron balance has shown that this depends on many factors and that there are many ways of reaching the critical state, that is to say obtaining a stable chain reaction. For example, the concentration of uranium in the ore is a fundamental parameter, but a very rich ore is not necessarily favored if the deposit is too thin or there is not enough water. There are not, therefore, limits imposed on each parameter (although there are, in general, thresholds); it is the combination of the ensemble that must be considered.

In the reactor zones, criticality was achieved throughout, at any time, in all circumstances, and despite the great diversity of characteristics and situations: to seek how this was obtained and maintained for Oklo is the fundamental physical problem. One will only begin this study in the present chapter: for the time being one leaves to the side the problems of equilibration of activity during functioning (the subject of chapter 9), and the spatial interferences between reactors (chapter 10 studies). One considers here only the initial and final states of isolated reactors. We also limit ourselves to analysis and comparison of situations, leaving for later interpretations concerning the origin of the situations and the progress of the phenomenon in its entirety.

In the first part of the chapter, we approach the problem from a very general manner, only taking into consideration a small number of parameters in order to establish equivalences and ranges of compatible values. In particular, this allows comparison of the initial and final conditions of the reactors. On the other hand, after supposing in the first case that there was, from the beginning, the argillaceous gangue and the geometry we now have, we take into account that these reactions were able to start in sandstones that were more or less desilicified, from which we get new equivalences.

We then examine more concretely a certain number of situations typically encountered at Oklo, in order to determine if we can justify criticality in a homogeneous manner, or if we must admit, on the contrary, that it was reached under conditions that were not identical everywhere.

Moreover, it is interesting to inquire to what extent criticality could have been reached in uranium ore deposits with origins younger or older than those of Oklo. A reflection on the phenomenon of "natural reactors" is discussed in the appendix to the chapter.

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# 1 PARAMETRIC STUDY IN CLAY MEDIUM

## 1.1 CHOICE OF PARAMETERS - DEFINITION OF AN "EQUIVALENT" THICKNESS

The three principal parameters that characterize the composition of the ore deposits for a given environment are the relative amounts of uranium, of gangue, and of water, as well as the neutron capture capacity of the gangue. In practice, we use the parameters  $t$ ,  $x$ , and  $m$  defined in chapter 5, and reviewed briefly here. " $t$ " is the concentration of uranium (or grade) of the ore, translated to present day (to adjust for radioactive decay). " $x$ " represents the neutron poisons (rare earth elements, boron) and more generally, an increment of capture capacity of the gangue in comparison with a "standard" composition (in the following material, we keep the composition given in the preceding chapter, which represents suitably the average of zones 1 and 2). Finally, " $m$ " defines the "free volume" occupied by water ( $m/(1+m)$  is the porosity of the deposit). The total quantity of water present is fixed when, in addition, the fraction of structural water in the clay is given and the densities are given.

We will only consider the initial and final conditions, and assume that the temperature is still - or again - that of the environment. In the following, we adopt, as in the reference example,  $T = 160^{\circ}\text{C}$ ; the margin of error in this value (which might need to be lowered by about  $20^{\circ}\text{C}$ ) is of little consequence with respect to criticality. We will also not vary the parameters related to heterogeneity, the "standard" hypotheses defined in the preceding chapter being considered as representative of the mean. Finally, in nearly all the calculations, we adopt an age of 1950 MA, considered as the best value; we examine only the effect of variation of this parameter, this question being reviewed more thoroughly in the appendix.

Two other parameters will eventually be used: in the initial condition, we consider either a deposit completely argillaceous - that is with a gangue such as we now see - or a sandstone, more generally a sandstone in the process of being desilicified - that is a deposit with some quartz remaining. There is then a supplementary parameter " $Q$ ", which designates the ratio of quartz to clay in the gangue. In the final condition, the irradiated deposit will be characterized by the parameter " $e$ ", defining the isotopic depletion of the uranium (transposed to present time). This result implies a calculation of the residual poisons and fission products accumulated at the end of reaction.



There remains, finally, to characterize the dimension of the multiplication medium and then the escape of neutrons: a certain number of conventions and simplifications are needed to represent this complex ensemble of results by means of a single parameter. This is what we will now examine; this will also be the occasion to note that the geometry of the reactors at Oklo poses certain problems.

### **Simplification of the geometry**

To proceed in a simple manner, it would be necessary to suppose that the multiplication medium is homogeneous, that it has a definite geometric form, and that it is surrounded by a void. In reality, one is very far from this abstract concept; the characteristics of the terrain vary in all respects without precise limits. We can in the first instance adopt the model of a "flat cake reactor" in which we suppose the escape of neutrons to be important only in a single dimension. The distribution revealed along a core that crosses a reactor zone is then considered to represent the small dimension and we assume that the properties in the core are uniform in the other two dimensions. In reality it is not at all like this, of course, and the exchanges of neutrons with the lateral environment is often important. The situation will be different depending on whether the core is surrounded by rich ore or by poor ore and whether it is in the center of the reactor zone or on the edge.

Another difficulty comes from the fact that vertical drill cores cut the strata obliquely to the bedding. (It is the same problem with samples taken from horizontal faces of a mining face). To be consistent with the hypotheses used, one must multiply all the distances in the drill core by the cosine of the dip of the strata. This "recipe" is hard to apply because the dips are poorly known, and moreover the dips are poorly defined because the beds are deformed and the thicknesses of each partial environment is not constant.

Another adaptation is needed to recover the geometry of the reactors; since that time, there has been a compaction of the terrain. We have seen that the spectral indices suggest that the porosity at the time of reactions was much greater than today, which means that the ore mass occupied a much larger volume. In other words, in addition to the compression that accompanied the desilicification rather rapidly, there was an additional compaction. To evaluate the ratio of corresponding volumes, we must take as a reference the mass of the argillaceous gangue, which is assumed to be unchanged. This ratio, RV, consists in the general case of two factors, one a function of Q (cf. table 10 of the previous chapter), the other calculated for  $Q = 0$ . [Note: Q is the weight ratio of quartz to clay in the rock before reaction]. The second factor, proportional to  $(1 + m)$  is difficult to evaluate, because the densities of the minerals are poorly known and it is difficult to measure the present porosity of the ore deposit in situ.

The theoretical densities are the following: uraninite, 10.9; galena, 7.5; quartz, 2.65; phyllosilicates, 2.6 to 2.85 depending upon composition; but

the real densities are always less than the theoretical values because the crystals are imperfect. In particular, it is probable that Oklo uraninite, which has lost a good part of its original matter after it formed (the majority of lead formed by decay having been lost by migration), has a rather low present-day density. A series of measurements were made of the density of ore samples as a function of their uranium content, but these measurements are not precise, giving results that are dispersed and difficult to interpret. One finds densities are systematically lower when samples are from cores rather than from blocks extracted from the quarry, which leads us to believe that the coring device on breaking up the ore has caused an abundance of voids, giving an apparent porosity that is much greater than the in situ value.

We have adjusted all density data from core samples using the following formula:  $1/d = 0.50 - 0.44t$ . Even if one uses rather low values for the mineral densities, this adjustment leads to apparent porosities of at least 20%. On the other hand, direct measurements of porosity on rocks - consisting of measuring the weight loss in an autoclave of an ore sample originally saturated with water - gives values ranging from 2 to 12%. (It should be noted on this subject that the porosity is larger in the faciès-piles and the argiles de pile - that is the desilicified old sandstones - than in the sedimentary argillites (pélites), which seems to show that even after 2 billion years the compaction of the former is not yet complete.) Comparison of the numbers [cores versus quarry blocks] seems to show that the breaking up due to the coring has increased porosity on the order of 10 to 15%.

It is not sufficient to just evaluate the ratio RV; what we are interested in is the thickness of the reactor "cake". We can suppose (but it is not strictly justified) that the compaction has occurred principally in the direction perpendicular to the stratification. On the other hand, the increase in porosity due to coring occurs essentially in the longitudinal direction. (One sees effectively an elongation of the cores in comparison with the sides revealed in the hole from the coring.) We are led, therefore, to multiply the lengths revealed along the boreholes by the ratio RV defined by using the densities from the cores. (This ratio is smaller than that obtained from the in situ ores, but it allows us to take into account the elongation of the core).

For the density of the ore at the time of the reactions, we use 10.5 for uraninite, which it was when first formed, and 2.5 for quartz and phyllosilicates. (Even though these values are debatable, the important issue is to use the same values when calculating the area of migration). By comparison with the formula given above, one finds then the following values for RV as a function of t, for  $Q = 0$  and  $m = 0.6$  (recall that RV is proportional to  $1 + m$ ).

t	10%	20%	30%	40%	50%	60%
RV	1.206	1.213	1.223	1.234	1.248	1.267

In other words, for this value of  $m$ , we must dilate the thicknesses by about 20 to 25%, which we can see is a very important correction.

In the end, we are led to multiply all lengths taken from boreholes by two factors:  $\cos \alpha$  and  $RV$ , both of which are poorly known. *This is, certainly, the most important source of uncertainty in the evaluation of criticality at Oklo.*

### Gain from the reflector

At this point, in order to begin the parametric study, it is desirable to characterize simply the corrected distribution of concentration of ore. We note that there generally is from place to place in the very rich ore a very steep decrease in ore grade. We can then, very roughly, consider that there is a certain thickness of rich ore, in which we can homogenize the uranium concentration and isotopic depletion, surrounded by a poor ore zone. This poor zone, even when there is a coefficient of neutron multiplication much less than unity, influences the neutron balance, because it sends the neutrons back into the multiplication medium: one says that this plays the role of a reflector. The result is that the thickness of rich ore necessary for criticality is less than if the rich zone is surrounded by a void. The difference is called the "gain of the reflector".

It seemed interesting to do a systematic study of this parameter. It depends essentially on the properties of the reflector - principally the amounts of uranium, water, and poisons - and much less on the properties and thickness of the multiplication medium. The study was done using the CRITO code, which we will discuss in much more detail in the next chapter. This code, when combined with a series of media of proscribed thicknesses, allows us to calculate a  $k_{\text{eff}}$ , which is the number by which one must divide the  $k_{\infty}$  of all the media so that the ensemble will be critical.

One proceeds in the following manner: one takes a reference medium (for which one knows the  $k_{\infty}$  and the  $M^2$ ) and a series of reflectors. [Note:  $M^2$  is the "migration area". See p. 231 for further discussion.] One chooses each time the thickness  $H$  of the multiplication medium so that  $k_{\text{eff}}$  shall be near unity, the reflector being much thicker on both sides.  $H_e$  [the critical thickness] is then given by

$$k_{\infty}/k_{\text{eff}} = 1 + M^2\pi^2/H_e^2$$

where the gain  $\delta = \frac{1}{2} (H_e - H)$  (the gain is defined for each of the interfaces); one verifies that this quantity varies only slightly when one changes  $H$ , and thereby  $k_{\text{eff}}$ . Results are given in Table 1 as a function of the properties of the reflector -  $t$  (uranium content),  $x$  (amount of poisons, as equivalent  $^{10}\text{B}$ ), and  $m$  (free volume occupied by water):

**Table 1. "Gain of the reflector" as a function of t, m, and x.**

	x = 50 m = 0.4	x = 20 m = 0.4	x = 50 m = 0.3
t = 2%	$\delta = 8.8$ cm	$\delta = 9.7$ cm	$\delta = 9.2$ cm
5%	9.4	10.6	9.9
10%	10.6	12.5	11.2
20%	13.9	17.9	
25%	17.4		

The gain of the reflector increases with t, and decreases with x and m (the reflector is better if there is more uranium, less poison, and less water). One sees, nevertheless, that the gain is insensitive to m (there are two effects from m acting in opposite directions), and that the gain stays about the same, 10 cm, in a range of variation that is rather large for t and for amounts of poison (t = 2 to 10% and x = 20 to 50 ppm <sup>10</sup>B). One can, therefore, use this value as a good order of magnitude. For example, for the reference environment from the preceding chapter, for which  $H_e = 63.1$  cm, a real thickness of 43 to 45 cm for the region that would be critical would be sufficient, even though it were surrounded by relatively poor ore.

When the uranium content of the reflector increases more, so that its  $k_{\infty}$  approaches unity, the gain increases much more quickly. In particular, if we increase t to 25%, we only need a thin layer of reference material for it to be critical, which is easy to understand because the reflector itself is close to being critical.

This concept of reflector gain is useful for simple characterization of a distribution, but one must be able to define without too much ambiguity the thickness of the rich zone and its average characteristics. One could also proceed in a more rigorous fashion, even if the rich zone is not clearly delimited. For this one uses the CRITO code to treat the real distribution (for t and e) by choosing for the other parameters (x, m, etc.) values more or less identical in all the partial environments. Then one deduces a  $k_{\text{eff}}$ . The BINOCLE code then allows us to associate average values to the whole environment ( $t_{\text{av}}$ ,  $e_{\text{av}}$ ) which represents the rich zone, and for the same choice of the other parameters, find the value of  $B^2$  which leads to the same  $k_{\text{eff}}$ . Since  $B^2 = \pi^2/H_e^2$  we have by definition that  $H_e$  is the equivalent thickness of the corresponding medium. Although the result depends a little on the choice of parameters, this is a rather precise method of globally characterizing the distribution.

In most of the boreholes that cut the reactor zones 1 and 2, the thickness of rich ore is between 30 and 60 cm. The dip for those two zones was between 45 and 30°, from which the  $\cos \alpha$  is 0.7 to 0.85. On the other hand, from the values of  $m$  deduced from the spectral indices,  $RV$  is of the order of 1.20 to 1.25. From this one concludes that the equivalent thicknesses are most often between 50 and 75 cm.

## 1.2 "INITIAL" CRITICALITY. EQUIVALENCES BETWEEN PARAMETERS.

In the following discussion, one considers the media to be defined by four parameters:  $t$ ,  $x$ ,  $m$ , and  $H_c$ . One presumes the uranium to be unirradiated, so  $e = 0.72$ , and further that  $Q = 0$ . This last point is not really a restriction, because the results remain valid even if the gangue contains quartz, so long as we adjust the values of  $t$  and  $x$ . In any case, we will consider more specifically the case of criticality in sandstones in the following section.

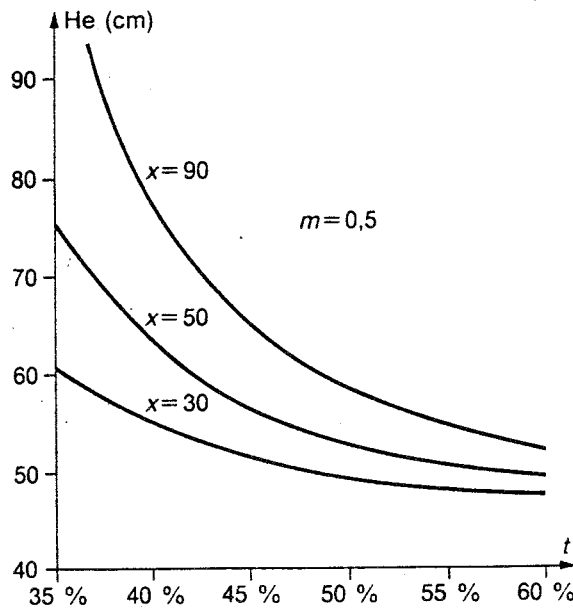
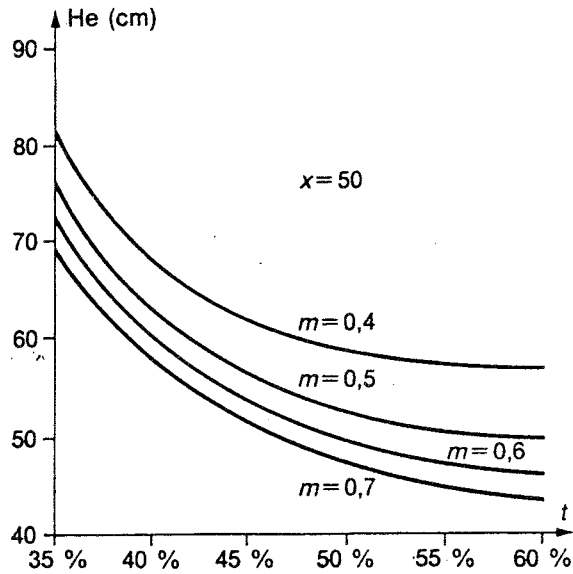
For a given  $m$  and  $x$ , the criticality condition imposes a relationship between  $H_c$  and  $t$ : the thickness of the rich mineral zone must increase as the ore content decreases. Figures 1 and 2 illustrate this relationship for different combinations of  $m$  and  $x$ .

First let us consider the reference case from the previous chapter ( $t = 40\%$ ,  $m = 0.5$ ,  $x = 50 \text{ ppm } ^{10}\text{B}$ ,  $H_c = 63.1 \text{ cm}$ ). As one increases the uranium content, one can decrease the critical thickness to 50 cm, but not more. Nothing is gained for increasing uranium above 55%. For the inverse problem, as one decreases the ore grade,  $H_c$  increases very rapidly, with more than 120 cm for  $t = 30\%$  and if the grade is less than 28%, criticality cannot be achieved even for an infinite thickness ( $k_\infty \leq 1$ ). One sees from this that the condition for criticality carries with it a compromise between the properties of the rich ore zone (average concentration and thickness), and that there is a threshold for each of these. For the chosen values of  $x$  and  $m$ , the U concentration must be above 28% irrespective of thickness, and  $H_c$  must be above 50 cm, regardless of the ore grade.

In figure 1, we have varied  $m$  between 0.4 and 0.7, while keeping  $x = 50$ . We have seen previously that measurements of spectral index give most often values of  $m$  between 0.5 and 0.65; the range of compatible values is therefore rather narrow. By contrast, the conditions for criticality depend strongly on the choice of  $x$ , as shown in figure 2, while the value of this parameter is not known *a priori*.

The rare earths present in the ore have a neutron capture equivalent of about 15 ppm  $^{10}\text{B}$ . If we suppose that the remainder of the poisons are due to boron itself, varying  $x$  from 25 to 75 corresponds to assuming a content of

50 to 300 ppm of natural boron in the gangue. This is essentially the order of magnitude that one finds in the present ore deposit; other considerations lead to values of the same order, as we shall see later. In any case, one notes that this level of poisons is compatible with an initial condition for criticality.



**Figure 1 & 2.** Critical thickness of the rich layer as a function of uranium content for different values of the amount of water (m) and poison (x).

Table 2 demonstrates this point more precisely: we give the critical  $x$  value for different combinations of  $t$  and  $H_e$  (while keeping  $m = 0.5$  constant). For zones 1 and 2, the average ore grade for the rich zone is always between 35 and 50%; we have said earlier that the values for  $H_e$  most often are between 55 and 75 cm. We see that criticality is justified for levels of poisons of the order of magnitude indicated.

**Table 2.** Critical value of  $x$  (ppm equivalent of boron) for different combinations of the ore grade and thickness of the rich zone.

	$t = 35\%$	40%	45%	50%
$H_e$				
50 cm	6.6	16.4	25.8	34.5
60 cm	28.8	43.7	59.5	76.5
70 cm	43.1	61.2	81.1	103.3
80 cm	52.7	73.0	95.7	121.5

These results presume that the age of the reactions is 1950 MA; the critical thickness would be thinner if the age were older. The advantage of an older age is more apparent when the ore grade is lower. Increasing the age by 100 million years allows reduction of the critical thickness by 3.4 cm when  $t = 50\%$  and by 7.3 cm when  $t = 40\%$ . (The gain is 2000 pcm [2.000%] in the first case and 2700 in the second; this is equivalent to 14 to 15 ppm  $^{10}\text{B}$ ). One sees, then, that the effect of age is important. One can say that, for the average properties of the rich zones at Oklo, *changing from 1800 to 2000 MA allows roughly a doubling of the amount of poison compatible with criticality.*

**Remark:** - It must be emphasized that the critical value of  $x$  has the meaning of a content of poison only in the case where a full range of conditions are met. We have assumed that the composition of the gangue is that of the argillites and then that the desilicification is complete, that the initial porosity is the same as the average during the course of the reactions (which is calculated from the spectral index), and that the compaction is stabilized, that the temperature is that of the ambient environment, and finally that the neutron exchanges with the environment are equilibrated. In reality, these conditions are never simultaneously fulfilled: either the reactions began in sandstones, or they resulted from a propagation phenomenon, that is to say that they began in an environment that was not necessarily stabilized, with a temperature generally higher than ambient, and involving exchanges of neutrons that had no reason to be equilibrated. This is, then, only a fictitious manner of defining the initial situation. We put ourselves into the conditions

that would have occurred if the reactors started in an isolated manner in their final geometry.

From this point of view, the critical  $x$  retains in any case an important significance: it is a reserve of reactivity that is necessary so that the reactors can function. Whether this reserve is invested in the form of neutron poisons, or in supplementary capture by silica, or in the form of temperature, it is still "capitalized on" in response to the effects of isotopic depletion of the uranium and the accumulation of fission products. The preceding conclusion can be stated in another way: it appears that at Oklo (at least in zones 1 and 2) initial criticality can be supported taking into account a suitable reserve of reactivity (this last term will be elaborated upon in the next section.)

### 1.3 CRITICALITY AT THE MOMENT WHEN THE REACTIONS STOPPED

It is easier to discuss the final condition of the reactors precisely because one avoids a great part of the uncertainties which weigh upon the conditions at startup. For one thing, it is less difficult to suppose that at the end of the reactions all the quartz had dissolved. Additionally, the neutron poisons at that time had largely been destroyed by the neutron flux (at least to the extent that one has the right to suppose that there had not been partial renewal by geochemical migration); the poorly known level of the initial value of the poisons then has much less importance. Finally, because the effect of a lowering of temperature is always favorable, the reactions continuing until the temperature has relocated it equilibrium value, it is justifiable in this case to reason using that temperature. As for supplementary parameters,  $e$ , which characterizes the final condition, is adequately known because one can use measured values of the isotopic depletion.

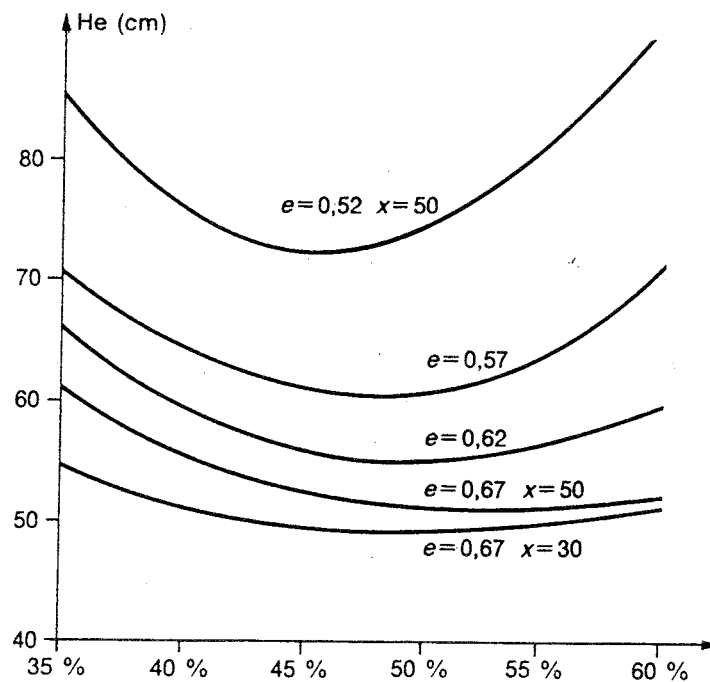
Figure 3 shows how the critical thickness varies as a function of the mean uranium content of the rich zone for different values of  $e$  (keeping  $m = 0.5$  constant). This time, since  $x$  represents the initial concentration of boron, the effect of variation of this parameter is much less as the value of  $e$  increases (for  $t = 40\%$ , the  $\delta H_c$  corresponding to 20 ppm  $^{10}\text{B}$ , which is 4.1 cm for  $e = 0.62$ , 1.1 cm for  $e = 0.57$  and 0.6 cm for  $e = 0.52$ ).

One sees that for a given value of  $e$ , an increase in ore content causes only a small decrease in the critical thickness and can even lead to a larger value. The reason for this is that neutron capture by the fission products is more important in the rich ores (cf. chap. 5, §2.7). It is for this reason that for the given value of  $m$  and for  $e = 0.52$ , capture by fission products represents 4.7% for ore with  $t = 35\%$ , and 9.5% for ore with 55%. In addition, the reactivity liberated by capture in the poisons for a given initial value of  $x$  is smaller if the ore is richer. *Because of this, in the region of normal concentrations, isotopic depletion of uranium depends most on the thickness of the rich zone. This is essentially what we see at Oklo.*

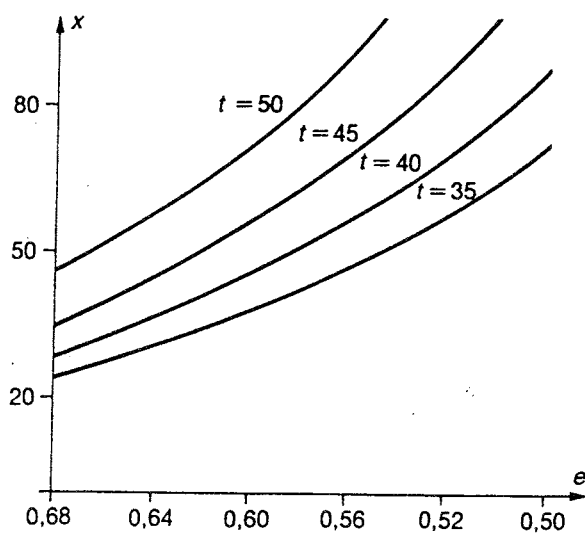
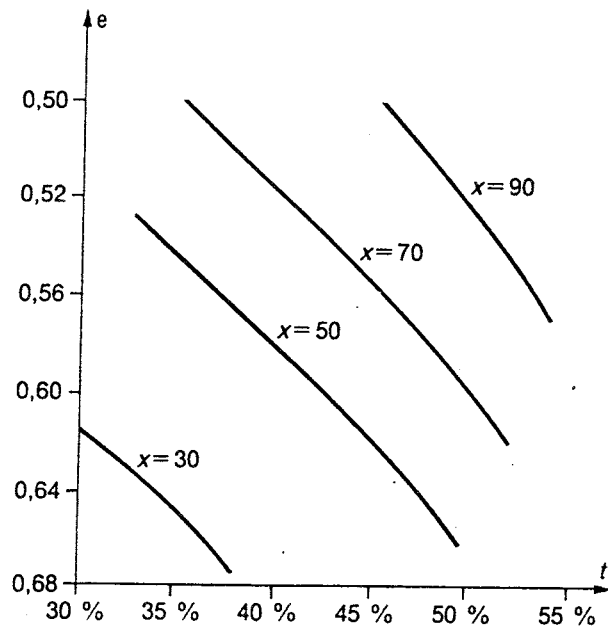


In zones 1 and 2 at Oklo, the average isotopic concentration of  $^{235}\text{U}$  in the rich zones is most of the time between 0.67 and 0.57% and does not go below 0.52% except in rare circumstances. Figure 3 shows that these values of the levels of irradiation are compatible with the characteristic  $t$  and  $H_e$  for the rich layers that are found in these zones. *It seems that for now that criticality in the final conditions is properly justified.* This will be shown in a more precise fashion in section 3 by examining in detail a certain number of particular distributions.

As we have seen above, there is an relationship between the critical value of  $x$  for the initial condition, which represents a reserve of activity, and the  $e$  in the final condition: figures 4 and 5 illustrate this relationship.



**Figure 3.** Critical thickness as a function of ore content for different values of the isotopic depletion



**Figures 4 & 5.** Relationship between isotopic depletion, the uranium content of the rich zone, and the critical value of  $x$ .

We worked in the following way: when one assumes  $t$ ,  $x$ , and  $m$ , one can deduce for the initial situation a value of  $H_e$  for criticality. There also exists a second value of  $e$  (in addition to 0.72) for which criticality is possible, everything else remaining constant (particularly the same value of  $H_e$ ). Keeping  $m$  in all cases at 0.5, there is for each value of  $t$  a relationship between  $x$  and  $e$ .

Figure 4 shows  $e$  as a function of  $t$  for different values of  $x$ . The form of the curves illustrates the remark made above: for a given  $x$ , one attains a level of irradiation that is smaller when the ore grade is richer. Figure 5 gives directly the relationship between  $x$  and  $e$  for different values of the ore grade.

We see, in particular, that for a value between 40 and 45%, *the initial reserve necessary to attain criticality for an isotopic depletion going from 0.67 to 0.54% ranges from 35 to 75 ppm boron 10*. This is another way to justify the order of magnitude of equivalent poisons in the initial conditions.

## 2 CRITICALITY IN THE SANDSTONES

One can pose two separate problems. On the one hand, it is interesting to study the comparison of the initial condition for a reactor for which desilicification is incomplete to that for the case considered above. This is accomplished by increasing the field of our investigations through the introduction of supplementary parameters. On the other hand, it is necessary to consider the problem of the initial starting of the reactors, that is to say, the achievement of criticality in the rich sandstones as they existed "naturally" at Oklo before they were modified by the convection currents associated with the nuclear reactions.

### 2.1 SANDSTONES IN THE COURSE OF DESILICIFICATION.

We saw in section 2.3 of the preceding chapter that in comparison to an ore that was completely converted to clay, the neutron balance of a sandstone in the process of desilicification is more or less drastically altered for the worse by the effects of the silica, but by way of compensation, to the extent that compaction is not instantaneous, it can benefit through the presence of supplementary water (characterized by the parameter  $ES$ ). One has said also that the escape of neutrons is different; it is this point that we need to examine more closely now.

The sandstones occupy a larger volume, and one knows that the evolution of the neutron escapes depends on the way in which compaction is achieved. We have seen, in particular, that if we assume that compaction is only in the dimension perpendicular to stratification, the compaction carries with it an increase in the amount of neutron losses, despite the decrease in the migration distance. We must remember that this is an extreme hypothesis, because observations of the terrain show that even though there has been effectively a profound decrease in the thickness of the beds caused by the reactions, there was also lateral sliding of strata. We will suppose this, nevertheless, in the following discussion.

In this hypothesis, one is led to multiply all the lengths revealed in a borehole crossing the reactor by a factor  $RV$ , which is a function of  $Q$  (or more generally,  $Q + ES$ ). But, to simplify the modelling, one can be content with, in the spirit of the preceding parametric study, transposing the "equivalent" thickness of the rich zone and supposing that the gain of the reflector stays in all case about 10 cm. This approximation is sufficient for qualitative analysis of the combination of these effects.

Table 3 gives an illustration of a particular case. Consider an initial condition (in the sense of the preceding paragraph) defined by  $t = 45\%$ ,  $x = 50$ ,  $m = 0.5$ , and  $H_e = 56.6$  cm, and contemplate different combinations of the parameters  $Q$  and  $ES$  assumed to represent successive stages to reach that initial critical condition. This does not involve, of course, studying the evolution of reactivity during a phase of functioning, which is necessarily more complex, but only to appreciate this particular effect, all other things being the same, as usual.

**Table 3.** Sandstone in the process of desilicification.

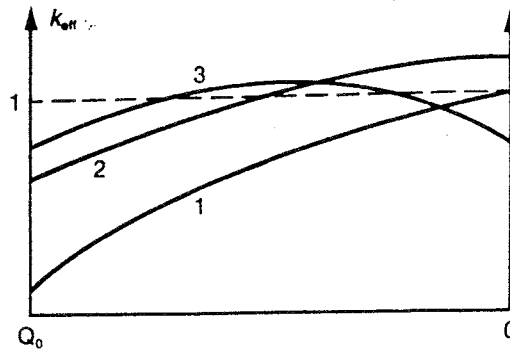
$Q$	$ES$	Porosity %	$RV$	$H_e$ cm	$M^2$ $cm^2$	$k_\infty$	$1+B^2M^2$	$k_{eff}$
8	0.5	11.8	4.64	189.8	209.8	1.0307	1.0527	0.9791
6	0.4	13.5	3.74	156.9	163.8	1.0571	1.0657	0.9920
4	0.3	16.2	2.84	124.0	132.6	1.0871	1.0852	1.0018
2	0.2	21.6	1.94	91.1	96.9	1.1234	1.1154	1.0072
0	0	33.3	1	56.6	53.7	1.1650	1.1650	1.0000

In this example, one has assumed that during desilicification there is "supplementary" water that is going to be reabsorbed. One admits that despite that the porosity increases constantly because of the decrease in total volume. The drop in  $H_e$  is very pronounced and since  $B^2$  varies with the inverse of the square of that quantity, the neutron escape factor,  $1 + B^2M^2$ , increases significantly despite the reduction in  $M^2$ . At the same time,  $k_\infty$  also increases significantly, but more slowly than it would if the medium water/uranium remained constant. Finally, we note that the evolution of both  $k_\infty$  and neutron loss are of the same order and that the coefficient of effective multiplication only varies slightly. In this example it even passes through a maximum in the course of desilicification: criticality is achieved before even half of the silica has been eliminated.

The effect of desilicification on the evolution of reactivity depends on many parameters and one can imagine a large number of cases. It is clear, in particular, that the amount of "supplementary" water and the manner in which it evolves during the processes could be common. On the other hand, the competition between the variation in  $k_\infty$  and that of the loss of neutrons depends on the values of  $t$  and  $H_e$ , respectively. In particular, if the thickness of the rich zone is too small, the penalty for neutron loss increases more during the course of desilicification. There can be situations where criticality is impossible in a zone that is completely converted to argillite, because the zone would be too thin, but is reasonable at an intermediate stage. This will

happen more easily when the ore is very rich because then, in the opposite sense, the penalty due to silica is smaller.

Figure 6 shows schematically the competition between  $k_{\infty}$  and neutron escapes (we consider only the variation in  $Q$ , the compaction being assumed to be done in a comparable manner in all three cases). With a large enough zone of ore with a rather low uranium content (curve 1), the elimination of silica is always favorable. When the ore is richer (curve 2), the situation initially is more favorable but the gain in reactivity is weaker. Finally, if one has a very thin zone with high grade, the effect of neutron escapes becomes most important at some time (curve 3) and the reactivity passes through a maximum.



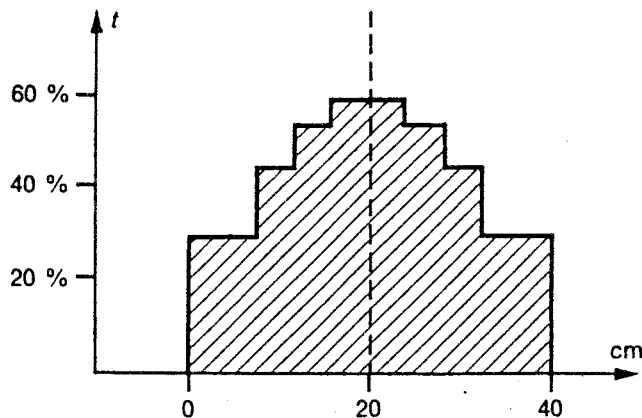
**Figure 6.** *Effect on reactivity of the elimination of silica.*  
*Curve 1. Thick zone, low uranium enrichment level;*  
*Curve 2. Thick zone, rich ore.*  
*Curve 3. Very thin zone, very rich ore.*

The result of these considerations is that initial criticality can be achieved at different stages of desilicification. In zone 1, for example, where one most often has ore with a moderate level of enrichment, a rather thorough desilicification must have occurred in order for the reactions to start. In zones 3 to 5, in contrast, one frequently finds thin zones of very rich ore where it is difficult to justify criticality in an ore that is totally converted to clay. Curve 3 shows that this can be achieved more easily before complete desilicification. Finally, in very rich zones, such as zone 2, criticality can be achieved during the course of desilicification, but in this case it continues to be advantageous to continue desilicification.

Actually, the preceding calculations only consider one aspect of reality, and they paint a simplified picture of it because we modelled using average characteristics:  $t$ ,  $Q$ , and  $RV$  are variable in the interior of rich zones, and the

partial environments can play different roles. We shall illustrate that with an example.

Consider the distribution represented in Figure 7: the rich zone has a thickness of 40 cm. It has in the center a thin portion with very high ore grade surrounded by ore with an average grade (30%). The CRITO code shows that criticality is not achieved when the ore is completely desilicified (with  $x = 50$  and  $m = 0.5$ ); it also is not achieved no matter what value of  $Q$  there is if one keeps the same ratio of water to uranium. On the contrary, one can obtain criticality, even for rather high values of  $Q$ , by calling upon a certain entry of water: in particular, the reactor is exactly critical when  $Q = 8.35$  and  $ES = 0.65$ . What is interesting is to see how the critical condition is achieved.



*Figure 7 Distribution of ore grade, t.*

Table 4 gives the thickness, the ore grade, and the  $k_{\infty}$  for each partial environment for the two extreme situations. We note that the environment with  $t = 30\%$  plays an important role in the neutron balance when the medium is completely argillaceous because then its  $k_{\infty}$  is close to unity. In contrast, when  $Q$  is large, this surrounding medium has a multiplication coefficient too low to be important and has no other use than a mediocre reflector. But, in this case, the very rich zone for which  $k_{\infty}$  remains much above unity, is sufficiently thick to assure by itself alone that criticality will occur. Criticality is then achieved in the two cases in a totally different way. We would not have discovered these results if we had used only the average grade of the rich ore zone; when  $Q = 8.35$  its  $k_{\infty}$  is less than unity and one would then conclude that the situation is considerably subcritical. The

situation becomes even more complex when we consider different rates of desilicification in different media.

**Table 4.** Characteristics of the media in figure 4 before and after desilicification.

Thickness	t	$k_{\infty}$	Thickness	t	$k_{\infty}$
4 cm	60%	1.184	15.4 cm	21.1%	1.097
4	55	1.175	17.0	16.9	1.070
4	45	1.130	19.5	10.5	0.986
8	30	0.981	44.5	5.5	0.772
$Q = 0, ES = 0, k_{\text{eff}} = 0.980$			$Q = 8.35, ES = 0.65, k_{\text{eff}} = 1.000$		

In addition, in the preceding we have assumed as in the preceding paragraph that the temperature is ambient; well, because desilicification is caused by circulating warm fluids, this is not a realistic hypothesis in the general case. There is another supplementary parameter to take into consideration. In particular, one can imagine that in the course of desilicification one passes through a stage where criticality would be started from the cold state, but where it was not achieved because of the effects of temperature. In this case, desilicification could continue and one must wait until a later lowering of temperature for the neutron multiplication coefficient to exceed unity.

One sees that the conditions in which reactions begin locally are very complex. One cannot, moreover, discuss them without considering the different couplings and the phenomena of propagation, problems which will be examined later. At this stage, we wish only to show that in certain cases, *criticality could be achieved during desilicification, sometimes with a rather favorable neutron balance, perhaps even more favorable than in a region completely turned into clay.*

## 2.2 FIRST STARTING OF THE REACTIONS

We have just seen that criticality could be reached rather frequently in ores that were still siliceous; the initial starting of the reactions, however, poses a different problem. One cannot take into consideration a preexisting substantial elimination of silica, accompanied by incomplete compaction. We must start with sandstones essentially undisturbed by hydrothermal circulations. On the other hand, we can assume that criticality was only achieved under certain favorable conditions, which we propose to identify exactly.



The hypothesis of ambient temperature is in this case totally justified. But there are still five parameters: in addition to their thickness and the level of poisons (which we will continue to link to the clay fraction), the sandstones are characterized by the ratio  $Q$ , by their uranium content  $t_g$  (function of  $Q$  and  $t$ ), and by their porosity  $p$  (a function of  $Q$  and  $m$ ). It is the porosity,  $p$ , which is the most significant, because it is inappropriate this time to use the parameter  $m$  from the argillaceous material as the reference.

What values of  $Q$  should we choose? In the "normal" sandstones of the bed  $C_1$  one finds a large range, from 3 to 1.  $Q$  depends on grain size (the lower values are in the fine sandstones), on the nature of the cement (which can be argillaceous or siliceous), and on their history, because they could have been subject to desilicification or alternatively to secondary resilicification. However, we note that the most mineralized levels in the  $C_1$  beds, and those that were preferentially the site of the "superconcentration" of uranium, are medium to coarse grained sandstones, for which  $Q$  is generally between 6 and 10.

On the other hand, it seems that the superconcentration, which was the result of circulation of water in the surroundings, was most often accompanied by some silica loss. In the rich sandstones, we find eroded quartz and we sometimes observe a lack of cohesion (or friability) which was caused by partial dissolution of the silica cement. We don't mean to imply that the loss of silica was significant from a quantitative standpoint, and we often find values of  $Q$  in the U-rich sandstones that are rather high. In reality, it is difficult to give a correct impression of the situation because the rich sandstones immediately around the reactor zones have had their compositions altered through the movement of silica associated with the convection currents. As for those that we find outside the context of the reactors, it is not sure that they are completely representative.

One does not know exactly, therefore, what the condition of the sandstones was at the time when the reactions first started. If we assume that natural desilicification was of little importance, it is necessary to assume a mean value of 8 for our modelling, but we can not exclude that in the process of natural circulations associated with tectonic processes, a significant fraction of the silica had been eliminated before the reactions started. In the following, we will consider a range of values of  $Q$  between 8 and 4.

This having been said, if for the same value of  $t$ , an increase in  $Q$  is unfavorable, it is the opposite if we keep the value of  $t_g$  of the sandstones the same, because then we keep the total amount of gangue constant and the quartz captures fewer neutrons than the clay. The problem is that we don't know  $t_g$ , but only the distribution of  $t$  in the reactor zones as they now are. Even though one can find isolated samples with ore content above 60%, the average value of  $t$  over a region of rich ore with sufficient thickness is only

very rarely above 50%, and in any case, never exceeds 55%. In other words, no matter what the value of  $Q$  is, there is an upper limit for the ratio of uranium/clay, and one cannot at the same time increase this independently of  $Q$  and  $t_g$ , the "authorized" region being limited by the relationship between these parameters. In the final analysis, it is better to keep  $t$  as the independent parameter.

From the neutronic point of view, one comment should be made concerning the poisons. We said that one has found in the present ore deposit a rather close correlation between the levels of boron and potassium, which is to say that the boron seems to be particularly associated with the illites. We know that circulations of warm water, at the same time as they desilicify sandstones, have caused a change in the composition of the clay minerals, principally through the formation of magnesian chlorites. It is possible that this substitution of chlorite for illite has been accompanied by a certain release of boron. It effectively seems that, despite the large dispersion and imprecision of the measurements, the boron levels in the present  $C_1$  sandstones, when one relates them to the amount of clay present, may be significantly higher than the levels of boron in the argiles de piles. In the following material, we will vary  $x$  within the interval 30 to 70 as for the argillaceous material, but *it is necessary to remember that these values may be underestimated.*

The calculations show that it is impossible for criticality to be achieved without a significant ingress of water. Table 5 gives the values for  $k_\infty$  as a function of the ore grade,  $t$ , and the porosity,  $p$ , when  $Q = 8$  and  $x = 40$ .

**Table 5.**  $k_\infty$  as a function of porosity for the sandstones.

Porosity	$t = 35\%$	$t = 40\%$	$t = 45\%$	$t = 50\%$
2.5%	0.8109	0.8312	0.8350	0.8226
5.0	0.8696	0.9056	0.9255	0.9291
7.5	0.9006	0.9484	0.9808	0.9775
10.0	0.9146	0.9726	1.0149	1.0424

In the sandstones, the open porosity is usually of the order of 1 to 5%; one sees this is totally insufficient for criticality and one needs at least 10%. *It is necessary then to assume that the sandstones were fractured by tectonic actions, and that supplementary voids were created, either by extension or by dissolution of silica.* The table also shows that one cannot get by with too

low a grade of ore: for  $t = 35\%$ , the optimum for moderation has already practically been reached for porosity = 10%, and there is no chance of reaching criticality. As we already knew, the benefit of entry of water is much stronger in rich ore.

Table 6 allows us to get a more precise idea of the minimal values of the porosity. We examine the critical levels for  $p$  for different combinations of  $t$  and  $Q$  and using two values for  $x$ . We assumed in all the cases that the thickness of the sandstones is about 2 meters.

One notes that when  $x$  is fixed, the critical porosity remains about the same size. It passes through somewhat of a minimum as a function of  $t$ , and only increases moderately when  $Q$  is increased. We note that the porosity must be at least 8 to 10% for  $x = 30$ , and 10 to 13% for  $x = 50$ .

**Table 6.** Porosity for the different combinations of parameters:  $Q$ ,  $x$ , and  $t$ .

$Q$	$x$	$t = 35\%$	$t = 40\%$	$t = 45\%$	$t = 50\%$	$t = 55\%$
4	30	12.5%	9.5%	8.5%	8.7%	
	50		13.0	10.6	10.1	
6	30		10.6	9.2	8.8	9.2
	50			13.7	10.7	10.3
8	30			10.1	9.0	8.9
	50			16.4	11.9	10.3

Table 7 presents the results in another form. We have examined this time the critical value of  $x$  for different combinations of  $t$  and  $Q$  and two values of the porosity. For porosity, we chose 9.1 and 13% (being the ratio of empty to filled of 10 and 15% respectively). The thickness of the sandstone was kept the same as in Table 6.

In these results, we have not considered values of  $x$  below 30 because, taking into account the presence of rare earth elements and the remarks made above, these levels of poison (below 30) would not be realistic. We see, moreover, that when the porosity is less than 10%, only very low values of  $x$ , which are not very plausible, permit criticality to occur. In contrast, for a porosity of 13%, criticality is possible in a rather large domain. *It is, then, certainly the entry of water into the sandstone that is the essential element.*

The two preceding tables confirm that the ore grade must be very high and, of course, the minimum value for  $t$  is higher the larger the value of  $Q$ . Table 8 clarifies this issue of minimum ore grade: we have examined this time the critical value of  $t$  for different combinations of  $Q$  and  $x$ , putting it in each case in the region of optimal moderation (which corresponds to a porosity of 15 to 18%). For the sake of consistency, we have kept the same thickness of rich sandstones that was used in the preceding examples, but it would be necessary, also, to consider variations in that parameter. We give both the  $tg$  of the sandstone and the  $t$  value of the argillaceous ore.

**Table 7.** Critical level of poison for different combinations of parameters.

Porosity	Q	t = 35%	t = 40%	t = 45%	t = 50%	t = 55%
p = 9.1%	4		30.4	36.8	35.4	
	6			32.5	36.9	
	8				34.6	32.4
p = 13%	4	31.8	51.0	69.2	84.5	92.7
	6		39.1	62.4	84.2	100.9
	8			51.1	78.5	102.5

**Table 8.** Critical ore grade in the vicinity of optimal moderation.

Q	x = 30		x = 50		x = 70	
	tg %	t%	tg %	t%	tg%	t%
4	11.8	34.6	13.9	38.5	16.0	42.0
6	10.7	38.8	12.2	32.0	13.8	44.9
8	9.9	42.0	11.1	44.8	12.4	47.2

Consistent with a remark made above,  $tg$  [note: the Naudet text now uses  $t_g$ ] and  $t$  vary in opposite directions as a function of  $Q$ . We see that only sandstones with more than 10% for  $tg$  (which, one recalls is the average over a two meter thickness) can be the sight of nuclear reactions. We also note that the  $t$  values [present day ore grade] are rather high and although we encounter such values at Oklo, they are higher than those often seen in the reactor zones. *This means that the reactions could not start in all locations: only the richest portions had the possibility of becoming spontaneously*

*critical.* In particular, when  $Q = 8$ , the minimal  $t$  is about 45% (it could even be higher if there were more poisons); the margin is very narrow. On the other hand, when  $Q$  is smaller, that is when there has already been elimination of silica, the margin is larger; the reactions could have started under less restrictive conditions.

In conclusion, we summarize from this section the following points:

*From what we know about the preexisting sandstones, it is possible to justify initiation of the reactions, but only under certain circumstances.*

*-- One absolutely necessary condition is that the sandstones had been fractured in order to allow entry of sufficient water: the open porosity must have reached the range of 10 to 15%.*

*-- In addition, the reactions could only start in limited portions of the zones that we know, where the uranium ore grades are very elevated.*

*-- Although this assumption is not essential, the conditions are less restrictive if we assume that there was a preexisting elimination of some of the silica.*

### 3

## DETAILED EXAMINATION OF SITUATIONS ENCOUNTERED AT OKLO

We will now examine in a more detailed and more precise manner how criticality was achieved at Oklo. The parametric study allowed us to bring out the approximate sizes, notably for thicknesses necessary for rich zones of ore as a function of the uranium content. We noted that these characteristics correspond well to those that we find on average at Oklo, and equally, that the correlations suggested by the calculations are more or less found. But this remains a little vague; the variety of situations encountered demands that one look at things a little more closely.

It is for the final condition that we can do this with the least uncertainty. If we have listed a distribution of values for uranium and for isotopic depletion along a drillhole, we can treat this using the CRITO code. We need to choose a certain number of parameters for this, but we have seen that these are either relatively well known (for example, the amount of water through the determination of the spectral index) or that the results are relatively insensitive to them (for example the level of poisons because these are for the most part destroyed by the neutron flux). A more important lack of information is caused by the transposition of lengths. The calculation furnishes a  $k_{\text{eff}}$  and we must find in all cases this coefficient in the neighborhood of unity.

In reality,  $k_{\text{eff}}$  can only be equal to 1 if the neutron exchanges with the environment are equilibrated. The characteristics of the rich bands vary greatly in the interior of the deposit, and the distribution revealed by a borehole is never perfectly representative. If we have crossed a region locally poor in U, the critical condition may only be guaranteed by the transport of neutrons from the interior, and it is normal to find a  $k_{\text{eff}}$  smaller than unity. Conversely, a local environment richer in U must have a  $k_{\text{eff}} > 1$ , because there must be a reserve of neutrons to supply the lateral escape of neutrons.

It is thus necessary to take into account the environment to appreciate the results. But even if one knew the environment well, one could not draw precise information from the study of only a single distribution. On the other hand, if one uses a collection of such calculations, it is easy to produce statistically a representative mean value. This must then give us information about the validity of the assumptions that have been made. Among other things, we can evaluate whether conditions were similar in the different sectors or if there were

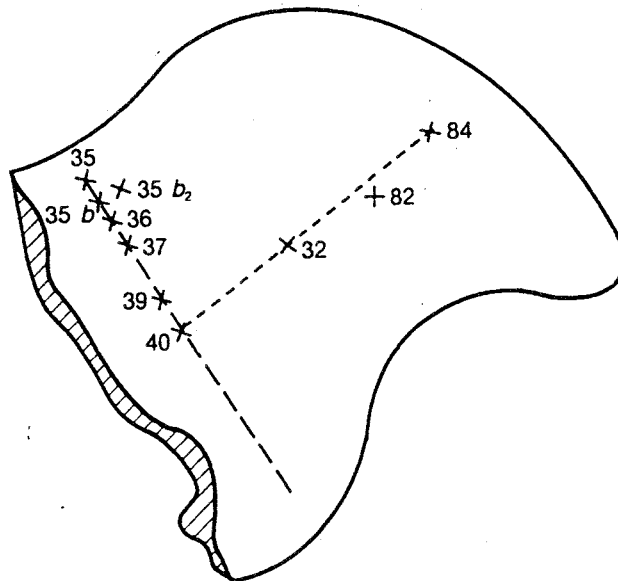
discrepancies. We have treated through calculation about 20 distributions. The results are presented, distinguishing between the different zones.

### 3.1 EXAMINATION OF ZONE 2

Zone 2, as we recall, is the most studied zone. It is massive and compact; it is the zone with the highest levels of isotopic depletion; and it is also the environment in which there has been the most complete conversion to clay. From the beginning of our studies, we have had the feeling that the conditions of irradiation in that environment must have been very homogeneous. The calculations confirm this.

The position of the boreholes considered here is shown in figure 8. Seven of them were in a line that crossed the reactor zone; the three last ones form another slice in the downhill direction. Figure 15 in chapter 7 [p. 358] shows the deformations and variations in thickness of the rich zone in the small portion which is between SC35 and SC37. Although zone 2 is the most uniform, there are important variations. Recall that the rich zone is split in two at the level of SC39 and SC40 and that it thins toward the downhill direction.

In order to treat the borehole data using calculations, we divide them into partial environments. Table 9 shows by way of example the division of SC36: each environment is defined by its thickness (measured along the borehole), its average uranium content, and its isotopic depletion. In addition, we place on either side a reflector consisting of a large thickness of low grade ore (1%).



*Figure 8. Location of the boreholes examined in zone 2.*

**Table 9.** Representation of SC 36

L, cm	t, %	U <sub>5</sub>	L, cm	t, %	U <sub>5</sub>	L, cm	t, %	U <sub>5</sub>
12.5	3	0.68	5	52	0.525	10	58	0.568
10	6	0.64	5	46	0.472	5	56	0.564
10	14	0.60	5	42	0.412	5	41	0.552
5	25	0.557	5	47	0.437	5	10	0.473
5	42	0.533	5	51	0.484	7.5	4	0.525
5	51	0.539	5	54	0.533	10	2	0.62

All the calculations have been done using the same conditions: "standard" composition of the gangue and heterogeneity, age = 1950 MA, temperature = 160°C, initial x = 50 (of which 35 ppm is actual boron and the rest is accumulated fission products that are calculated using the same "recipe"), m = 0.6 (a value that takes into account the results from measurements of the spectral index). In all cases we have used RV = 1.24 to transpose the length measurements (mean value consistent with m = 0.6). Finally, we have assumed that the angle  $\alpha$  of the dip is 45° at the level of the upper line and that it decreases to 35° as we go downhill.

**Table 10.** Critical condition calculated for ten boreholes in zone 2.

Borehole	H, cm	cos $\alpha$	H <sub>e</sub> , cm	t, %	e, %	k <sub>eff</sub>
SC 35	32	0.7	50	51	0.56	(0.958)
SC 35bis	79	0.7	88	52	0.49	1.020
SC 35 b2	76	0.7	85	46	0.47	0.997
SC 36	62	0.7	72	49	0.52	1.019
SC 37	55	0.7	69	44	0.54	1.022
SC 39	52	0.7	66	45	0.52	0.995
SC 40	(75)	0.7	(74)	(35)	0.55	1.005
SC 32	42	0.76	63	50	0.53	1.002
SC 82	32	0.80	51	48	0.62	0.998
SC 84	30	0.84	48	40	0.63	(0.953)

Table 10 gives the k<sub>eff</sub> calculated under these conditions by CRITO. For each borehole, we have noted for information the thickness H of the rich zone (in practice, the length of the portion for which k<sub>∞</sub> > 1), as well as its mean ore grade



$t$ , and its average isotopic depletion,  $e$ . We also give the  $H_e$  determined as indicated in § 1.1 (when the rich zone is well defined, the difference between  $H_e$  and  $1.24 \cos\alpha$  can be interpreted as the "gain of the reflector", but in SC 40 for example, where the rich zone is split in two, the total thickness has limited meaning and the average ore grade is not representative).

We note that, despite the diversity of combinations of characteristics ( $t$ ,  $H_e$ ,  $e$ ) for the rich zones, the values of  $k_{\text{eff}}$  (except for the first and the last) are closely grouped around unity. In addition, a careful examination of the boreholes shows, as we shall see, that the deviations can be suitably explained by effects of the environment.

S 35 is located at the edge of the zone. The rich zone is thinning and the ore was only able to reach a high level of irradiation thanks to its "powerful" neighbor, SC35*bis*, which donated neutrons to it; this explains the  $k_{\text{eff}}$  less than unity in that area. One can undoubtedly explain SC 84 located at the downhill edge of the zone in a similar fashion. On the other hand, SC35*bis* and SC36 are found in the place where the accumulation of uranium is the highest and they are surrounded by boreholes where the ore grade is much lower; this environment explains the  $k_{\text{eff}}$  a little higher than unity. SC35b2 is equilibrated; it pays back its great thickness by having a record isotopic depletion. It is interesting to compare this with SC37, somewhat lower in grade but for which the  $k_{\text{eff}}$  is still greater than unity. The latter sample had to donate neutrons to SC36*bis*, for which the thickness is insufficient for criticality. Note that the  $k_{\text{eff}}$  remains about the same value going from SC39 to SC82 and then to SC32, even though the thickness of the rich zone decreases considerably along this line. This decrease is compensated for, as it must be, by a lower level of isotopic depletion.

All of these remarks tend to reduce again the dispersion of the values found, and *one can then say not only that the results are very well clustered, and that the critical condition is justified with high precision, but that the results are remarkably homogeneous.*

### ***Discussion***

This conclusion deserves some comments.

First of all, what are the error limits in the calculations? One can detail them as follows. If one estimates that the age is known to  $\pm 30$  MA, this produces an uncertainty in  $k_{\text{eff}}$  of about  $\pm 500$  pcm [0.5%]. If we assume that  $m$  is in the interval 0.55 to 0.65, this translate into an uncertainty of  $\pm 800$  pcm [0.8%], to which we must add the effect on the neutron losses by the use of calculation of RV. A variation of 20 ppm in the initial level of boron 10 in the gangue has an effect on  $k_{\text{eff}}$  which is between 200 and 800 pcm [0.2 to 0.8%] depending on the isotopic depletion. The principal source of uncertainty comes from the geometry. An error on the order of 10% in the evaluation of "corrected" lengths translates

into a variation of 1300 pcm [1.3%] for a borehole such as SC36, but which increases when the rich zone is thinner. A difference of 20°C in the temperature represents 200 pcm [0.2%] if we limit consideration to "direct" effects. It is more difficult to evaluate the error that we introduce during assessment of the effects of heterogeneity of the environment. Even for very rich ores, it could be on the order of 500 to 1000 pcm [0.5 to 1.0%]. It remains for us to evaluate the uncertainties from the nuclear reaction constants themselves:  $\nu$  for uranium 235, adjustment of the effective resonance integrals for uranium 238, capture by fission products, etc.

As we can see, the list is rather long, but one notes also that each of the sources of error - at least if we have not been overly optimistic in evaluation of the sizes - has only limited consequences on the calculation of  $k_{\text{eff}}$ , in general less than 1%. Since all these sources of error are independent, their total effect, when combined quadratically, should not be greater than 2 to 2.5%. It is rather remarkable that the mean of  $(k_{\text{eff}} - 1)$  lies without doubt comfortably within these limits. One can say that *to within about 2% uncertainty, we have justified criticality to better than 1%*.

These results are certainly very satisfying. One could even observe - with a little malice - that the precision compares favorably with that for criticality experiments in the 1960s designed to test the form of the industrial nuclear reactors, which had a great advantage over Oklo, since these were in a well-defined multiplication environment. But this agreement, and this homogeneity are above all interesting because of their consequences on our study. In effect, *they tend to show that not only the choice of numerical values had the good fortune to be done correctly, but also that a certain number of implicit hypotheses are very likely justified.*

**Development of this idea**, underlining the most important points.

-- We know that there has been considerable discussion about the value of the age to use; it is thus that in 1975 the first calculation of criticality used 1780 MA. Such a value caused the  $k_{\text{eff}}$  to be lowered by about 3% and the deficit around the mean value to be visible. (At that time this was accommodated because the calculations were more simplified and in particular one had largely underestimated the capture of neutrons by fission products). Of course, there can be compensations here, and it is not a question of deducing a date from which to start the criticality calculations, but one can at least say that *these contribute to giving a credibility to the chosen value.*

-- Among the implicit hypotheses, there is one which is fundamental: this is that we assume that the uranium has remained in situ. It is clear that *if a significant fraction of the uranium has been leached away, it would not be possible to justify criticality* as we have done.

-- Another very important hypothesis is the one that deals with calculation of poisons. We have assumed that the isotopes eliminated by neutron capture have not been renewed by a mechanism of geochemical migration. It is thus that for the very irradiated ore zones we have only taken into account 4% of the initial boron 10. If one assumes, for example, that half the initial boron had been restored, that would entail a reduction of about 5% in  $k_{\text{eff}}$ ; in that case, criticality could no longer be justified. *The results tend, then to justify that such a renewal of boron has not taken place.* Moreover, if this had taken place, one would probably have seen much larger discrepancies between the boreholes; the same comment applies to dissolution of uranium.

-- We have also assumed implicitly that the environment was totally desilicified at the end of the reactions, and that in particular, the geometry was close to that we now see. If we take into account variable amounts of silica, the  $k_{\text{eff}}$  would have been smaller and the homogeneity would not have been maintained.

-- Furthermore, the calculations have been done using in all cases the same value for  $m$ , and we have chosen a high value taking into account the measurements of the spectral index. This is also a very sensitive parameter, since if, for example, we had taken  $m = 0.4$  rather than 0.6, the  $k_{\text{eff}}$  would have been lowered by about 5% (taking into account the indirect effect on RV). One can conclude from this two very important consequences.

First, the act of using for the calculations a value close to those deduced from the measurements of  $r$  implies that the porosity has not varied greatly during the reactions, since the measurements apply to the mean, even though the calculation is done for the final conditions. (In fact, we shall see in chapter 8 that the best value deduced from these measurements is 0.65; we have then chosen here a value slightly less than that). The fact that this magnitude is proven to be correct tends to show that the degree of compaction was little modified during the course of the reactions and that *the reactions were not prematurely stopped by an effect of collapse which reduced the amount of water available.* Everything appears, on the contrary, to indicate that they continued to their completion in the presence of a large amount of water (and this is moreover what justifies the very high levels of irradiation).

In addition, the excellent agreement among the results tends to show that *throughout the zone the porosities that must be considered are of the same order.*

This comment was already made in connection with the measurements of the spectral index (cf. chap. 5, § 4.2). The present calculations tend to confirm this proposal in a more tidy manner.

Given the very high values of the porosity, which can only be explained by calling upon imperfect compaction, one would have been able to understand that this continued during the reactions; the porosities ( $m$ ) would have been variable both in time and space. But we can understand also that once desilicification is

achieved, and the essential compression is realised (the reduction in volume has been, despite everything, considerable), one is lead to a situation of pseudo-equilibrium, evolving from that time only very slowly. The series of comments above argues for the second interpretation. In other words, the results suggest that *in zone 2, the reactions occurred in an environment already practically stabilized*. We will give other arguments in favor of this proposition in later parts of this study.

We shall see that everything that was just said about zone 2 does not apply everywhere.

### 3.2 EXAMINATION OF THE OTHER ZONES

Now that we have thoroughly examined the calculations for zone 2, it will be easier, proceeding by comparison, to appreciate the results from the other zones, even if the information for them is less numerous and less precise. We are led to distinguish three groups.

#### a) Zones 1 and 6

Zone 1 was explored at the same time as zone 2, and it was rapidly apparent that despite a certain number of differences, these two zones are essentially very similar. In particular, the correlations between isotopic depletion and the characteristics of the rich ore zones are to a first approximation comparable (in general the accumulation of uranium is less, or less concentrated, which translated into average isotopic depletions that are less pronounced).

The investigations in this zone were less systematic than in zone 2 and it happens that the rich zone is much more irregular. It is thus less easy to demonstrate homogeneity for the irradiation conditions. Nevertheless, as we shall see, the calculations confirm that the former conclusions can reasonably well be extended into this zone. Table 11 gives the results for four boreholes that have been treated by the CRITO code exactly under the same conditions as the preceding series [for zone 2].

**Table 11.** Critical condition calculated for four boreholes from zone 1.

Borehole	H, cm	$\cos\alpha$	$H_e$ , cm	t, %	e, %	$k_{eff}$
SC29	~40	0.84	53.4	38	0.62	1.024
SC61	~60	0.866	73.5	39	0.54	1.018
SC30	~64	0.866	72.8	40	0.52	1.001
Sc30bis	~30	0.866	55.4	39	0.59	0.981

We note that the  $k_{\text{eff}}$  values are still close to unity. It should be noted that we have, in general, selected for the detailed studies the richest boreholes in each section; it is then not abnormal to find for the two first cases a  $k_{\text{eff}}$  a little higher than 1. The SC29 sample is a borehole typical of zone 1, with a compact region of ore for which the grade is not particularly high; SC61 has, on the contrary, as for SC40 considered above, two rich zones separated by a rather marked furrow [zone with much lower uranium content]; SC30 and SC30bis are flanked on the roof by a secondary rich zone, from which the reflector gains are elevated, which makes them very sensitive to the choice for levels of poisons. Finally, despite this diversity of conditions, the balances are comparable and *we can consider that the results are completely compatible with those for zone 2*. In particular, it should be emphasized that criticality is justified for the same values of  $m$  and that observation must be compared with the fact that we have also noted a good agreement at the level of the spectral indices.

In zone 1 as in zone 2, all of bed  $C_1$  has been desilicified. This is an important peculiarity that distinguished these two zones from all the others. However, zone 6 represents an intermediate condition. It is only a link between zones 2 and 5, and its characteristics change with elevation; argillization becomes more and more important with the result that the top of zone 6 is very close to the base of zone 2. Even though we only have very limited data, it seems that this similarity extends equally well to criticality.

There is only one borehole in this sector, KN256; the core is rather well known over 30 cm, but there are two unfortunate gaps on each end. Radiographic measurements show that the total thickness of the rich zone is probably on the order of 35 to 40 cm. If we do the calculation using the average values of  $t = 45\%$ ,  $e = 0.64$ , from the known portion of the rich zone, and the same values for the other parameters as before, we find a critical  $H_c$  of 52 cm. For a  $\cos\alpha$  of 0.76, this  $H_c$  is compatible with an  $H$  in the neighborhood of 35 cm, which is acceptable. We can then say that this portion of zone 6 is effectively compatible with the preceding zones.

#### **b) Zones 3 to 5**

In contrast, the group 3-4 and the zone 5 (including the linkage through zone 6) appear noticeably different. Only a fraction of the bed  $C_1$  has been desilicified, and there are remnants of sandstones on both side of the clay zone, which is often rather thin. The total accumulation of uranium is clearly less than in zones 1 and 2, but it is also often more concentrated. On the other hand, the fact there are sandstones still present on the roof of the reactors has entailed, at the time of elimination of the silica, tectonic readjustments of a different nature from those one observes when the reactors are covered by pelites. These peculiarities pertain to both the mineralization and to the conditions of desilicification.

Since the beginning of the exploration of these zones, we noticed that, because of the smaller accumulations of uranium, criticality must have been much more marginal. We expected, even, to find frequent interruptions of the reactions and it was a surprise to find that the uranium throughout the zone was isotopically depleted. Certainly, we find large sectors where the depletion is very slight and we wonder if it is only contamination (it was not always possible to eliminate the uncertainty by an isotopic analysis). But even where there was incontestable evidence for reactions, the rich zones are very narrow. Before the calculations, it seemed difficult to justify criticality under conditions homogeneous with those of zones 1 and 2.

Data concerning the distributions of uranium are much less precise. There are few boreholes and, with the exception of a small group in a sector of zone 3, the drill cores are of bad quality. Often, one only has debris, which is inevitably not representative, is poorly located, and contains gaps. These zones are above all known by means of geologic samples (outcrops and mining fronts), but these don't lend themselves to quantitative balances. Most often one only has a series of samples for which the locations are not very well defined. In addition, the surfaces are usually not planar, particularly for the fronts, which further are generally not vertical, nor perpendicular to the level lines. It is thus difficult to reconstitute true thicknesses. Finally, we have said that the densities for ores in situ are not at all comparable to those from cores (the terrains having been disrupted through blasting).

In addition, the rich zone is very irregular. Its thickness varies enormously from point to point and it is most often chopped up by tectonic faults (cf. chap. 1, fig. 13). Another disconcerting remark is that it seems that there are not clear correlations here between the accumulations of uranium and the isotopic depletion. One can find very thin zones with significant depletion, or by contrast, wide zones with uranium that is practically normal in composition, without this being able to be easily explained by the effects of the environment.

Because of the lack of precise data and because of the irregularities it is not possible to do a quantitative study of criticality here comparable to that done for zone 2. It is necessary, rather, to consider as indicative "soundings" the results from the calculations that follow.

In a small number of cases, the characteristics are comparable to those discussed before. For example, in zone 3 the hole SC52 crossed a rich zone with modest grade but which was rather thin. The core is of good quality and one can analyze in detail the distribution; one can treat the data with the code CRITO using the same conditions as before. With  $H = 60$  cm,  $\cos \alpha = 0.64$  ( $\alpha \sim 50^\circ$ ),  $t = 37\%$ ,  $e = 0.64$ , one finds  $H_e = 69$  cm and  $k_{\text{eff}} = 1.043$ . There is, then, in this case a rather large excess of activity, which is explicable because the environment is much poorer in U and it is found that even that boreholes SC53 and SC 54, situated on

either side, have hardly been effected by the reactions. It would have been the lateral escape of neutrons that prevented the level of irradiation from becoming larger despite the favorable local conditions. In the same way, the mining face 166 in zone 4 has a rather regular rich zone. The section GL1853 to 1859 shows an average grade of 39% and an isotopic depletion of 0.62 over a thickness of about 33 cm. One finds, under the same conditions as before, a critical value of  $H_e$  of 56 cm, which is a priori compatible with the geometry.

On the outcrops, as we have said before, the thicknesses of the rich zones are usually extremely irregular. We studied a section that was particularly broad and rich on outcrop number 6 and the junction between zones 5 and 6 (GL1935 to GL1945). We found an average grade of 54% over nearly 40 cm (average  $e = 0.60$ ). But most often the conditions are much less favorable. For example, all along the length of outcrop # 5 in zone 3-4, the thicknesses are modest. We can consider the section from GL1630 to 1636 as representative, where we find an average grade of 45% over about 27 cm (average  $e = 0.66$ ). If one treats these two sections using the same conditions as before, we find for the first one  $k_{\text{eff}} = 1.021$  and for the second  $k_{\text{eff}} = 0.947$ . If we suppose that the excess in the first case is justified by surroundings that are rather poor, then in the second case the deficit is representative of the average over a large distance.

Likewise, the series of boreholes SC86 to SC93 only encountered very mediocre accumulations of uranium. The richest is SC89 in zone 5. The core is not in too bad condition and we have analyzed the distribution in detail. We have also treated the data using the CRITO code. With  $H \sim 35\text{cm}$  ( $\cos \alpha \sim 0.7$ ),  $t = 42\%$  and  $e = 0.63$ , one finds  $H_e \sim 49\text{cm}$  and  $k_{\text{eff}} = 0.972$ . The deficit, although modest, must be considered as very significant, because the core is surrounded by ore that is markedly poorer.

But what is even more astonishing is the existence of a series of boreholes, KN243, KN245, and KN267, also in zone 5, in which the thickness of the rich zone is only 20 to 25 cm. The average ore grades are on the other hand rather elevated (50 to 54%), the isotopic depletions going from 0.66 for KN243 to 0.62 for KN267. If we adopt as average characteristics (representing those from KN245)  $H = 24\text{ cm}$ ,  $\cos \alpha = 0.7$ ,  $t = 52\%$ ,  $e = 0.64$ , we find a  $k_{\text{eff}}$  in the vicinity of 0.915!

*Thus, even though one can find in zones 3 to 5 situations rather comparable to those in zones 1 and 2, there are also vast sections where one observes a systematic deficit of reactivity and where it does not seem possible to justify criticality under the same conditions as for zone 2. This means that certain choices of parameters or certain assumptions must be modified.*

In the first place, we noted in chapter 5 (§4.2) that the measurements of spectral index made in zones 3 to 5, although few in number, gave dispersed values, sometimes with results that could only be interpreted by appealing to very high

porosities. For example, we found for a sample from KN245 an index  $r$  of only 0.12, although the grade of the ore is 52%; this leads us to a very large  $m$ , on the order of 1.15. We also found values of  $m$  in the vicinity of 1 in SC55 and SC89 and the front 127. One has then the impression that conditions were not uniform and that there was sometimes important ingress of water.

On the other hand, we have seen in paragraph 2.1 that in the case where the rich zone is both very narrow and very high grade, we can find a situation more satisfactory for reaction when the desilicification is incomplete, so long as there is enough water. This remark applies particularly to the environment of KN245. The reactions probably began in this sector in a sandstone environment and we can even imagine that the desilicification and compaction were not accomplished at the end of the reaction. Table 12 gives values of  $k_{\text{eff}}$  for several combinations of  $Q$  and  $m$ , keeping the other parameters constant and varying  $RV$  and  $H_e$  in a consistent manner.

We note that we can effectively justify criticality in the final conditions if we assume that there was more water [than for zone 2] (here with  $m$  in the vicinity of 1). In addition,  $k_{\text{eff}}$  can become close to unity even with a non-zero value of  $Q$  (for example  $Q = 1$ ). We are led to conclude that in certain parts of zones 3 to 5, contrary to what we said for zone 2, *the reactions took place in large part if not entirely in an unstabilized environment*. Other arguments in favor of this conclusion will be presented during the course of this study.

**Table 12.** Examination of critical conditions in the case of KN245.

$Q$	$m$	$t, \%$	$e, \%$	$H_e, \text{ cm}$	$k_{\infty}$	$k_{\text{eff}}$
0	0.6	52	0.64	40	1.204	0.915
0	0.8	52	0.64	42.5	1.229	0.975
0	1.0	52	0.64	45.2	1.237	1.015
1	1.0	52	0.64	52.3	1.216	0.997
2	1.0	52	0.64	59.3	1.195	0.983
3	1.0	52	0.64	66.5	1.175	0.976

### c) Zones 7 to 9

We have already mentioned several times the peculiarities of this sector, notably the low level of uranium. These are usually on the order of  $2 \text{ kg/dm}^2$ , which is in general about 2 times less than in zones 3 to 5, and 3 to 4 times less than in the



richest region of zone 2. The thicknesses of the ore with elevated grade are nearly always reduced, sometimes even ridiculously, and above all, the ore grade is not high, rarely exceeding 30 to 35%. For example, a section of the front 305 has successively about 6 cm at 9% and 12 cm at 16%, 3 cm at 25%, 6 cm at 32% and 9 cm at 25% (giving about 1.8 kg/dm<sup>2</sup>).

Since the discovery of these zones, we have wondered how criticality could be explained, and detailed exploration has only reinforced the difficulties in explanation by confirming the insufficiency in the amount of uranium. It should be noted, that contrary to what we had hoped, even if the desilicification is not always complete, quartz is generally not present in significant quantities and without exception, the gangue is argillaceous, with capture being only a little smaller than that for zone 2 (notably a little less iron).

Because of these low grades, the approach that allowed us to justify criticality in zones 3 to 5, consisting of assuming an incomplete desilicification accompanied by a strong entry of water, can not be used here. Indeed, with an ore with too low a grade, the addition of silica always has unfavorable effects and overmoderation is very quickly attained. Table 13 shows as an example the results obtained for the section from front 305 of which we were just speaking.

Table 13: Examination of critical conditions for a section from front 305.

Q	m	x	e	k <sub>eff</sub>
1	0.8	50	0.72	0.833
1	1.1	50	0.72	0.834
0	0.8	50	0.72	0.852
0	0.8	50	0.64	0.883
0	0.8	30	0.64	0.899

For this distribution, the addition of silica is immediately unfavorable and overmoderation is reached as soon as  $m = 1$  (the gain is insignificant between 0.8 and 1.1). We see that under the best conditions, the deficit is enormous (on the order of 15%) in unreacted ore with  $x = 50$ . We can improve the initial neutron balance, of course, by lowering  $x$ , but the gain is illusory because  $x$  is strongly decreased by radiation. We have in that section  $e = 0.64$  and at that level the poisons are for the most part destroyed. We only gain 1800 pcm [1.8%] when, for example, we change  $x$  from 50 to 30. For the reactor in the final conditions, there is in any case *a considerable deficit and we cannot find a means of avoiding it*. Well, this section is not exceptional.

The situation is worse still when we envisage the initial starting of reactions because criticality is much more difficult to achieve when Q is high. We find nowhere in this sector ores rich enough over a sufficient thickness to justify starting reactions in sandstones. We are then led to think that *the reactors are without doubt no longer in the conditions under which they functioned.*

Because there is throughout an apparent deficit of uranium, with insufficient rich ore, could one think, for example, of a leaching with departure of uranium entrained in solutions of circulating water? It would be necessary then to explain why the ore did not behave the same way as in the northern sector. One could imagine, for example that the reactions had taken place for a long time in the sandstones, while in zones 1 to 6, argillization blocked the circulations very early. But we showed that in certain portions of zone 5, desilicification happened during the course of the reactions, and that did not lead to a massive departure of uranium, since there is still ore with more than 50% uranium and we could justify criticality there. There are, moreover, two reasons to think that uranium was rather less vulnerable in zone 7 to 9; these are that the reactions took place in the presence of active organic matter, and that the temperatures were lower. Naturally, once the reactions stopped, the two sectors experienced the same history (the only difference being that the zones 1 to 6 were slightly higher and thus more vulnerable in the very recent times.

In addition, if the uranium had been largely displaced during the course of the reactions, one would have found more abundant evidence for contamination of the environment, for which there is no evidence. In particular, uphill from zones 8 and 9, isotopic analyses were done in the ore partly desilicified by convection currents, with the aim of understanding the extent of contamination. In the front 299B, for example, even though the first samples were slightly depleted, one found within two meters of the reactors (GL2801) uranium with natural composition. Transport of depleted uranium seems rather to be less than uphill from zone 3 (which is not abnormal, considering the lower temperature). Finally, the hypothesis of entrainment of uranium in water in a manner greater than for the other zones has no support.

Because the presence of abundant organic matter is the most remarkable characteristic of the sector, one then wonders if there is a relationship between this fact and the problems with criticality. In the first case, the organic matter, because it was there when the reactions occurred, perhaps in the form of hydrocarbons that were not yet degraded, was of a type to improve the neutron balance. However, it participates, as for water, essentially through its hydrogen; well, we have seen that even for optimum moderation, we were still far from criticality. It could also have played a role as padding by opposing too rapid a compaction during desilicification (cf. chap. 5, §2.4), but for this effect to have been important, we must assume that the organic matter at the time of reaction was much more abundant than it is now, which is hardly compatible the hypothesis suggested in chapter 4 of a not too severe thermal history. Moreover, the organic

matter that was destroyed in situ left its uranium in the reactor zone. In sum, the calculations lead to a deficit of uranium no matter what hypothesis is used concerning the amounts of water or organic matter present during the phenomenon.

Another possibility is that the organic matter, which apparently was displaced after the start of the reactions, migrated outside the reactor zones taking along its uranium; this could have emptied the reactors of part of their contents. But, this time as well, one must find depleted uranium outside the reactors. Certainly, in the mining face of 1/4, for example, we have found a small veneer of rich ore with depleted uranium in a corner tectonically isolated (GL2491, 0.633%). One could think that it was brought there by migration of organic matter. One also finds pockets of organic matter containing uranium in fractures in the heart of poor sandstones. They could reasonably have migrated from the reactors during the time of reactions; however, the uranium is normal except when one is a short distance from the reactors (F299C, GL2832, F299D, #2948). The evidence is, thus, a little thin. Even if this phenomenon could have played a significant role, it does not seem that it could have been enough to explain a deficit of the size we are facing.

We cannot see any other possibility for explanation except to call on deformation of the terrain: *the reactors are no longer in their original geometric configuration*. We are further lead to this conclusion since from the beginning of investigations we were struck by the amplitude of the sliding and the collapsing of strata which had notably caused the famous "troughs" (chap.1). The rich bed could have been sometimes eroded and other times wiped out or thinned by deformation. This is the only possibility to think of in the case of "reactors" of a few centimeters thickness that one finds notably on the outcrop GL13. We know on the other hand that in several cases we find the reactor mounted on top of clays and sandstones, and we can wonder if sliding has not sometimes dissociated the argillaceous parts of the reactor from their sandstone portion.

We shall return to this problem in chapter 13. We shall see that we can only explain the deformation of the terrain if we suppose that desilicified portions were not compacted gradually by the pressure of the overburden, but on the contrary, vast spaces remained open until this produced a collapse of the total section. This implies that the pressure of the adjacent terrain was not too high and this would be an indication that the reactors functioned near the surface.

These observations prove in any case that the different zones did not behave in identical ways. Finally, we acknowledge that the difficulties encountered in explaining criticality for the other zones, and especially for zones 7 to 9, only makes more astounding the coherence of the data obtained for zone 2.

## APPENDIX

### THE IMPORTANCE OF AGE IN THE PHENOMENON OF "NATURAL REACTORS"

We have wondered about the frequency with which phenomena of the Oklo type might have been produced in very old uranium deposits. In this matter, the age of the deposit is an essential parameter. Table 14 recalls the evolution in time of the content of the fissile isotope of uranium as we go back in time.

**Table 14.** Concentration of uranium 235 as a function of its age.

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800 MA - 1.38%	1600 MA - 2.61%	2400 MA - 4.9%
1000 MA - 1.62%	1800 MA - 3.06%	2600 MA - 5.72%
1200 MA - 1.90%	2000 MA - 3.58%	2800 MA - 6.66%
1400 MA - 2.23%	2200 MA - 4.19%	3000 MA - 7.76%

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It is clear that the more we go back in time, the easier it is to achieve critical geometry and comparable compositions. But another aspect of the problem must be considered.

We know of no uranium ore body older than 2800 MA. Some people think, furthermore, that uranium was not extracted from the mantle in significant amounts until the era of the large granitic intrusions, which were emplaced principally from that time (ref. 189). On the other hand, the deposits with ages older than 2000 MA are found to be, for the most part, of the same type (quartz pebble conglomerates) with generally rather low ore grades. In these deposits, it seems that the uranium is detrital and that concentration occurred gravimetrically. This could only have happened in the near-total absence of oxygen in the atmosphere.

Most authors think that the level of oxygen in the atmosphere, which was until then rich in CO<sub>2</sub>, only began to grow significantly from an era between 2200 and 2000 MA (ref. 188). It is from that date that the geochemistry of iron was completely changed. Although the question is controversial, it seems that until then uranium could only be transported as valence 4, and that the only

mechanisms of concentration were crystal fractionation and gravity. Neither of these two mechanisms could lead to very high concentrations and this is what explains that we only know for that era uranium ore deposits with very low grades. (These are generally less than 1 part per thousand, and the uranium is usually only exploited as a by-product, as is the case in the Witwatersrand in South Africa).

Conditions seem to have become radically different after a time on the order of 2000 MA. Development of photosynthesis led to the dissociation of carbon dioxide into oxygen and organic carbon. From that time, surface waters were oxidizing and could contain uranium in the hexavalent form (while considerable amounts of iron precipitated in the ferric state). At the same time, the much more abundant organic matter made reducing fronts which constituted geochemical traps. It is the generalization of these redox processes that allows the formation of vein and sedimentary ore deposits, and this is when the often-described cycle of changes of these deposits by successive mobilizations began. In other words, it was the beginning of a new stage in the development of life that had made possible the accumulations of uranium that were much greater than those that formed up until then.

It seems that the uranium deposits of the Francevillian are among the oldest of this new generation. The problem is then posed differently depending on whether one considers uranium deposits more recent or older than those of Oklo. We made some calculations to investigate these two aspects of the question.

### **Uranium deposits younger than Oklo.**

In the initial condition, such as we described in paragraph 1.1, criticality imposed a relationship between the four parameters  $t$ ,  $H_e$ ,  $x$ , and  $m$ . If one lowers the age of the deposits, this relationship is modified and it is necessary, for the same values of the three other parameters, to have a higher ore grade, or a greater thickness of the rich zone, or a gangue that captures fewer neutrons (in particular one that contains fewer poisons), or to have more water (for the cases where optimum moderation had not yet been reached). That is, we must combine conditions that become more and more exceptional. How far can this go? Does it remain plausible?

Figure 9 shows the degradation when the time scale decreases. We chose to represent the critical  $x$  as a function of age for different combinations of the other three parameters. The first curve ( $t = 40\%$ ,  $H_e = 60$  cm,  $m = 0.5$ ) passes through a point near our reference case; the critical  $x$  value decreases to about 25 ppm when the age decreases by 200 million years, and it goes to zero at an age of about 1550 MA. The next curve ( $t = 45\%$ ,  $H_e = 70$  cm) is still representative of Oklo conditions, but is already exceptional; the critical  $x$  stays above zero a somewhat longer time. We consider then conditions even more favorable, taking the grade to 50 then 55 and 60 %, the equivalent thickness to 80, then 100 and

120, and finally  $m$  to 0.8 and 1.2. Each time the critical  $x$  is larger for a given age, and we can then in exchange accommodate ages that are younger. Nevertheless, we note that it becomes more and more difficult to lower the age limit. For the most favorable conditions that were considered,  $x$  goes to zero at 850 MA; one could go a little further, but not much, by choosing combinations that were still more favorable.

From curiosity, we studied what was the absolute age limit for a natural reactor. To do this, we considered an environment formed uniquely from uranium dioxide and water, with no gangue and no poison. We tried to place ourselves at the optimum for moderation, and assumed dimensions large enough so that neutron escapes were negligible. We note that the optimum  $Y$  value decreases with age; it is on the order of 0.13 in the vicinity of the point being examined. For  $H_e = 200$ , our code gives us a critical age of 425 MA and we can decrease that to

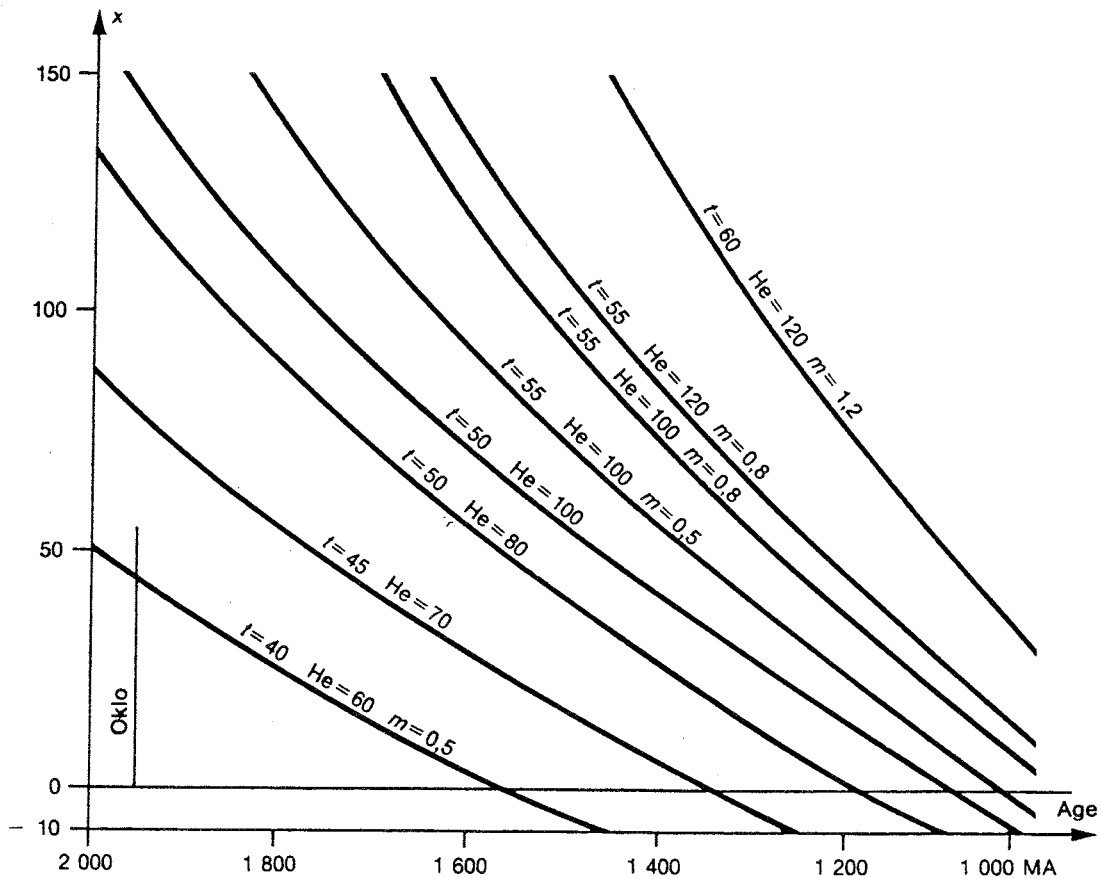


Figure 9: Critical  $x$  as a function of age for different combinations of  $t$ ,  $H_e$ , and  $m$ .

395 MA for an infinite medium (the isotopic composition of uranium in this case is 0.99% <sup>235</sup>U). One could undoubtedly gain a little more by calling upon heterogeneity, by separating the water and the uranium more. It is unnecessary to say that this case is purely theoretical and that it can clearly not be realized in nature, but it is interesting to note that even under these extreme conditions, we are still far from the present day.

On the contrary, the curves from figure 9 suggest that there are plausible combinations of parameters that permit going down to ages of the order of 1000 and perhaps 900 or even 800 MA. It is still advisable to examine this plausibility a little more closely.

First of all, how far can we lower  $x$ ? Note that the rare earth elements (despite their name) and boron are very common elements and that it would hardly be reasonable to imagine ore deposits that did not contain them. But besides that, we have seen that these poisons - or more generally the reserve reactivity they represent - are necessary to permit the natural reactor to function, because it is necessary to compensate for the burning out of uranium. If one does not include at least a little reserve in the balance, even if criticality were attained it could not be maintained and we would not reach a significant level of irradiation.

Setting aside that comment, we can imagine a capture capacity weaker than at Oklo. We have transferred to  $x$  that possible difference. We saw that the capture in the clays is equivalent to 35 ppm of boron 10, and that quartz captures 4 times less for an equal weight. In a sandstone for which the  $Q$  is high, even if we keep the same composition for the clay fraction (including the minor elements and the poisons), we can, with that definition obtain an  $x$  value near zero, or even slightly negative. It must be noted, however, that we know of no ore body anywhere that has such a gangue composition with, at the same time, a level of uranium of 50 to 60%. At Oklo, this level of ore grade was only obtained as a consequence of the last stage of concentration consisting of eliminating silica, which at the same time concentrated all the elements associated with the clay minerals. It seems that each time we find an extremely rich ore, a process of this type was involved, for example by means of circulating hydrothermal fluids.

One can say more generally that when an element becomes concentrated by some process, there are often others that behave the same way. It is for this reason that we often find uranium accompanied by a whole series of elements of which some strongly capture neutrons. In particular, vanadium is frequently associated with uranium; there are important quantities of vanadium in the nearby, and probably contemporary, deposit at Mounana. It is a stroke of luck for Oklo - still unexplained - that the levels of vanadium are so low. We can also note that the level of manganese is also very modest, although there are considerable amounts of it in the neighboring levels of the Francevillian. In the uranium deposit at Cluff in Canada, for example, which locally has very rich ores, there is a paragenesis that is very rich in neutron-capturing elements, in particular tourmaline with a high

concentration of boron. We must admit that at Oklo, apart from iron, there are few "neutron-loving" elements, and finally, not only would it not be realistic to consider values of  $x$  that are too low, but we have to consider that keeping a value of the same order as at Oklo already places us in favorable conditions.

Having said that, Figure 9 shows that this requirement is compatible with ages as low as 1000 MA, for the combinations of  $t$ ,  $H_e$ , and  $m$  that are only a little higher than at Oklo. Other comments, however, should be made. One will note, first, that the values of  $m$  considered imply the existence of ore in which water occupies up to half of the volume, and this can only happen in exceptional cases. At Oklo this was accomplished through a desilicification that was rapid enough so that compaction could not occur immediately. One wonders if this might not be the only process that would allow water to enter an ore body that rich.

But this leads us to another comment. It is legitimate to imagine that what happened at Oklo could have happened other times, but we must then transpose all the conditions; one of these is starting the reactions in sandstones that were not already desilicified. We have seen that this is not so easy. The conditions are rather restrictive and in any case one can justify criticality in these sandstones despite the penalty of the extra silica only by having a large amount of water come in and a reduction in the escape of neutrons through the means of the clay environment. But if we suppose that this latter condition is itself close to optimal for moderation, and that its thickness is such that the escape of neutrons is very small, there is not much more to be gained, and it is not even certain that in this case we can justify criticality in the sandstones. In other words, *"it is not reasonable to gamble on all the parameters at the same time"*.

We shall note again that a combination of parameters such as  $H_e = 120$ ,  $t = 60$  represents an enormous accumulation of uranium, more than twice as much as one finds occasionally at the best at Oklo. We consider always a "pancake" reactor with neutron escapes in a single direction. It is necessary for the uranium accumulation to be maintained over several meters in the other two dimensions. Also, if we consider a largely spherical pocket, the diameter of the sphere must be twice  $H_e$  and there must be at least 2 meters of 60% ore in all directions. This is not unthinkable, but the least one can say is that it is not a condition that one frequently encounters in uranium ore deposits.

These comments show that when we descend down the time scale, even if we remain in domains with characteristics that are not completely unreasonable, the probability of finding them together becomes smaller and smaller, and in the limit, we can doubt the plausibility of such a combination of circumstances. At the same time, this examination gives us a better appreciation of criticality at Oklo. This deposit has undoubtedly benefited from a number of favorable conditions, but which do not appear to be very exceptional, and criticality can be justified with a certain "elasticity". On the contrary, conditions become more and more stretched if one moves to younger ages.



In conclusion, we can say that nothing prohibits natural reactors from having been able to form up to a date on the order of one billion years ago. If in the vicinity of 2000 MA age, however, we can imagine the phenomenon was relatively frequent, then for dates much closer to present day, they could only have been produced under exceptional conditions and, therefore, with a very low probability. It is, without doubt, not very realistic to think of ages less than 1500 MA.

We know a certain number of ore deposits, notably in Canada and in northern Australia, which also contain very large accumulations of uranium, and with ages comparable to Oklo. This was, at least, what one believed a few years ago, placing them in the range of 1600 to 1800 MA. Subsequently, while the age of Oklo increased to about 2 billion years, it became apparent that the Canadian and Australian deposits were rather between 1100 and 1600 MA. Under these conditions, it is less astonishing that the first investigations were revealed to be disappointing. It seems that the Gabon deposits are the oldest that one knows among those that consist of very high concentrations of uranium.

### Deposits of uranium older than those at Oklo

If we go to ages older than 2000 MA, we find conditions that were more favorable to starting nuclear reactions because of the composition of the uranium, but we come up against another handicap, which is the establishment of sufficient concentrations of uranium.

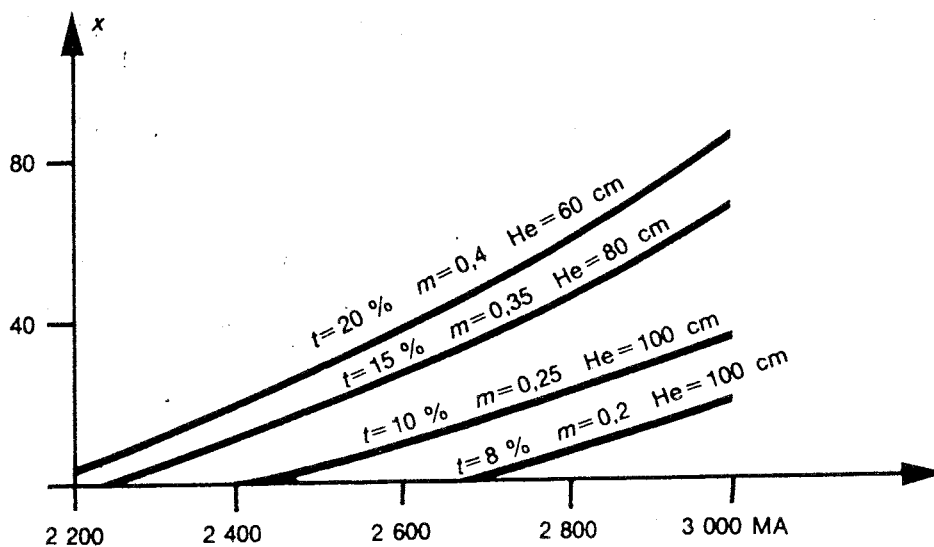


Figure 10. Critical value of  $x$  as a function of age for different combinations of  $t$  and  $He$  ( $m$  near the optimal value).

Figure 10 gives the critical values of  $x$  for different combinations of  $t$  and  $H_e$  and for sandstones with ages from 2200 to 3000 MA. We are systematically located close to optimal moderation, which only requires rather modest values of  $m$ , taking into account the low grade of the ore. We see that in that range of ages, it is possible to bring the value of  $t$  to about 20%, even for rather low thicknesses with  $H_e = 60$  cm. We could even descend to values of 10 to 15% on the condition that the thicknesses are chosen to be very large and the  $x$  values are smaller. But it seems that even at 3000 MA it is not realistic to consider an ore grade of less than 10%.

The preceding calculations relate to an ore of the Oklo type, as do the examples in figure 9. If we acknowledge that conditions were different at that time, it is perhaps more prudent to consider directly the sandstone ore deposits as they could have formed in the earlier time. Table 15 gives some results. To produce the largest reduction in ore grade, we have considered sandstones for which the  $Q$  value is elevated (that is, in which the fraction of clay in the gangue is small). One is located there, also, in the vicinity of optimal moderation and we suppose very large thicknesses. We calculated critical  $t_g$  values for several combinations of parameters.

**Table 15.** Critical ore grade in % for sandstones as a function of their age.

$Q$	$x$	$H_e$	2000	2200	2400	2600	2800
8	50	250	11.35	8.60	6.42	4.97	3.94
8	30	250	9.60	7.24	5.52	4.32	3.42
10	30	250	9.27	6.89	5.20	4.04	3.18
10	30	380	8.50	6.25	4.80	3.78	2.99

We see that one can effectively strongly lower the minimum ore grades. At 2800 MA, criticality can be realized in sandstones for which the grade is only 3 to 4%. This may seem to be very low, but it is not certain even so that concentration of this order could have been reached over thicknesses of several meters using only processes such as gravimetric accumulation, rather than calling significantly upon mechanisms with oxidation and reduction. Geologists think that these [oxidation-reduction] processes were very rare until the growth of oxygen in the atmosphere.

*Finally, it would seem that the phenomenon of "natural reactor", being stuck with these two requirements of high concentrations of uranium and a suitable isotopic composition, could only have taken place in a rather narrow "slot" in the history of the earth.*

## CHAPTER 13

### SOME PROBLEMS WITH THE OVERALL FUNCTIONING OF THE REACTOR ZONES

Throughout this study we have analyzed in sequence different aspects of the functioning of the reactors: how criticality was achieved; what were the control mechanisms; how the reactions, once started, reached neighboring areas; what role was played by the removal of heat by the expedient of thermal coupling; how did desilicification take place? Now we need to put together all these partial conclusions to give a more global picture. One important result, which has not yet been examined in detail, must be added to the collection of results. That is, the behavior of the terrain during the tectonic readjustments that followed desilicification. We have seen that there are a variety of situations, and the neutronic study has confirmed that conditions of functioning were not the same everywhere. We shall show that the manner in which compaction of the desilicified sandstones was achieved played a determining role in this respect.

On the other hand, we have inquired, particularly in chapter 4, about the circumstances in which the nuclear phenomenon fit itself into the geologic context and, more particularly, about the chronology of the events that led to the start of the reactions, and we have even wondered if there were not several distinct phases. We would like to proceed a little further in understanding the phenomenon as a whole.

It is very evident, even if the harvest of information was rich, that we could not reconstitute everything in detail and that many questions remain without answers. We will focus the discussion on these problems and try at least to set out what is well established, what is conjectural, and what is not well understood.

Among these problems, we will examine first those concerning the functioning of zone 2, the zone that furnished so many interesting pieces of information, and which allowed us to push very far our physical analysis. The problem in this case comes because we must explain how the apparent geometric and geochemical stability, which made possible the exceptional coherence, is compatible with the disturbances that caused the desilicification. We shall see that it is not that easy. We must also understand the reasons why the other zones had a different behavior. We will return to the problem of the initiation of the reactions and try to find out in each case the most probable points of initiation for the reactions and the conditions under which they were propagated.

The most troubling problem and the most difficult remains, without doubt, that of the peculiarities of zones 7 to 9. We will examine again the reasons why they must be considered separately: the idea that they functioned later and in a different context cannot, in our opinion, be dismissed. We must, however, place the nuclear events into a satisfactory chronology to fit the regional geologic history.

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# 1 FUNCTIONING OF ZONE 2

## 1.1 AN EMBARRASSING PROBLEM

When the evidence emerged that the reactor zones came from desilicification of rich sandstones and that this had disturbed their environment, one had trouble accepting this because we had gathered at that time, at least for zones 1 and 2, many reasons to think that the reactions developed in a medium "geometrically stabilized and geochemically preserved". Continued studies only reinforced that conclusion. It seemed that the only way to reconcile the two ideas was to call on a functioning by instalments, that is to suppose that the desilicification occurred progressively and that the reactions followed with a time interval sufficient each time for the terrain to have been completely desilicified and compacted at the time when they started. As we shall see, the justification for this time interval is not evident and the first, tentative explanation has been shown to be incorrect. We will discuss this problem by considering, more particularly, zone 2.

### *The reactions took place in a stable environment*

The idea that in the zone where the reactions occurred the environment was stable is a fundamental conclusion of the study of the physics. It seemed useful, before going further, to bring together and review the arguments that were presented in the course of this study, if only to correctly appreciate their limitations.

First, we saw in chapter 6 (§3.1) that in this zone criticality at the end of life could be justified in a very homogeneous manner in the current geometry, calculating the amount of water as if the porosity had been the same throughout. Certainly, this only applies to the situation at the conclusion of the reactions, but we note that the order of magnitude of the porosity that is suitable is the same as that given by the spectral indices, which characterize the entire period of functioning. This suggests that one arrived rather early in the course of the reactions to a state of pseudo-equilibrium in the compaction of the desilicified terrain that, even though it hadn't finished, only evolved very slowly. The observations, in any case, contrast radically with those of zones 3 to 5.

That idea was confirmed in chapter 7, since we could justify in a very systematic manner the form of the curves for level of reaction by making the same assumption. This time, it involved detailed distributions along the length of boreholes and the average amounts of water during the life of the reactors. Even in extreme cases (§2.2, chapter 7) we showed that there were no favored circulations of water in

possible fractures. The distribution of water was established in a very precise manner for borehole SC36 (chapter 8, §2.1), where we found that the average porosity, although extremely high, was nearly the same everywhere. These results could not be explained if the compaction had not arrived very early at an equilibrium.

Chapter 8 brought us even more decisive reasons for that stabilization, because one could interpret with exceptional coherence a large number of measurements by reasoning based on the geometry and composition of the argillaceous environment that we find today (only with a porosity uniformly larger). In particular, calculation of factors of conversion (and then of restitution) is very sensitive to the presence of quartz, which increases the resonance capture. The comparison of the isotopic depletion of uranium with the analyses of neodymium and thorium lead to values of the factors of restitution that agreed with the total absence of quartz.

This result is all the more significant since when one reaches the border regions, one finds values of the factors of restitution and curves of isotopic depletion of uranium that can only be interpreted by assuming the presence of a fraction of residual silica during functioning of the reactors. As the study of convection showed that desilicification proceeded more quickly in the core, and that it only reached the periphery after the "stopper" of clay had formed, these results give a coherent picture. They prove that the desilicification was effectively achieved in the core of the reactor at the start of the reactions, while it was only partially achieved in the border regions. This sequence of events, which lets us also explain that there were isotopic perturbations of uranium in the border when there were not in the core, has been confirmed in all the boreholes of zones 1 and 2 (chapter 7, §3.2).

We saw, on the other hand, in chapter 12 (§4.1) that one could only explain the near total absence of solution of uranium in the core (demonstrated in core SC36 through the balance of thorium and neodymium) through the means of assuming an extremely slow speed of circulation of fluids, implying a permeability on the order of that for the clays. In a general manner, this early argillization explains the geochemical stability of a certain number of elements (from which comes the notion of a "preserved" or "protected" environment). We also saw that the smallness of the circulation velocities is also capable of explaining the symmetry of the remobilization of the rare earth elements.

*There are, then, a number of impressive arguments, such as neutronics and geochemistry, that lead us to think that in zone 2 the reactions were each time developed in terrains that were already nearly totally desilicified and compacted.*

*It is not easy to explain this.*

This idea runs into two kinds of problems. In the first instance, the results that have just been recalled could suggest that from the first start the reactions proceeded very slowly, allowing a sufficient delay so that the vicinity of the reactor could transform and stabilize before the reactions arrived. This is why at the colloquium in Paris we said that the total duration of functioning of zone 2 was probably much more than

that of the functioning of each reactor individually [172]. But the thermal study presented in Chapter 11 (§2.2) showed that is not correct. The fact that a modest neutron flux was sufficient to maintain the temperatures necessary for control implies that the heat losses had been reduced and this can be justified only if we assume that nearly all the zone functioned at the same time. We must suppose, then, that if there was propagation, it was very rapid, leaving little time for the terrain to desilicify and stabilize.

In addition, the very idea of a time difference between desilicification and the reactions is a problem. While desilicification is only effective near the heat source, the neutron flux spreads out from the core all the more effectively when the  $k_{\infty}$  is close to unity. We saw in chapter 10 that the zone of overflowing of neutrons can extend itself rapidly with the destruction of the poisons and the removal of a fraction of the silica, thus extending the region where criticality is achieved. In zone 2, the ore is extremely rich. If a little water entered or if a fraction of the silica was removed, that was sufficient to raise the  $k_{\infty}$  to near unity (cf. fig. 6 in chapter 6). Once the reactions were started, they could then take over the whole zone before the desilicification and consequently the stabilization of the terrain had come to completion.

In reality, the mechanisms of propagation were complex, in particular because of the role played by temperature. In an attempt to respond to this objection, we have shown that it is possible to define conditions of propagation that allow explanation of the start. But, as we shall see, the proposed solution turns out not to be satisfactory.

#### *A false solution*

When a reactor started, it possessed a reserve of reactivity that allowed it to function until a certain level of irradiation. That reserve was normally held in the form of poison, but it could also be in the form of residual silica or temperature. For example, in the case of SC36, we saw that to compensate for the irradiation in the argillaceous ore, it was necessary to have poisons equivalent to 115 ppm of boron if one kept the porosity, and more if one increased  $m$ . This is certainly more than the real level of poisons. The difference could be silica, but not necessarily. Even if entirely desilicified, the reactor would not initial be critical if it were very hot. One then thought that it would be necessary to adjust this parameter. What might have impeded the reactions from expanding rapidly was perhaps *the harmful effect of temperature* lowering the reactivity in the vicinity of the core.

This effect could be reinforced when the convection currents carried a significant fraction of the heat. It tends to transfer towards the upper end the maximum temperature. The propagation of the reactions is then facilitated towards the bottom, which received the cold currents, but it is retarded towards the top where the temperatures remained very high over a long distance. If we then suppose that the reactions started at the base of the zone and progressed upward, the progression would have been strongly retarded, leaving each time enough time for desilicification to be achieved.



We studied this mechanism through calculations simulating the reactor, which were presented in chapter 10 (§3.3). In the example studied, the environment situated above had, in the beginning, a coefficient of multiplication less than unity. This coefficient was then increased to simulate desilicification, but the environment did not become critical because of the temperature. This actually increases rapidly during reactor operation, which sends upward very hot water, and the propagation of reactions during that phase is insignificant. It is only when the control mechanisms lower the temperature of the core enough to compensate for the irradiation that the reactions can extend upwards. We observe effectively *a large spreading out*, with the reactor zone above not really starting until the end of the time that represents a large part of the operating time.

We know now that *this solution does not work*. It is not possible to call on a spreading out in time that is so great, because we have to assume that the reactions extended rapidly into the whole zone. Also, the hypothesis of intense convection currents that carried away the major portion of the heat can no longer be retained. The role of convection in the removal of heat seems to have been modest. Convection only moderately deformed the temperature curve in the upward direction; the direction of the currents could not play such a strong role as in that model.

Finally, the third objection. We saw that convection favors the progression of desilicification laterally and towards the base, but not upwards, where the temperatures decreased in the direction of the currents, and where, consequently, the water, if it was already saturated, has rather a tendency to precipitate silica. Certainly, we showed that a progression towards the upward directions is ponderable, but only on the condition that phenomena of undersaturation associated with by-passes can be called upon, which is more plausible in the context of an encasing sandstone. In addition, which we will now examine, there are other reasons to cast doubt on the idea that desilicification proceeded upward.

## 1.2 PROPAGATION UPWARD OR DOWNWARD?

The hypothesis that propagation was from the base of the zone, envisaged in that solution, answered another preoccupation. We showed in chapter 6 (§2.2) that even in very rich sandstones, initial criticality could only be achieved with a degree of fracturing bringing the open porosity to exceptionally high levels, close to 15%. We then rightly thought that the points of initiation of the reactions had been few, limited to the small, strongly tectonized zones. In particular, it is easier to conceive of the creation and maintenance of a large porosity in subvertical regions, strongly fractured, where the pressure of the adjacent terrains is less directly exerted. Zone 2 has a shallow dip and is tectonically relatively calm. We wondered then if this zone, despite its rich ore, had been able to start spontaneously, or if it hadn't, rather, been *lit* from the outside.

The observations in zone 6 seem to indicate a progression toward the top (cf. §2.4). At the base, where this zone is detached from zone 5, there is precisely one portion strongly straightened out with elevated levels of reaction possibly appearing as a plausible starting point. In the upper part, it joins more or less zone 2. One imagined then that desilicification and the reactions in their progression from zone 5 had spanned over this joining area and continued up to zone 2.

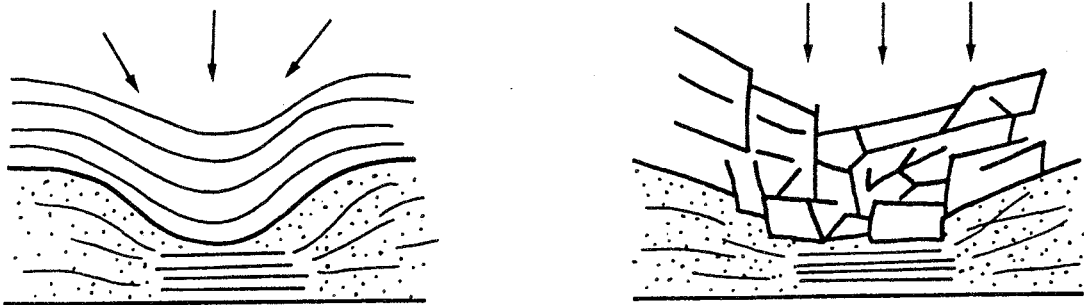
This idea had another advantage. We could think that during the desilicification phase, at the time when the permeability was very high, these currents had displaced little by little a certain number of the elements in the sandstones above. This could have been the case for uranium (from which an explanation of the fact that the top of the zone is much richer), but also for boron. We know that this poses a difficult problem (chapter 12, §4.3). The situation is more satisfactory if we suppose that the progression was toward the top, because the reactors cannot receive during operation unirradiated boron, as would risk being the case if the reactions extended towards the bottom.

In fact, we have taken into account subsequently that the *hypothesis of lighting of zone 6 runs into serious objections*. The study of convection showed that the intensity of the effects decreased very rapidly as we got further from the source of the heat. In the light of these results, the interval between the sufficiently rich portions of the two zones (even if it is not well known) seems too large for there to have been an efficient interaction, at least in this sense. In addition, if one could explain in this fashion the manner of functioning of zone 2, one could not do the same for zone 1, which posed comparable problems in the matters concerning the chronology of desilicification and reactions. The lowest part of that zone is subhorizontal, poor and little affected tectonically. It certainly does not seem to be a good candidate for the initiation site of the reactions. In this case, there is no auxiliary reactor nearby to facilitate the task.

But above all, in the matter of direction of propagation, there is a more important argument that seems to us to be decisive. It comes from the *observations of the terrain*. We know that between the sandstones of C1 and those of FB there is a bed of pelites. In the vicinity of zone 2, the thickness of the pelites is of the order of a meter, but at the emplacement of the reactors we observe a strong gliding towards the downward direction. The pelites are laminated and are thinned above the zone, while their thickness is on the contrary several meters below. To be able to explain that, we must analyze the behavior of the terrain at the time of desilicification. We shall see that this analysis will give us some insight.

When silica is eliminated, the reactor sandstones lose about 4/5 of their volume and the terrain above them must drop to fill up the "voids": that is, to effect the compaction of the remaining material. The rocks in the immediate vicinity, which support the weight of the overburden, fold to fill as closely as possible the "hollows". Since the latter occupy a small surface area (they can be nearly as deep as they are wide) the rocks must be subjected to considerable deformation. If they are pelites, a relatively plastic material, they molded themselves into all the available space. If, on

the other hand, they were sandstones, a more rigid material that cannot undergo large deformations without fracturing, we get a series of ruptures with piles of blocks and crushed material. In this case, the filling is first done in a less complete and more discontinuous fashion because the blocks can show some resistance. Each rupture passes the stress to the rocks above, which yield in their turn, so that the readjustments occur gradually. This difference in behavior, shown schematically in figure 1, can be seen notably on the front 119 (figure 3 in chapter 4), where we see angular displacements in the sandstones, although the pelites were subjected to considerable folding to come to mold themselves around the deformed parts.



*Figure 1. Difference in behavior of pelites and sandstones under compaction.*

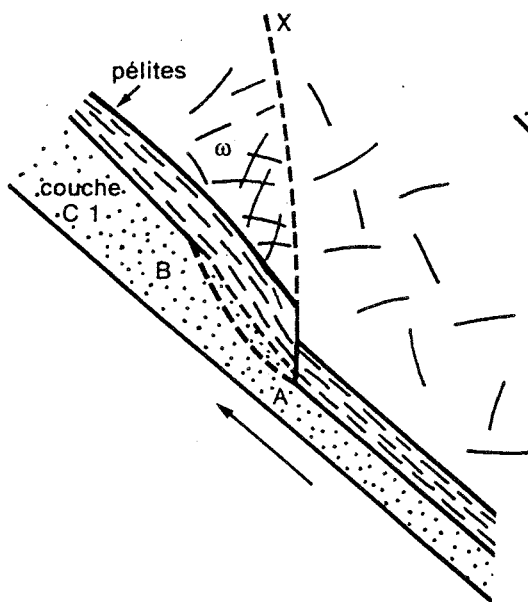
When the reactors were covered by a relatively thin bed of pelites and then by sandstones, the conditions are particularly favorable for a good compaction because the pelites form a kind of plastic joint, coming to flatten themselves out very closely onto the desilicified portions under the pressure of the sandstones that break away above them. The analysis always becomes a little more complicated when we have to consider simultaneously a rather steep dip and gradual elimination of the silica. Figures 2 and 3 represent schematically what happens when desilicification progresses by flow upward and downward respectively.

In the first case, the compaction is accomplished in a discontinuous manner, because the pressure is controlled by the sandstones above, which "unhook" in successive panels. Consider, in figure 2, the portion AB, which is in the process of desilicification. On the downward side, the compaction is effective, because the sandstones have collapsed, equilibration has had time to occur, and the terrain pushes with all its weight. On the upper side of the breaking plane Ax, the sandstones are still in place, but the hatched portion w, which is badly supported underneath, is largely deformed and has a tendency to detach itself although incompletely dissociated. The corresponding pelites are not totally under pressure and they are beginning to contact the sandstones. It is only when the block in question collapses, gradually causing new ruptures, that the pelites are going to come to pack down on the underlying strata. *With respect to compactions, therefore, there are*

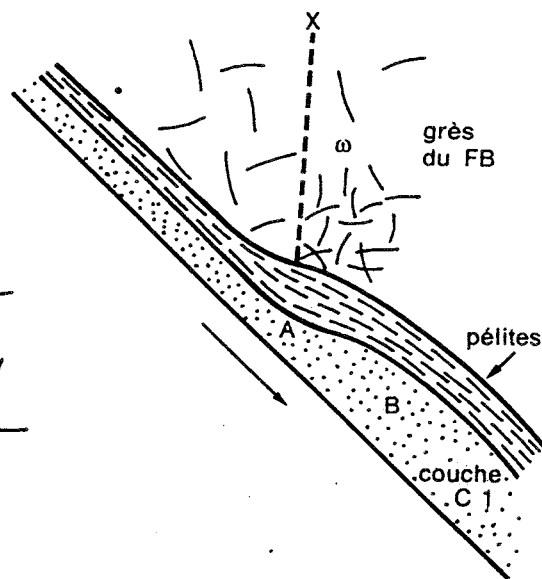
*discontinuities in the readjustments, and if the progression is slow, there is no generalized sliding, because the pelites are blocked at each stage.*

If the propagation occurs in the downward direction, the situation is totally different. We see in figure 3 that the sandstones that detach from Ax press strongly on the pelites and these are pushed downward to the extent that there are "voids" to fill. Consequently, when the portion AB is desilicified, even if the sandstones above do not come into position immediately, the compaction is effected through the means of the pelites, which transmit the pressure exerted by the entire collapsed sandstone lying in the upward direction. *They act as a go-between to the extent that the sandstones above collapse in their turn. In this case the compaction occurs without retardation and is done continuously and there is a permanent sliding of the pelites in the downward direction.*

We can further note that in the latter case, the sliding of the pelites has the chance to be more extensive when the progression is more rapid (compared to the time for equilibration of the overburden) and that by contrast, the propagation upwards takes place with discontinuities that are more marked when it is slower. If we then return to our observations of the terrain, we see that they do *not at all correspond to a slow progression upward*, but they evoke, rather, as in figure 3, *a relatively rapid progression in the downward direction*. This is, then, another reason to reject the operational scheme for the reactors that had been proposed.



**Figure 2.** Progression of Desilicification uphill.



**Figure 3.** Progression of desilicification downhill.

### 1.3

### THE SEARCH FOR AN ACCEPTABLE SOLUTION

Let us begin by drawing the conclusions of this discussion. Propagation towards the bottom is not only *the most appropriate direction* for desilicification but also that best consistent with the field observations. Moreover, if one considers that there was not an external "illumination", the most probable hypothesis is that of starting the reactions high in the zone. It is there that the ore is by far the richest, that the dip was the greatest, and that the "dome" structure was the most pronounced, leading to the greatest susceptibility of provoking the extensional stresses necessary at the first criticality. Finally, an equally important point, the scenario we just analysed for the case of progression toward the base is the case that best explains that compaction was achieved *without delay*.

Another conclusion is required: because the overall duration of the reactor functioning could not have been much longer than that of each reactor, we must conclude that *desilicification was very rapid*. We saw in chapter 12 that one could effectively justify, for plausible values of permeability and temperature, high speeds for removal of silica at least immediately below the reactor. We cited figures like 20,000 years, which is only a small fraction of the duration of reactor functioning.

If we now return to the arguments given in paragraph 1.1, it is evident that the margins of error do not allow us to draw conclusions that are too strict. We showed that the average amount of silica present in the core during functioning was practically none, but taking into consideration the margins of error, we can only assert that it was less than a very low threshold, such as 5% of the original silica. This does not mean that there was not more silica present when the neutron flux became significant, but only that the amount that remained then must have been eliminated very rapidly. In addition, the definition of neutronic parameters in general gives a lower weight to initial conditions. (For example, the spectral indices, which give us information about compaction, are measured from fission products that are gradually produced. The same is true for the ratio of radiogenic thorium to fission neodymium, because these two quantities are not present in the beginning.) The results do not allow us, therefore, to exclude *a short phase of coexistence*, most particularly at the start of irradiation.

Under these conditions, when we consider that desilicification was very rapid, we can ask whether it is really necessary to keep our initial hypothesis, namely that of a spreading out of functioning, matching a delay between desilicification and reactions. Even if we had to suppose that the reactions extended nearly instantaneously throughout the zone, could not a sufficiently short period of stabilization suffice to explain the measured results? We do not think so.

In the first place, a value such as that given above characterizes a speed of desilicification at the lower edge of the heat source, where the temperature gradient is highest, but we have seen that the speeds decrease very rapidly with separation distance. If the reactions had extended through the whole of the sandstones, we certainly could not justify an overall speed of desilicification of that order. In

addition, that speed varied greatly through time, because there the permeability collapsed with argillation. It takes infinitely more time to remove that last grams of silica. So, even if the majority of silica was removed in a short time, we must in any case consider a small residual percentage during a very long time, and that contribution could already lead to the limit of what is compatible with the measurements.

Above all, what we are concerned with is less the removal of silica than the geometric stabilization that accompanied it. If the reactions extended immediately throughout the zone, this would have been done in a chaotic manner, more broken, and with more significant deformations. It is much more satisfying to explain the results by calling upon a *regular progression* and a compaction achieved gradually. We think that a scenario such as that represented in figure 3 is necessary to explain the situation observed. In this case, the desilicification proceeds to the lower limit of the zone under optimal conditions and compaction is achieved each time without delay thanks to the mechanism shown. The values cited make it possible that the speed of progression was large enough to justify the conclusions of the thermal study and the phenomenon of gliding in the pelites, while being moderate enough to permit a good compaction at each stage. (We could allow, for example, that the duration of extension to the entire zone was of the order of one quarter to one half of the duration of functioning of the individual reactors).

It remains necessary, however, to show that *the reactions were prevented from extending faster than the desilicification*, and remained each time set back, and that the flux remained low in the parts not yet desilicified. We return now to our initial problem, but this time under less arduous conditions. In effect, if the desilicification is rapid, it is sufficient to justify a short time interval, and as we shall see, that is much easier.

We have emphasized that the effects of temperature hamper propagation. This remains true even without an amplification from the effects of convection currents. In the border regions, there is at the start a disequilibrium in reactivity, because the poisons are destroyed less quickly than in the core, while the temperature is largely determined by the neutron balance in the core. We showed in chapter 10 that this situation can evolve in either of two ways. If the antireactivity in the border is large, the flux is strongly lowered, the poisons are destroyed very slowly, and the disequilibrium is not only not absorbed, it is accentuated. If, on the other hand, the border is made more reactive, the flux rises, the destruction of the poisons accelerates, and it is possible to have propagation there.

Consider the situation at a given moment in the border. There, where the silica is not yet removed, the medium is largely subcritical because there are the effects of the silica, the poisons, and the temperature; therefore, the flux in the non-desilicified part is very low. This situation evolves very rapidly, especially if the ore is rich, due to the removal of silica, but there is in any case a delay, of which the kinetics is determined by the coupling between the destruction of the poisons and the neutron flux. If the desilicification is too slow, the flux rises before it is finished. This is why

we have imagined in our "false solution" the intervention of convection currents that permit the increase, by pushing them further, of the negative effects of temperature, blocking more firmly the propagation. If one assumes, on the other hand, that desilicification proceeds rather quickly, it is no longer necessary, because even if the delay is not very large compared to the duration of the reactions, it can be relative to the time needed to remove the silica. In this case, the integral of the residual silica by the flux, during the time when they coexist, may only be a very small part of the total.

We must note that this line of reasoning assumes that the compaction of the terrain is accomplished without delay, because an increase in the amount of water present in the neighborhood of the frontier brings with it additional reactivity that would extend the flux further. We have seen that this was the case. Another element must be considered. In all of the lower part of zone 2, the thickness of the rich bed (and with it the  $k_{\text{eff}}$ ) decreases with depth. This is also of a nature to retard progression. There was possibly also another factor that played a role, as was suggested in paragraph 4.3 of chapter 12. This consists of the possible leaching of boron at elevated temperatures in the ore, which began to be affected by radiation damage. Even if the boron was not redeposited, it was present in the circulating fluids. Consequently, as long as they were important, that is before the desilicification, there was a little supplementary poison in comparison with nonirradiated boron, which lowered the flux and inhibited the propagation. By contrast, the circulations disappeared nearly completely (they were diverted laterally) when the ore was argillized. All these reasons contribute to explaining that *the reactions remained always delayed* with respect to the process of desilicification.

#### 1.4 CHARACTER RELATIVE TO STABILIZATION

The scenario of descending propagation can suitably explain the observed situation, but one must make a few more comments concerning compaction. We know that desilicification of the borders of the reactions was less rapid; one could then argue that the core was not in direct contact with the pelites, but that it was through the intermediary of the sandstone that had not been completely desilicified. To this we can reply that since a certain degree of decohesion had been achieved, these borders could transmit pressures plastically, without the intervention of rigidity and significant retardation of compaction.

In their sliding toward the downhill direction, the pelites certainly had a tendency to entrain the neighboring argiles de pile. We can think that this phenomenon was limited, precisely because of the delay in desilicification of the border regions, and because below there were still rigid sandstones remaining, but nevertheless, the less stabilized the upper border was (because of contact with the pelites) the more immediately the lower border was flattened by the pressure against the adjacent terrain. To this difference, moreover, is added another coming from the fact that the convection currents were not only directed from the downslope direction upward, but also from the wall towards the roof (figure 11 from chapter 12), in a manner that caused a much greater contamination of the upper border region. It is this that

explains the observations found in numerous drillholes, and notably in SC36 where the curve of isotopic depletion of uranium remains very representative in the border of the wall, although it is difficult to explain in the border of the roof.

On the other hand, even if the compaction was done very efficiently and almost instantaneously in small dimensions, this does not exclude longitudinal *slidings* as we have just seen, and also *deformations* on a much larger scale, in particular in the lateral sense. These are very visible in the outcrop 2' (figure 4 of chapter 1), most particularly in the extreme south (region of cuttings 2'N, 2'N', etc.) where we observe essentially an overfolding of the rich ore zone. The profiles obtained by combining the indications from the different drillholes show all the important flexures. Particular spectacular, even disconcerting, is the mining front 30/6/76 in the base of the zone (fig. 4). One gets the impression that the reactor has been folded back on itself (it actually consists, based on the indications in the drill holes, of a strong lateral flexure that appears this way because of the oblique geometry of the front). We see clearly a whole series of deformations and glidings and even disconnections in a setting that nevertheless remains plastic.

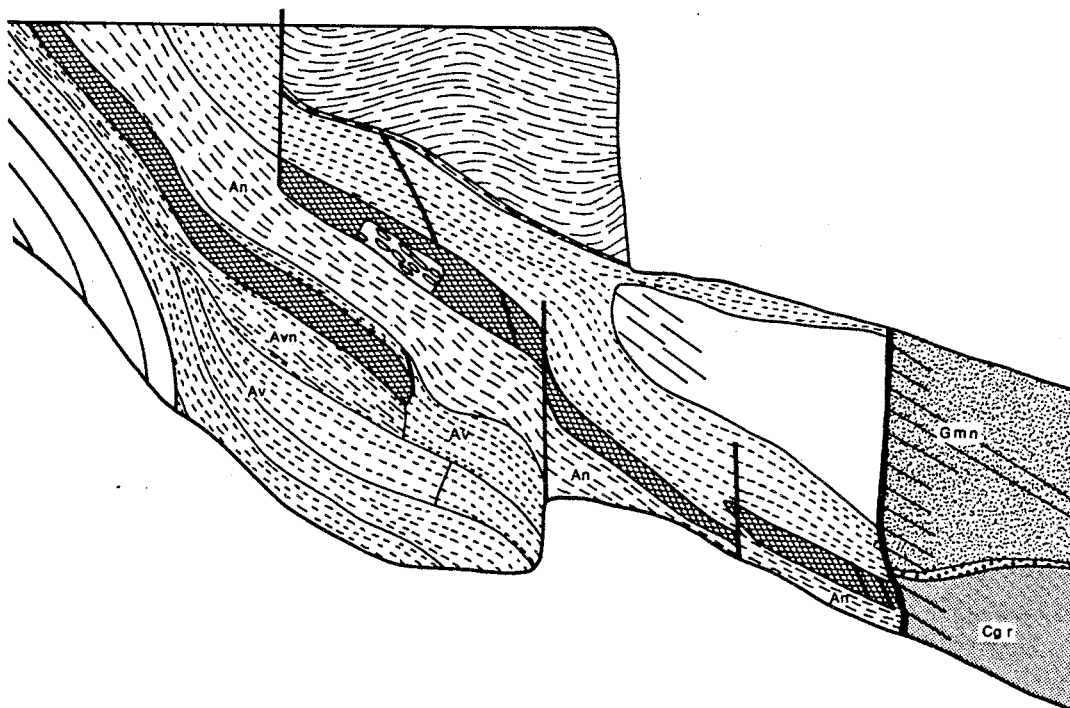


Figure 4. Front 30/6/76: base of zone 2 (after ref. 16).



It is then rather *a relative stabilization*, certainly much less perfect than we believed for a long while. It is because from the neutronic point of view we call upon essentially only the distributions perpendicular to the bedding that we had that impression of stability. It is an effect of the precocious nature of the compaction in the small dimension that has permitted coherence in the measurements along the drillholes. The deformations in the other dimensions have consequences that are less easily recognized.

Finally, it seems that this realization of a reactor environment "stabilized and protected" in the middle of an environment that was highly disturbed - which remains astonishing, even if it is necessary to make it relative - was only possible through a *concurrence of exceptional favorable circumstances*, which are only fully united moreover in zone 2 (and to a lesser extent in zone 1). These were: starting the reactions at the top of the zone, very homogeneous mineralization, dip sufficiently high to encourage convection and compaction but not so much as to cause faulting, optimal thickness of the bed of pelites playing the role of plastic "joint", etc. - and also, as we have seen, subtle adjustments of the different kinetics that come into play in the propagation.

In addition, it is probable that the scenario described is only applicable in a part of the zone. In effect, one can think that when the reactions started, they extended nearly immediately through all the very rich ore that constituted the upper part, causing relatively significant geometric upheavals. It is without doubt only when they overflowed to the underneath, that part in the form of a "tongue" in the process of thinning out (the only part that remained at the time of discovery of the phenomenon), that a more regular propagation, such as we envisaged, could start. We have a sort of proof of this in the problems that we encountered in the sections of the outcrop number 2, and that we did not find again a little lower on the outcrop 2' and above all in the drillcores. (It is thus that in the section 2P the reactor is terminated abruptly against the poor sandstones in which the reaction levels are very weak. There was clearly displacement. All the same, we did not understand well the section 2L that seemed cut in two, etc.). On the other hand, in the bottom of the zone, the slidings by accumulating assumed a greater importance, as shown in figure 4. It is therefore an extraordinary good fortune that the circumstances of discovery led to concentration of studies not only on zone 2, but on that part of the zone that is located in the most favorable position.

## 2 OTHER PROBLEMS OF THE NORTHERN SECTOR

### 2.1 FUNCTIONING OF THE ZONES ENCASED IN SANDSTONE (ZONES 3 TO 5)

In zones 3 to 5, there always is on both sides of the argiles de piles some non-desilicified  $C_1$  sandstone (chap. 1, fig. 7b). This situation must be put together with the fact that the conditions that produced the superconcentrations of uranium in the "rich sandstones" only affected here the central part of the mineralized bed. The ore values in the residual sandstones are actually, in general, of the same order as in the corresponding levels of "normal" bed  $C_1$  (certain drillholes outside the reactor, such as KN248 near zone 5, moreover, give a good picture of the distribution of uranium before the start of the reactions). As we saw in chapter 12, the fact that these sandstones were practically not desilicified proves that their average permeability was lower by at least an order of magnitude as compared to the reactor zone. This is not astonishing because the superconcentration of uranium is found along tectonic fractures. It is because these fractures were limited to the central part of the  $C_1$  bed that there was at the same time transport in of concentrated uranium and reduced desilicification at the exterior.

Although in zones 1 and 2 a larger extension of the mineralization and fracturing allowed the desilicified sandstones to become rather quickly in contact with the pelites, the reaction zone was here subjected to the pressures from the adjacent terrain through the means of sandstones that remained intact. Because of this we can think that compaction was not achieved under the same conditions.

The physics study has provided evidence that *a whole series of important differences* can be examined in the light of that observation. First, the criticality study in the final condition of the reactor showed that we could not account for this by considering the situation as homogeneous as for zones 1 and 2, and in vast portions we could only justify criticality by calling upon much larger amounts of water. Measurements of the spectral indices confirmed these two points. We found dispersed results and certain of these implied a very large average porosity (such as 60%). On the other hand, it is in these zones that the microthermometry of fluid inclusions showed elevated temperatures, or more precisely, low densities. These very low densities are acceptable from the neutronic point of view only if we assume that there was at the same time enough water, and thus a considerable volume available at the moment of trapping. All of these things make one think that the compaction occurred in an irregular fashion, that it did not instantaneously accompany desilicification, and that certain places and times there were very high values of porosity.

We have shown in chapter 6 that when we have very thin portions of ore that is very rich, which is often the case here, the reactivity of the setting can be more favorable before complete desilicification, especially if compaction does not immediately follow. The simulations presented in chapter 6 show that the starting can effectively be produced in the course of desilicification, and therefore in a medium not already stabilized. In this case, the temperature can increase strongly if the compaction follows with a measurable delay, but inversely, one can obtain a premature stoppage of the reactions when the volume available for water is rapidly reduced. We note effectively in zones 3 to 5 the existence of extended zones with weak levels of reaction. The reactions were stopped at a moment when the evolution of reactivity in the fissile nuclei and the poisons still had a positive result. One can only explain this by calling on an intervention of this sort.

It is necessary also to explain how one has at the same time transitory elevated temperatures (as indicated by the fluid inclusion measurements) and very moderate average temperature differences (as attested to by the duration of the reactions). These two results would be incompatible if we supposed that the porosity remained on the same order during functioning of the reactors. There are, therefore, a whole collection of arguments to affirm that the reactors encased in sandstone, contrary to zones 1 and 2, experienced *irregularities in their compaction*.

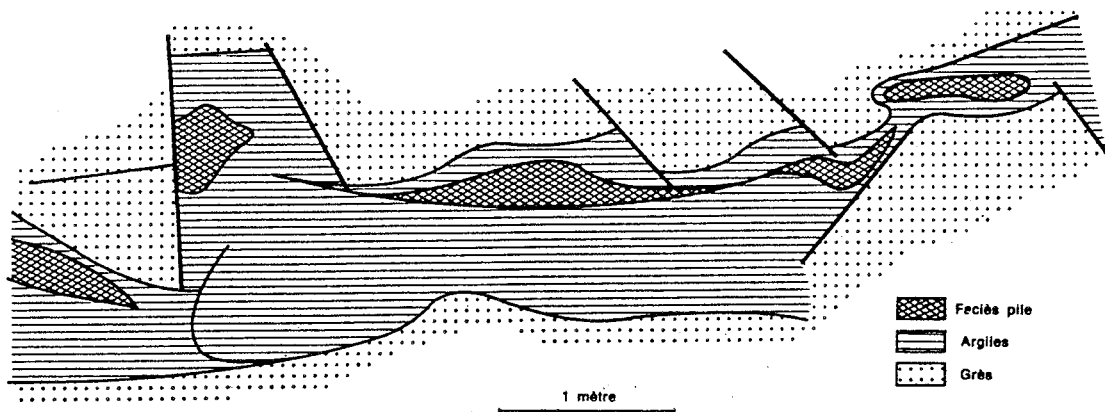
These irregularities can be explained by the behavior of rigid blocks that could resist for a certain time before falling, and not transmit completely therefore in the beginning the pressure of the overlying terrain, in contrast to the pelites that molded themselves immediately onto the sandstones, which were in the process of desilicification (fig. 1). A delay in compaction translates into a larger porosity and, therefore, into a lowering of the density of water. Inversely, a "recapturing" [rapid collapse] reducing the porosity translates into a prolonged lowering of temperature, or eventually even into a premature stopping of the reactions.

The field observations confirm completely this behavior. If one looks at figures 11 and 12 of chapter 1, one sees a series of faults. The blocks of sandstone glided along the fracture planes, and were driven more or less deeply into the argilites, (the blocks are on the scale of a meter, and the relative displacements on the order of a decimeter). It is clear that this was a tectonic readjustment, the consequence of the desilicification. The fractures were produced by the pressure of the overlying terrain, and the relative displacements translate into the differences in compaction. One easily sees that this occurred in a discontinuous manner.

The existence of these delays in compaction can, moreover, be considered as a new proof that at the moment of the reactions *the overburden was moderate*. If the pressure had been very high, it is probable that even the sandstones would have behaved plastically.

Figure 5 shows a detail, particularly "in an uproar", of the outcrop GL6: even where the faciès pile is continuous, the displacements are visible in the sandstones that are

driven into the argile de piles, but there are also displaced portions of the reactor and even an isolated piece. It is evident that the reactor could not have functioned with that geometry. At the same time, certain portions are very thinned and have undoubtedly been laminated by the blow. These observations tend to confirm that the functioning took place in a *non stabilized medium*.



*Figure 5. Detail of the outcrop GL-6 (zone 5).*

This behavior equally had consequences on the propagation of the desilicification. In the first place, the fact that the reactions took place in sandstones that were incompletely desilicified and not yet compacted, but with significant permeability, and that the temperatures could have been momentarily very high, favor convection. On the other hand, the currents, following the fractures, which constituted a by-pass, could find themselves not saturated at the top of the reactors, especially if the temperature reached the region where  $dCs/dt$  is negative, and all the more so if there had been vapor. On the other hand in the very rich sandstones it was enough to remove a small fraction of the silica, as long as the compaction was not immediate, for criticality to be reached. The reconciliation of these remarks leads to being able to more easily justify, in this environment, the possibility of propagation in all directions, *including uphill*.

Correctly, we noted in chapter 12 (§3.2a) a certain number of observations where it seemed that there had been desilicification above the reactors. This always involves the zones encased in sandstones. On the other hand, we see later that it is improbable that in zones 3 to 5 the reactions started at the top. One is then led to suppose that in this case the propagation was not exclusively toward the base or laterally, but that it could have been also upward.

We even wondered, to justify criticality at the end of the reactions, if in certain cases these were not stopped before complete desilicification, the situation for reactivity

having been more favorable then. It was necessary to suppose then that the elimination of quartz had continued after the stop of reactions, because we only find completely argillized faciès pile. That implies almost certainly a progression towards the top, because one can only think that those currents induced at the downward part of a functioning reactor have the capacity to carry to completion desilicification. In fact, table 11 in chapter 6 shows that even for a rich zone as thin as KN245, we can justify final criticality through an entry of enough water, without residual silica. With this discussion of the speed of desilicification in mind, we can think on the other hand that, in zones 3 to 5 the duration necessary to remove silica certainly represents a much greater fraction of the total duration of operation than in zone 2, but that nonetheless, desilicification must in general have been finished at the time when the reactors stopped. There is not, therefore, possibility to deduce from this a constraint on the direction of propagation.

## 2.2 START OF THE REACTIONS

We saw in chapter 6 that obtaining criticality in the sandstones not yet transformed by hydrothermal circulation required very restrictive conditions. It was necessary to have a sufficient volume of very rich ore (of a sort that only limited portions of the reaction zones could provide), and above all it was necessary to have the entry of significant amounts of water (the porosity had to approach 15%). Such a high value can, without doubt, only be achieved through a tectonic episode that caused open fractures to appear.

The date of the reactions - 1950 million years - is also the date of metamorphism of the neighboring Ogooué series (intrusion of pegmatites) and we noted that the coincidence is probably not without significance (chap. 4, §1.1). F. Gautier-Lafaye showed that at that time a whole series of events affected the Francevillian - what he calls the "major tectonic phase" (ref. 36) - and that almost surely a similar event occurred further to the west. In addition, we noted that the pressure, which probably was unequilibrated during the maximum of burial, seemed to return practically to hydrostatic during the period of the reactions, and that this reequilibration, connected to the uplift of the basin, could have been facilitated by large scale fractures. The start of the nuclear reactions seems then to occur in *a context of tectonic events of very large scale*, and it is probably the consequence of this.

It is probable also that it was at that time that the superconcentration of uranium was produced, which formed the "rich sandstones", the fractures serving as the drains for the mineralizing fluids. It was then possible here to have a certain simultaneity in the realization of the two requirements for criticality: concentration of uranium and entry of water. However, if it is clear that when that supplementary uranium was deposited there were fluid circulations, and thus water, that does not mean that there was enough water. The cracks do not necessarily produce openings of the order of magnitude indicated, especially if they resulted from shearing or compression. There must be an extensional setting.

In this respect, the study of the Oklo ore deposit shows that the rich sandstones were put in place along large shear zones oriented NW-SE. We noted that in the sector 1-6 they were in one piece (because the areas of rich ore connect group 3-4 to group 5-6 as well as zone 2 to zone 1, and that zones 6 and 2 are connected). The ensemble has the indicated orientation and is aligned with other zones of rich sandstones. But in addition this sector was structured by transverse faults (called by the miner "crochon", "dôme", "dressant") that could have caused distensions. These are undoubtedly the structures that played the principle role in starting the reactions.

A tectonic episode generally consists of successive phases, in the course of which the faults can remobilize and give rise to wider openings. As we previously noted, criticality could then have been achieved either by a last transport of uranium into a zone already open, or by a widening in a zone already very rich in uranium. Do we have information that would allow a choice between these possibilities?

Some people have thought that the uranium was emplaced not only just at the start of the reactions but also during the reactions, and even that its transport could have been strongly increased by the convection currents (ref. 36). Concerning the last point, note that the "thermal siphon" invoked constitutes a very modest engine compared with regional circulations. It could arguably provoke local remobilizations (we will give some examples later), but it undoubtedly could not by itself collect much uranium from long distances. The thermal study leads us to think, furthermore, that at the time of the reactions, the regional circulations were no longer very intense, since they did not seem to have carried away a significant fraction of the heat. But in any case, since we showed that the extension of the desilicification was very rapid, that emplacement of uranium after the start of the reactions, if it occurred, could only have been very limited.

Consider particularly zone 2, with the scenario for operation that we have assumed. We saw that it is the early argillization that prevented heterogeneities that we would have noted if the new uranium had been able to be introduced in the heart of the reactor during functioning. That does not exclude that the uranium was still being added to the lower side, since we assumed a stepwise progression downward, but it could only do that for a short time and only in the part of the zone that was the least rich. It is therefore necessary to admit that this enormous accumulation of uranium that constitutes the upper part of zone 2, and in which the reactions were very rapidly spread, was already there in essence when the reactions started.

Actually, one could be surprised that in such a rich ore the reactions had not started earlier, during the course of its emplacement. It is evidently because beforehand there was not enough water. In a sufficiently open environment, there comes a moment when an increase in uranium is no longer beneficial, because it accentuates undermoderation. On the contrary, it is when the ore is very rich that the entry of water is the most effective. We think, therefore, that at this stage it is the entry of water, rather than transport in of more uranium, that played the decisive role. Moreover, the top of the zone is not just the richest ore, it is there where the structure of the "dome" is most clearly developed. It consists of a vertical thrust that in making the bed bulge caused distensions. It is undoubtedly a later thrust that rendered the medium critical.

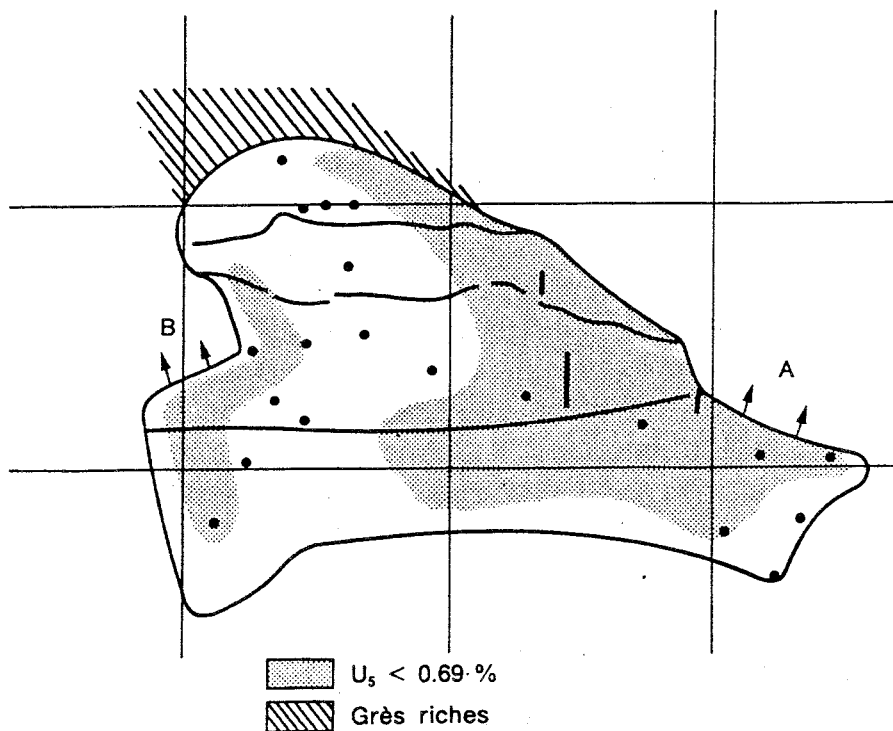
We are then tempted to *favor tectonic effects in the starting of the reactions*. In these conditions, the last fractures could have been produced on "the heels" of the end of the period of enrichment of uranium, but we think that one can not exclude that they were produced a little later at a time when the deposition of uranium had already ceased.

This argument does not apply to the other zones in the sector 1-6, which are much less rich, and where the early argillization could not be established so clearly, but the same conclusion is suggested for different reasons. It is essentially the functioning in a stabilized environment that, in the case of zone 2, led us to the idea of a propagation mechanism. But that idea of propagation imposes itself also in the case

of the other zones, because the portions where uranium accumulation is sufficient for spontaneous starting are very few. We note that these small special portions are, in general, located with the tectonic structures we have discussed. The fractures have also, of course, favored the transport of uranium, but we can think that there again it is the entry of water that played the decisive role. Let us examine where these privileged zones are located.

Concerning zone 1, the major part of what remained at the time of discovery of the phenomenon consisted of ore grades too low for one to envisage criticality before a thorough desilicification. In contrast, the upper part had very rich ore, a steeper dip, and deformations and fractures connected with the proximity to the "crochon" fault. The physical study suggests on the other hand that the functioning was analogous to that of zone 2. One can imagine there, also, a startup due to a new tectonic thrust, and a propagation *downward* under the enclosing pelites, as described above (with, however, some peculiarities as we will see in the following section).

Let us examine now the case of group 3-4. In figure 6, we noted the portions where the isotopic depletion produced a concentration of <sup>235</sup>uranium below 0.69%. We see that they form two groups in one piece (which justifies in hindsight the distinction made between the two zones in the beginning), with a large band of desilicified ore at their junction where the isotopic depletions are weaker, and for which some could have been due to contamination. This suggests two distinct "reactors" that, in their propagation, met and became more or less connected. Each of them possesses a



**Figure 6.** Zones 3-4. Regions of significant isotopic depletion.



small, strongly tectonized, region (A and B), with relatively rich ore and elevated levels of reaction, which bring together a certain number of favorable characteristics for the start of the reactions.

In these two small sectors, in effect, the bed is strongly tilted. At A, it consists of a very strong flexure, which is the one called the "dressant". One recognizes it in particular on front 127 (figure 12 of chapter 1), where we see the bed literally plunging vertically, cut into several pieces. We also found it in KN230 where the drillhole follows the rich bed for a large distance because of its near verticality. At B it is a series of very marked detachments that produce locally elevated dips (fronts 123 and 128, figures 23 and 27 in reference 16). These faults are capable of having caused distensions, and one can estimate on the other hand that a subvertical opening would close less quickly under pressure of the terrain, because it would be less directly applied. Outside of these two small sectors the conditions are much less favorable for starting the reactions, because the thickness of the rich ore is less and the tectonics are more calm.

In this hypothesis, the reactions were propagated from A and B mostly laterally and towards the base but a little also towards the top. We saw in the context of the encasing sandstones that propagation is facilitated in all directions, and that we could not exclude that it was also towards the top. (The upper limit of the zone is poorly known, but the drillhole SC51 is clearly uphill from point B). At the base of the zone, the front 185 showed a complex situation where the argiles de piles and the non-desilicified, rich sandstones coexist (fig. 30 from reference 16). These rich sandstones, moreover, extend towards the downward direction (without being

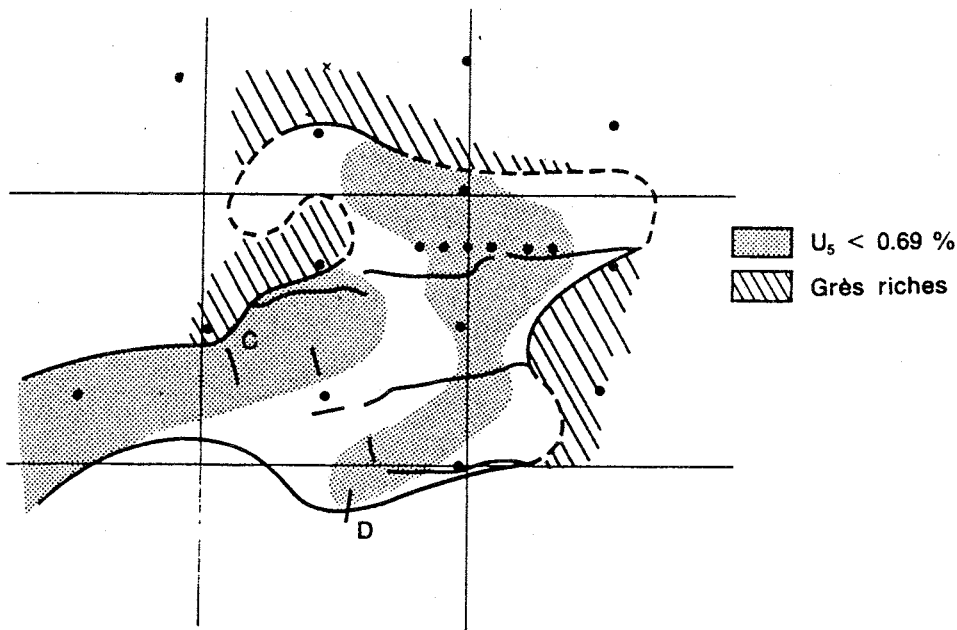


Figure 7. Zones 5-6. Zones where isotopic depletion is significant.

limited by a fault as is the case at the base of zone 2). These observations are compatible with the arrival in this region of reactions propagating toward the base and stopping because the levels of uranium in the ore were insufficient.

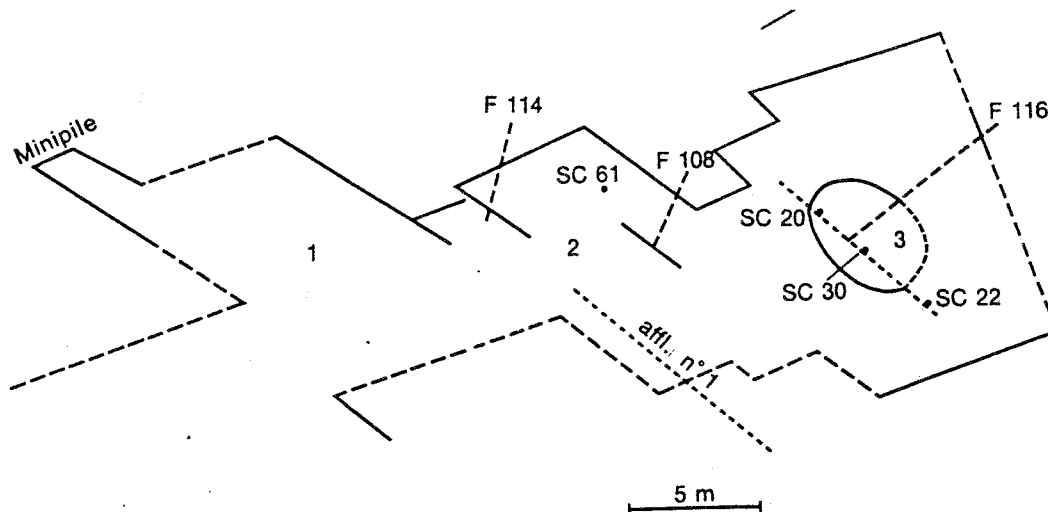
A similar analysis can be done for the group 5-6. There, too, one can distinguish two groups in a single piece with isotopic depletion significant in the interior of them (fig. 7). We also can recognize two small sectors (C and D) where the bed is strongly tilted, but these are not really comparable to the previous case. The first was recognized in the mining front of 30/9/76 (fig. 2 from chap. 4). It is there that we find the largest isotopic depletions, and it is possible that there was a significant flexure here, but the environment is actually not very well known. As for the situation shown in D by the front 119 (figure 3 from chapter 4), it seems to correspond rather to the arrival of the reactions bumping into the fault that bounds the rich sandstone sector. In this case, it was sliding along this fault after desilicification that tilted the bed.

Nearly all of zone 5 contains only small thicknesses of rich ore. It should be noted that the rich sandstones rap themselves generally around the zone. In several places it seems that the reactions stopped because the thicknesses or the ore grade became insufficient. It seems that we cannot exclude that the reactions propagated by fanning out from a single ignition point (which could have been point C) and that they "stepped over" the poorer band that separates the two groups in figure 7. In reality, we are missing too much information from this zone to be able to reconstruct its history. We will return a bit later to the case of zone 6.

## 2.3 THE CENTRAL REGION OF ZONE 1

Zone 1 shows many analogies with zone 2. As in zone 2, the sandstones of bed C<sub>1</sub> were completely desilicified, and it has then operated beneath an enclosing pelite. We supposed in the preceding section that the reactions had started in the upper part and had progressed downward according to a mechanism analogous to that for zone 2, which is of the type that supports a rather good homogeneity in the physical study results. It shows, however, several important differences from that region: the dip is clearly less, the ore is less rich and more irregular, the zone is broken up into a series of compartments, and finally, the bed of pelites between C<sub>1</sub> and the FB sandstones is noticeably thinner (30 to 40 cm, as can be seen in the neighboring drillholes). All this explains that the propagation was *more irregular*, and that the stabilization of the reactor environment was a little worse.

A small portion of this zone shows remarkable peculiarities. This is the region of SC30. It is framed by two lines of strike and outlined by the drillholes SC19 to SC22 (fig. 14 of chapter, and figure 8 below). The rich bed having been strongly moved left, the dip is particularly shallow here. This region poses two problems (which have been mentioned several times). The amounts of clay are enormous, much larger than that which could come from desilicification of the sandstones of C<sub>1</sub>. Secondly, significant amounts of uranium seem to have been displaced.



**Figure 8.** Schematic diagram of the structure of zone 1 (after Gauthier-Lafaye).  
 1. Region of rich ore exploited before the discovery  
 2. Region of SC 61      3. Region of SC 30

The thickness of the mineralized clays, among which we can consider as being the base of  $C_1$  and the sandstones of FB, sometimes reaches four meters in this region. Total desilicification of the sandstones from  $C_1$  can give a maximum of 1.2 m. We saw that there are also great thicknesses of clays at the base of zone 2, but they consist essentially of pelites that slid downhill. Here the pelites are thinner, we are in a region that is nearly flat, and nowhere are the clays identified as those, if only because they have elevated levels of uranium while sedimentary pelites are practically always sterile. We discussed in §4.2b of chapter 12 the possibility that new silicates were formed from the silica from the quartz. Although this possibility cannot be excluded, we concluded that it was not quantitatively important. We were then led to the conclusion that the clays in this sector come from the *desilicification of the FB sandstones*.

In this hypothesis, what would become of the pelites? As we said, they are hard to identify. There is often a continuous region of very rich ore ( $> 10\%$ ) over the core for more than a meter. But above all, the pelites normally constitute an impermeable barrier to the convection currents. It is the pelites that prevent desilicification of the FB sandstones. We then thought, considering their thinness, the pelites had been able to be nearly completely eliminated, pushed downward during the collapse of the overlying sandstones, putting these in contact with the  $C_1$  sandstones. This does not exclude that higher up, where the dip is steeper, there could have been a generalized slippage of the pelites according to the model in figure 3. But they could not do this in the region in question because they were blocked in the passage by tectonic faults. Figure 8, which illustrates schematically the structures of the zone, shows clearly that for the very rich portion exploited in 1971, as for the little region of SC61 (where the distribution of reaction levels resembles most those of zone 2), there are disconnections at the lower edge that have probably interrupted the process of descent. In other words, we could have here at the same time continuity of

desilicification, and *successive stages in the tectonic readjustments*, because of the compartmental character of the zone.

Our idea is then that in the region of SC30, taking into account that special conformation that made it so that the sandstones already detached above could not press directly on the terrains that were not yet stabilized, there would have been, following probably a temporary delay, a *brutal collapse* of the roof sandstones, which would have pushed the pelites downward and crushed the reactors under an accumulation of very fractured blocks. These provided innumerable bypasses for the convection currents, allowing them to desilicify external regions. Judging from the thickness of the clays, the volume involved is enormous.

The FB sandstones are not generally mineralized. The fact that there is a great deal of uranium in these clays constitutes a second problem. It is almost obligatory to assume that the uranium is emplaced *during the course of the reactions*. There are considerable quantities involved. At the level of SC30, they were apparently higher than those that existed initially in bed C<sub>1</sub>. Figure 9 shows the partition of the "supplementary" rich ore as it is revealed in the series of drillholes.

An independant proof of the idea that it involves displaced uranium was discussed in paragraph 2.2 of chapter 3. We indicated that one finds in the two drillholes SC30 and SC30bis, over more than 1.2 m, an isotopic composition that is almost perfectly uniform (0.687%). This consistency suggests that it is probably depleted uranium from the reactors that was homogenized in solution, carried by convection currents, and redeposited thanks to the still-intact reducing potential of the FB sandstones. This is the clearest example we have found of a displacement of uranium in the

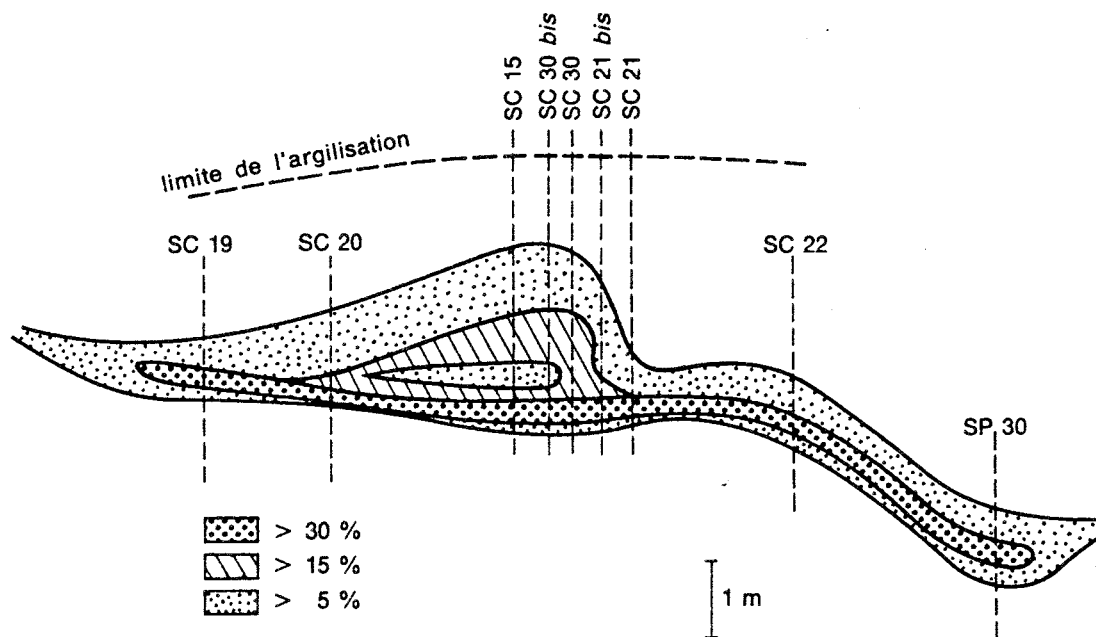


Figure 9. Partitioning of uranium in a cross-section of zone 1.

course of the reactions. It is explained by the exceptional importance of the fractures (and thus of the induced currents) formed *outside* the source of heat, this being as usual rapidly blocked by argillization. (It is essentially the border uranium that has been displaced).

These observations can be integrated into a *scheme of operation*. Because it is depleted uranium that was deposited, this implies that there had been reactors operating below the zone in question at the moment when this one had not yet been desilicified. This is not contradictory with the hypothesis of propagation towards the base, because we know that this was done very rapidly, while on the contrary, the desilicification progressed more slowly at the borders. (It is even more true here because to the C<sub>1</sub> sandstones was added the considerable thickness of the FB sandstones fractured later). Neither can this be considered as a desilicification above the source of heat, because we have noted that the spreading out in time was very relative. At a time when the reactors had already reached further below, the reactor to the right of the portion considered continued to operate, and maintained the gradients permitting desilicification of its own borders.

This chronology has been confirmed by the study of distributions of reaction levels in SC30 and SC30*bis*, presented in paragraph 3.2 of chapter 7. In effect, we showed that the curves (at least the most significant parts, because apart from that there is only contaminated uranium) could only be explained if one assumed that during the major part of the irradiation *all of the uranium* was there, and that on the other hand, the silica had not been eliminated quickly. In other words, the uranium was put in place as soon as the sandstones fractured, and the desilicification would have taken much more time. The uranium closest to the core (emplaced when it had already been partially depleted) received besides an additional irradiation because there is continuity between the core and the part with uniform isotopic composition.

This small region is then particularly interesting, and the corresponding drillcores very precious, but it is a pity that no geologic samples were taken (the front 116 is lower). We can regret even more that the project to maintain *in situ* a cutting through zone 1 in the bottom of the quarry was not carried to completion.

## 2.4 PROBLEM OF ZONE 6

Zone 6 *sensu stricto* is that thin band that is detached from zone 5 and was raised obliquely to come to connect more or less with zone 2. In the base, we find ore encased in sandstone as in zone 5. In the upper part, by contrast, the desilicification of C<sub>1</sub> was total as in zone 2. We said in paragraph 1 that we imagined at one time a propagation of reactions towards the upward direction, which would have continued as far as the connection, but that we had to give up this hypothesis. Is it necessary to think the opposite, of a propagation toward the base of the same sort as in zone 2, or can we maintain the idea of an extension toward the upper part from zone 5?

This zone presents several peculiarities that can appear embarrassing. In the first place, we notice that immediately below the band in question, we have poor ore that remains entirely sandstone (and with normal uranium), while above (where the ore is equally poor) there is a large band where the  $C_1$  sandstones are totally desilicified and where the uranium is depleted isotopically through contamination (figure 10). It is not easy to justify a *dissymmetry* so marked, if one thinks that the desilicification was done preferentially toward the downward direction from the heat source. (It is rather that observation that from the beginning supported the opposite idea).

We think now that this situation resulted in reality from the combination of two independant circumstances. If the border below the passage is absolutely not desilicified, it is probably because the fractured passage that served to drain the mineralizing fluids is terminated by a tectonic fault that separates an intact compartment, with a permeability at least an order of magnitude lower, from the broken up zone. This situation has effectively been observed on the front 10/1/76 (figure 11 of chapter 1) where we see zone 2 abruptly terminated against a fault, with perfectly sound sandstones on the other side (see also figure 4 above). It is possible that the same fault extends to the border of zone 6. Near zone 5, in any case, the fact that KN251 and 252 which are extremely close to the reactors remain sandy can only be explained by the interposing of a tectonic fault.

On the other hand, if the sandstones of  $C_1$  lying above the reaction zone have been desilicified, it is probably because, considering the dip, they became detached from the reactors at the time of desilicification to come to fill the "voids" creating many fractures that have been used by the convection currents. The explanation would be quite similar to that we suggested for the FB sandstones in a part of zone 1. It would

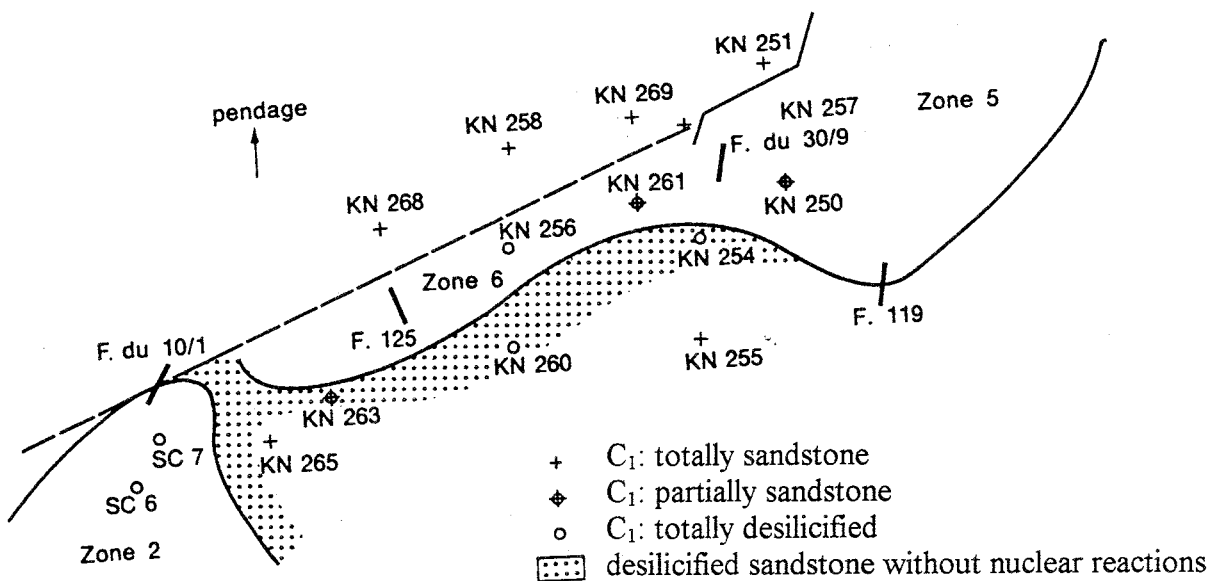


Figure 10. Zone 6: dissymmetry between the upper and lower borders.

also be the same for the chronology (this desilicification would have been done more slowly, at the time when the reactions had already extended throughout the zone). The contamination by slightly depleted uranium would also have been done under similar conditions, except that the amounts are smaller and the uranium came to be emplaced in sandstones that were already mineralized.

Furthermore, the situation observed in the cutting intersected by the three boreholes KN258, KN256, and KN260, is also disconcerting. In the first, on the downhill side, where one has normal ore, the thickness of the pelites is 1.70 m. One finds, moreover, a thickness of the same order in all the normal boreholes on the upward side as for the downward side of zone 6. (We note in passing that this thickness at Oklo increases from north to south, because it is only 30 to 40 cm in the neighborhood of zone 1, and on the order of a meter near zone 2). At KN256, which crossed the core, not only is there a great thickness of clays coming from C<sub>1</sub> (nearly 2 meters), but we find four meters of pelites. And the most astonishing thing is that we find the same situation in KN260, a poor drillhole entirely desilicified (three meters of pelites). One can be astonished by such an accumulation in the high part, because KN260 is several meters above the core (even if it is a little under the top of the zone).

This can only, in effect, be a result of general *sliding*. We find a large thickness of pelites in KN254 (which is, like 260, an entirely desilicified poor drillhole), and what is more astonishing is in KN261, which has preserved sandstones of C<sub>1</sub> over the reactor. On the other hand, front 125, on the top, only shows a very reduced thickness of pelites. A displacement so large is surprising, considering the size of the pelites in the zone and the small difference in height in the direction of the sliding.

It must be emphasized that the information necessary to try to understand the functioning of zone 6 is very insufficient. We only encountered two frontiers on the upward side and the downward side, (front 125 cuts neither of them) from which arise the speculations to which we had to resort to concerning the borders. We also do not know the transition between the lower part of the encasing sandstone and the upper part of the encasing pelites, and one is even not certain whether there is continuity. Above all, we do not know the possible connection between zones 2 and 6. At the level of GL1, we know there is an interruption (and moreover there is normal ore again in KN265) but one also knows that the firing 125, a little below, mined rich ore between the two zones. We do not know to what extent this ore was desilicified, if there had been reactions, and if the case arises, what was the extent of the interruption between the zones. Despite the importance of the missing information, we can risk proposing a scenario.

The base of zone 6, as far as KN261, resembles completely zone 5. We know that there were elevated reaction levels at the front 30/9 just at the edge and we even suggested that it could have been a starting point for the reactions. We don't see then why these would not have extended into this little portion of zone 6, which is, moreover, nearly horizontal. On the other hand, it is much more difficult to suppose that the reactions had continued their progression to the top, giving rise to so large a

desilicification (all of C<sub>1</sub> from KN256). And above all it would be hard in this case to justify the movement of the pelites.

We are then tempted to think that, in most of the zone, the progression was done *descending from zone 2*. The "jumping over" that we found implausible in an upward progression is much more acceptable in a descending one, considering the fault encountered on front 10/6. The convection currents induced by the reactions when they arrived at the base of zone 2 could only have come from this fractured corridor containing still rich sandstones. If the desilicification had been sufficient to restart the reactions, these would then have continued following the same scenario as in zone 2.

In this hypothesis, the sliding of the pelites is much easier to understand. It is the same process that starts again, beyond the turning point, but with a pressure of terrain that, from the new start, is considerable. Figure 3 showed in effect that all the sandstones that detached above continue to press during descent through the intermediary of the pelites. It is not, then, the few meters of level difference of zone 6 that must be taken into consideration, but the difference between the height of zone 2 and zone 6, which is close to 25 meters. This is an *enormous pressure* that continued to be exerted throughout desilicification.

Finally, this hypothesis leads us to conclude that *all the zones encased in pelites (1, 2, and 6) functioned the same way*, which justifies the homogeneity of the observations and the results of the physics studies. On the other hand, the real connection was not between zones 6 and 2, but in the base of zone 6 at the junction of two different modes of propagation. The singularity of KN261 would then be explained by the fact that it was situated at the place where the descending front would have encountered a region already desilicified but badly stabilized, which caused a significant geometric upheaval.

All that, of course, could be disputed. We are aware that our conjectures concerning reconstitution are fragile, especially for zones 5-6 where the information is insufficient. But we think that the idea that underlies them is well-founded, namely that in all this section *the ignition points for the reactions were less numerous, and that the phenomena of propagation tied to desilicification played the essential role*. It is these concepts that justify the continuity of reaction zones over distances without common scale with the diffusion length for the neutrons. As we shall see, it is not sure that events were the same in the south.



## 3 SECTOR OF ZONES 7-9

### 3.1 REMINDER OF CERTAIN PECULIARITIES

We were led, on several occasions, to discuss the peculiarities of this sector, and each time it seemed that these could be interpreted by calling upon functioning conditions closer to the surface than that for zones 1-6. Let us recall and comment on these observations.

a) First, it appeared that the maximum temperatures achieved during operation had been nearly certainly much lower than those of the north sector (chapter 4, §3.3). In effect, the measurements from fluid inclusions gave average homogenization temperatures lower than about 150°C. In addition, an isotopic analysis of lutetium confirmed the existence of a difference. A whole series of observations led to the same conclusion. The desilicification of the core was less advanced than in zones 3 to 5 (which have comparable geometry), because there is residual quartz and even sometimes intact sandstones in the core. One finds, on the other hand, organic matter very loaded with uranium not segregated at the heart of the reactors, which seems to be incompatible with elevated temperatures. Finally, the absence of magnesian chlorites would also be explained in this way. There are, therefore, a group of consistent indications. We are led to say that the temperatures could not have exceeded 250°C in this sector.

If we assume that the ambient temperature was of the order of 150°C as for zones 1 to 6, and that the pressure was the same and that conditions were far from boiling, this leaves only an extremely constrained margin for the neutronic control of the reactors. The greatest lowering of maximum density could hardly exceed a quarter of the margin used in the other sector. The conditions being otherwise comparable, one does not understand why the requirements of control had not involved, at least in certain cases, greater increases in temperature. On the other hand, it is equally difficult to justify, for differences in temperature that are so small, and so for convection currents that were even more limited, the disappearance of considerable tons of sandstones outside the reactors.

If, on the contrary, we assume that the reactors functioned a few tens of meters from the surface, this hypothesis implies that the ambient temperature was clearly less elevated and the pressure weak enough that boiling could occur at low temperature (such as 250°C for example). It is then boiling that could limit the rise in temperature (and therefore one could bring to bear an effect of a "ceiling" analogous to that observed in the northern sector). In this hypothesis, we use for control a larger margin in the single phase, and above all the possibility for supplementary lowering of

the density by vaporization. On the other hand, these conditions allow us to explain at the same time the weak desilicification of the core (chapter 12, §3.2a) and the removal of large volumes of sandstones as we shall see soon.

b) A second group of observations concerns expressly the details of the desilicification. Not only is it paradoxical, by contrast with the limited nature of desilicification of the core, that considerable tons of sandstones have disappeared, but the fact that the zones of desilicification are geometrically separated from the core is only encountered in this sector. Figure 11 shows for example a band of argile de pile separated from the core by sandstones that remain intact. We note also the existence of desilicified portions at the end of fractures, as well as the peculiarities of the "wall figures" (chapter 4, §3.3). The study of convection showed us that in a single phase the portions of the ore susceptible to desilicification could only surround very narrowly the sources of heat. On the contrary, these observations could be explained if there had been boiling, trapping bubbles of vapor (using the fractures, especially), and dissolution of silica at the time of condensation of the vapor. This is what was shown in paragraph 3.2c of chapter 12.

We must consider, however, whether this explanation is consistent with another result. The duration of the reactions in zones 7 to 9 were not shown to be particularly short. This means that not only could the amount of energy possibly carried away by vaporization have been only a small fraction of the total, but also - considering the reduced dimensions of the reactors - that the average  $\Delta T$  remained extremely modest. Even for a limited pressure of some tens of bars, the boiling temperature is of the order of 200°C above ambient. The duration of the reactions force us to admit that the reactors functioned most of the time with differences of temperatures clearly lower.

This remark does not exclude that there could have been boiling at certain times. This was necessary if the requirements for neutronic control were comparable to those for zones 3 to 5. In effect, the decrease in density up to boiling in the conditions assumed here is only half that assumed for the northern sector. We showed, on the other hand, for zones 3 to 5, that when the compaction was sufficiently irregular, the temperature data were not incompatible with a lower average. But the problem that occurs is to decide whether vaporization could have a size sufficient to explain the observations.

Note first that the enthalpy needed to lower the density of liquids through partial vaporization could have been at least equal to and possible greater than that giving the same result through raising temperature, if we assume that the amount of vaporization was augmented by migration of the bubbles. We can then allow that, even if the heat carried by convection remained very minor (for example 15% of the total), a non-negligible fraction of that (for example 3% of the total) could have come from vaporization of the water. But, even such a limited contribution is enough to explain the volume of sandstone that disappeared.

In effect, the weight of uranium 235 missing from zones 7 to 9, known from the passage through the factory of the extracted mineral, is 48 kg. Returning to the time of the reactions, we deduce that the fission energy liberated was of the order of  $10^{16}$  calories (being 1,300 MW-yr, as compared with 16,500 MW-yr. released from the northern sector). One could vaporize  $7 \times 10^8$  liters of water with 3% of that energy. If each liter vaporized dissolved at the time it condensed 0.4 g of silica (saturation limit at 220°C), that would be 280 tons, corresponding to desilicification of about 120 m<sup>3</sup> of sandstones. This is about the order of the volume of sandstone eliminated under abnormal conditions, that is to say outside the reactors.

A vaporization of this magnitude is then quite plausible under the assumed conditions, and it is in fact the only explanation that one could propose for these strange occurrences of desilicification. It implies that the pressure had been low and that the overburden was slight at the time of the reactions.

c) A third oddity of the zones 7 to 9 is the impossibility to justify criticality in the geometry in which we find the reactors. The importance of this result comes from the fact that the calculations of criticality have been perfectly consistent on zones 1 and 2, and cannot therefore be suspect. It is by comparison that this impossibility is appreciated. In the case of zones 3 to 5, we noticed that the conditions were not always comparable, but that we could make do here by supposing the desilicification, and above all the compaction, had not finished at the start of the reactions. But for zones 7 to 9, which also have an encasing sandstone, we remain far from agreement even playing to the extreme the supplementary degrees of freedom (chapter 6, §3.2c). We also cannot arrive at an explanation by invoking a departure of uranium, or by calling upon the intervention of the organic matter. We are then obliged to conclude that the geometry of the reactors has not been conserved.

This is a very important conclusion. It is obviously necessary that the reactors were critical; this is incontestable. Why then would there have been geometric modifications beyond those we find in the northern sector? In the case of zones 3 to 5, we saw entire portions of the faciès pile thinned, sometimes displaced, to the extent that locally we could not explain criticality except by invoking deformations (figure 5). But it always concerned *local* peculiarities, connected to successive detachments of beds, at the scale of meters. Here it is nearly throughout that the thicknesses of rich ore are insufficient.

We were then led to suppose that the behavior of the terrain had been different, that there had been modifications of the geometry that were much more significant, and that this could be explained by less overburden. It is this point that we would like to develop, returning to the observations from the geology.

## 3.2 A KEY: THE BEHAVIOR OF THE TERRAIN

Figures 11 through 14, taken from reference [19] show reconstructions that have been done by regrouping sometimes superposed fronts. We noted in chapter 1 that in zones 7 and 8 the faciès-pile is formed at the expense of sandstone from the upper

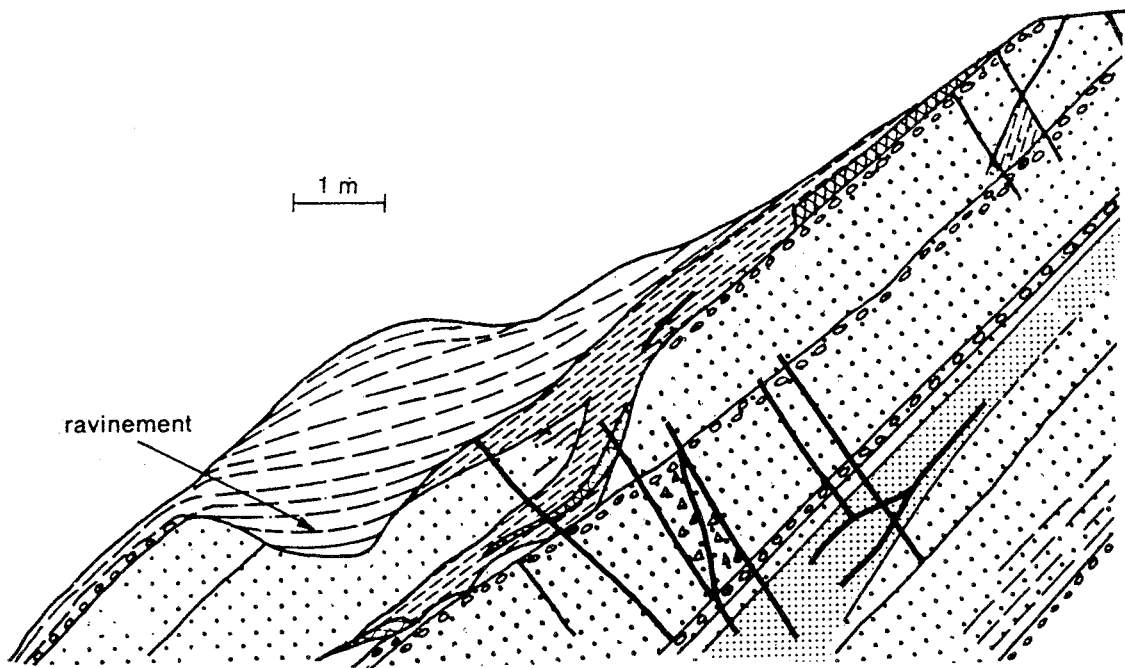


Figure 11. Front 299 A and front 1/4/78 (zone 7)

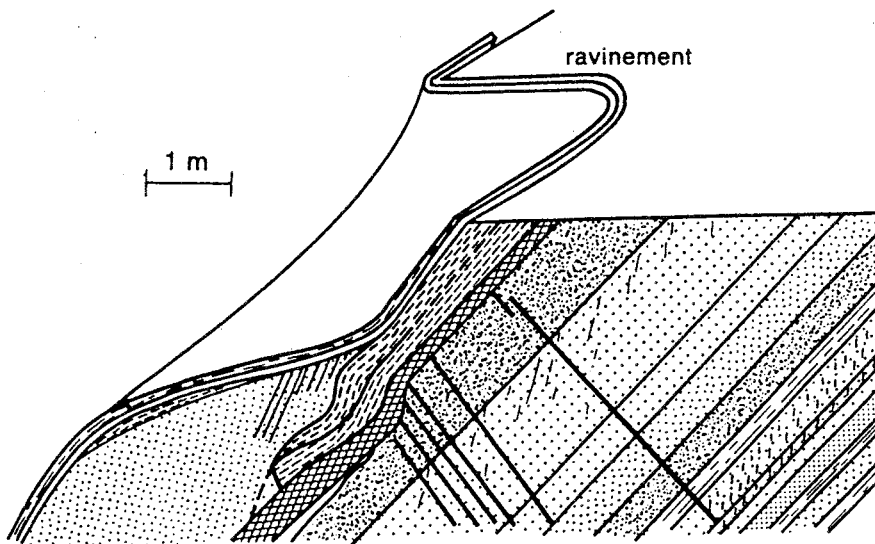


Figure 12. Front 299D (zone 8)

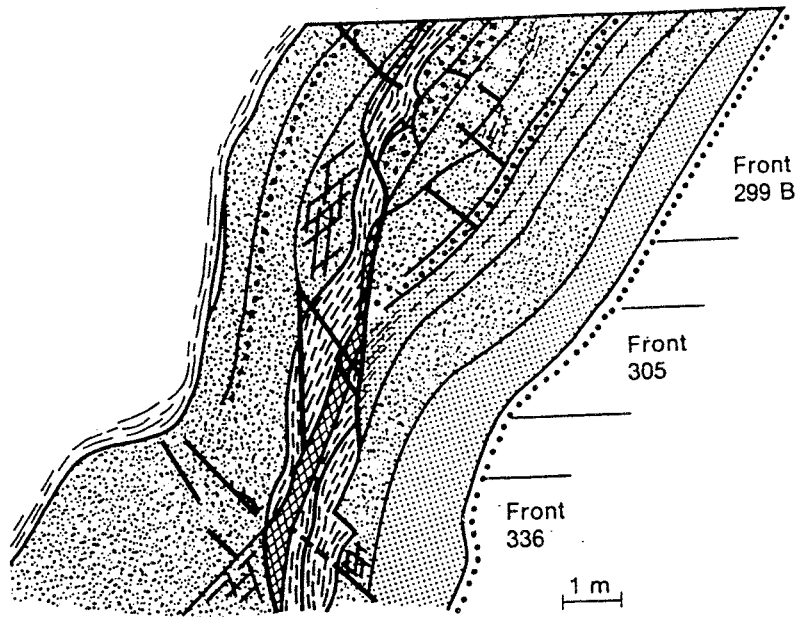


Figure 13. Composite of fronts 299B, 305, and 336 (zone 9).

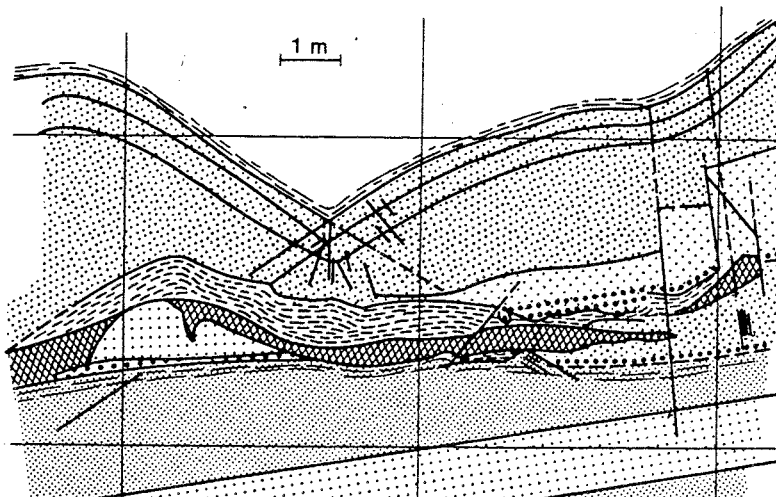


Figure 14. Outcrop GL 11 (zone 9).

part of the bed C<sub>1</sub>, while in zone 9 it is by contrast in the lower portion of C<sub>1</sub> that the reactors were formed. The observations are different in the two cases.

In the first (figures 11 and 12) we note that there are sandstones at the roof of the reactor that have remained in the lower part of the reactor zone, but that these "disappear" a little higher. This certainly is a disappearance because we do not find the argiles de pile that would have been formed by desilicification. These clays have undoubtedly been entrained in the large furrow that at the same time took a part of the pelites, and also probably a bit of the faciès-pile (causing the thinness of the remaining portion). These disturbances, which have moreover eroded the residual sandstones, have created in both zones 8 and 7, the characteristic "troughs".

We saw concerning zone 2 that when the argiles de pile came in contact with the pelites, the ensemble glided downward. But because of the fact that the pressure of the overlying terrain was very strong and, thus, that there could not have been at any time large open surfaces, the clays found themselves each time blocked against sandstones that were not yet desilicified. We never saw in sections 1 to 6 the clays "disappear" by passing over residual sandstones. Even if we think that part of this disturbance was caused at the time of mining, this proves that the compaction was not very strong.

But the situation observed in zone 9 is still more surprising. In this case there are always sandstones remaining between the reactors and the pelites, but the deformations are no less spectacular. In figure 13, three fronts have been superposed in a way that the distance from the top to the bottom of the figure is about 10 meters. We see that an enormous rockfall has profoundly deformed the terrain *over all that distance*. This collapse sank into a corner, as shown in figure 14 (the outcrop GL11 is a cutting at the roof of front 305). The ensemble of reactor/argiles de piles was crushed and probably laminated, flattened like a crepe. The most remarkable thing is that the fine wall sandstones have themselves been damaged. A part of them has disappeared, probably entrained lower down, and the argiles de pile, folded to vertical came to be plated onto the remains. It must be emphasized that in none of the twenty or so fronts in the northern sector have we seen significant deformations of the fine sandstones, which were always strongly held against the wall conglomerate by the pressure of the overlying terrain.

The size of these disturbances can only be explained, in our opinion, if we assume an overburden pressure sufficiently weak that the desilicified sandstones remained "open" (that is to say insufficiently compacted) rather long, over relatively large surfaces, provoking through this veritable rockfalls on a large scale. On the contrary, when the pressure is strong, even if the breaks are discontinuous, the overlying terrain is constantly pressing very strongly, prohibiting any significant displacements.

This behavior is completely analogous to what we saw in the mine workings conducted at several hundreds of meters depth, when the temporary supports are withdrawn (striking down). In addition, and perhaps as a consequence, the compaction seems to have never been very strong, giving an insufficient cohesion and new slidings at the time of exploitation of the quarry.

Finally, in the Oklo reactors, the terrain had *three types of behavior*, causing three distinct modes of functioning. In zones 1, 2, and 6, the pelites would have exerted immediately a very strong pressure, coming to mold themselves onto the desilicified sandstones and stabilizing the reactors very quickly. In the zones 3 to 5, the compaction would have been more discontinuous because of the rigidity of the bridging blocks, but these being also subjected to a strong pressure, could not resist long, and came continuously to be plated onto the reactors. Finally in zones 7 to 9, we had a discontinuous compaction, but much less strong because of the small thickness of overburden, from which resulted subsequent movements of much greater amplitude.

### 3.3 CHRONOLOGY OF THE EVENTS

There are now a converging collection of reasons that suggest that the reactors in zones 7 to 9 functioned under a smaller overburden. Since we know that the nuclear phenomenon was produced during the course of an uplift of the terrains, this would say that this sector functioned later. This hypothesis evidently poses a series of problems that it is advisable to examine.

First, it is clear that if we speak of a separation in time, if only because of the criticality, it can only have been a few million years (perhaps one or two tens of millions at the maximum), in any case an interval insignificant compared to the age of the reactors. The first question to ask is then whether the uplift of the terrains could continue *at that time* as far as the level assumed.

According to what was found for the pressure, two or three kilometers of overburden disappeared between the time of functioning for reactors 1 to 6 and the present day. When? The geologists have identified at the beginning of the Tertiary a rejuvenation of relief, followed of course by erosion, a consequence of distant events. But it seems that the consequences were rather limited. Through time, similar circumstances could have occurred, progressively eliminating the sediments in question. We know, however, that once the structural adjustments of the Francevillian were finished, the geologic history of this region became very calm. It is then plausible to go back to that very ancient time, which had significant events, for the essential part of the disappearance of the overburden ("on the heels" of what we already know).

It is necessary, however, that this hypothesis be compatible with the excellent geochemical preservation of the uranium deposit and more particularly of the reactors, which implies a certain burial. The study of lead, notably, has shown that some alterations were produced recently, probably following the already mentioned Tertiary episode, which brought the ore deposit closer to the surface and reactivated the old fractures. These alterations remained moderate, nonetheless, until quite recently the Okelo Néné dug its bed by the implacement of the quarry, placing the reactors in a very vulnerable position. These observations lead us to think that the

few hundreds of meters that lasted until the Tertiary were able to be enough to protect the reactors from superficial circulations.

Finally, it seems acceptable to assume that at the end of the "major tectonic phase" of the Francevillian and the peneplanation that followed it, the overburden was not more than on the order of four to five hundred meters for example (we recall that between the two sectors of reactors there is only about 50 meters of difference in strata level, which can be neglected in this discussion). Consequently, if the reactors of sector 7-9 functioned at the end of that period, the pressure could have been less than 40 to 50 bars.

If we hold to this hypothesis, we must explain why the reactors did not function at the same time as the others and why they began at that time. Initially we can propose two types of explanation: either all the uranium was already in place at the time of functioning of zones 1 to 6, but the fractures were insufficient and it was at the occasion of a new tectonic episode that ignition took place, or the rich sandstones formed later, or at least a certain amount of additional uranium was emplaced in a later phase, and it is only at that time that criticality was reached.

The first explanation could seem to be the most natural, because for zones 1 to 6 we have favored the idea that it is essentially fracturing in the already-existing ore deposit that led to criticality. However, the tectonic faults that are observed in sector 7-9 are analogous to the "structures" of sector 1-6 and probably contemporaneous. We have no reason to think that they have not played the same role at that time (moreover, the entry of water here is less necessary, because the ore was contained in organic matter still rich in hydrogen). One does not have, in addition, evidence for a late tectonic event. Moreover, this first explanation ignores the peculiarities of the rich sandstones of this sector, which could have played an important role. It is necessary, therefore, to examine the second hypothesis.

The rich sandstones remaining intact are abundant, notably around zone 5, and even more in sector 7-9 (where the reactors effected only a small part of a more vast ensemble of ore), and we can then compare in the two cases the primitive state of the reactors. We know that in the "normal" ore from Oklo (but also in all the ore deposits or uraniumiferous sites in the Francevillian), the uranium is associated with organic matter. We find equally in nearly all the deposits, in general connected with shear zones favoring circulations, a richer "oxide" ore, where the uranium is precipitated in microfractures without association with the organic matter. The rich sandstones of sector 1-6 are the clearest example of this. This having been remembered, the origin of the rich sandstones of sector 7-9 was the close overlapping in the same ore of these two types of mineralization, and also the exceptionally elevated quantities of uranium associated with organic matter.

Recent geologic studies have advanced the understanding of the origin of the mineralization. It was shown that all the uraniumiferous sites in the Francevillian had as their origin concentrations of hydrocarbons, for the most part migrated in the FA from the ampelitic levels that formed the source rocks. The mechanism of trapping



of uranium by the hydrocarbons seems now well understood, notably because of the studies done at the University of Orleans (ref. 60, 61). The uranium came to be fixed in the form of complexes of valence 6 by the free radicals formed in the process of maturation, and later it was changed to valence 4 causing simultaneously oxidation of the organic matter (cf chapter 4, §2.4). On the other hand, there remains much that is not well understood about the phenomenon of enriching concentration through oxidizing layers even if the principles and the modes are clear. Questions remain under discussion as much for the chronology (did it consist of a later stage or was it quasi-contemporaneous?) as for the size of the remobilization and the mechanisms of emplacement of the mineralization (respective roles of pyrite and organic matter, etc.).

It is therefore hard to arrive at a completely justified conclusion concerning the differences observed in the two sectors. Must we envisage in zones 1-6, which forms a structural trap in the top of the deposit, a gas pocket that would have preferentially collected the reducing gases from the hydrocarbon ores, causing the scarcity of residual bitumen? Must we think the absence of organic matter in this sector is caused by its complete destruction by the oxidizing layers, which were by contrast allowed to often remain in sector 7-9? Or rather still, did the maintenance of organic matter in the sector cause a new later coming of oxidizing fronts?

Lacking certain answers to these questions, we cannot exclude that the rich sandstones were formed at the same time under comparable conditions and, therefore, that all the uranium for zones 7-9 was there. We give, however, our preference to the idea that *at the moment of initiation of the reactors 1-6, the rich sandstones of sector 7-9 were not yet in the condition where we now find them*. This seems to us to better account for observed differences concerning the sandstones, and above all it has the advantage of being able to supply a plausible explanation for the late start of the reactors in this sector.

This does not signify, of course, that the rich sandstones were completely formed at a more recent date, constituting a better preserved model of what had happened earlier. A series of observations argues for a different analysis. Taking into consideration the relatively low oxygen content (O/C ~0.14), the proportion of uranium separated from the organic matter present is probably small, and we cannot undoubtedly justify by this mechanism all the uranium present outside the organic matter. On the other hand, there is a lot of hematite and we would be astonished if the iron had been oxidized in the presence of active organic matter. It is more plausible that the hematites were formed later. Moreover, it seems that the distribution of ore in black zones loaded with uranium and ferrous iron and red zones poor in uranium and rich in hematite, is not correlated with the distribution of organic matter (samples 1950/1 and 1950/2). Finally, with the exception of the organic matter, the rich sandstones resemble very much what one sees in the rest of the ore deposit. It is then more satisfying to think that they were formed at the same time and under the same conditions as those of the norther sector (with however lower uranium levels), but that they have been invaded *later* by a new arrival of organic matter, which provides a site for trapping of supplementary uranium.

The study of organic matter in the Francevillian has shown their diversity and it is natural to think that their migrations were staged in time. As we indicated in chapter 4 (§2.4), the French Petroleum institute (ref. 58) believed they recognized significant differences between the organic matter of zones 7-9 and those of the environment of zones 1-6 (levels of oxygen, behavior during pyrolysis), suggesting that they had perhaps experienced a less severe thermal history. The levels of uranium of this organic matter is also very exceptional (exceeding sometimes 30%) and the proportion of uranium not unmixed is unusually elevated. (It is moreover essentially by using the organic matter from sector 7-9 that the group at Orleans was able to show the correlation between their structural organization and the degree of demixing). The hypothesis of an origin a little later of the organic matter is then plausible.

We emphasized that the starting of the reactions in sector 1-6 was situated in an important tectonic phase that caused a large amount of fracturing to occur. It is strongly possible that new movements of hydrocarbons took place on that occasion, if the source rocks were still active. It could then have been at this time that the organic matter in sector 7-9 was implaced, then under a still considerable overburden, but nonetheless after a significant uplift. The "oxidized" rich sandstones were already in place, but with levels of uranium insufficient for chain reaction and that arrival could not by itself furnish immediately the necessary addition.

In effect, we have seen that it is in the course of maturation of the organic matter that free radicals are created that can trap uranium from solution, and so there was needed a certain time for these conditions to occur. But, in addition, there was needed, of course, inward transport of uranium. We said above that at the moment when the reactions of sector 1-6 were produced, the circulations that had been the source of the superconcentration were perhaps no longer very active, or even that they had already ceased. All the more so, one can imagine that was the case after the maturation of the organic matter. In this hypothesis, a new circumstance was needed so that transport of uranium occurred. One can justifiably propose a completely logical explanation.

When the ore deposit at Oklo was discovered, the mineralized bed reached the outcrop, with naturally an ore strongly oxidized and leached in the neighborhood of the surface. One saw that several tens of centimeters of overburden had been eroded during the Tertiary period and it is not forbidden to think that the ore deposit extended beyond that. In other words, when, a little more than 1950 million years ago, the uplift of the terrain had been sufficient, one can consider that the ore deposit appeared at the surface and, in this case, the last few tens of meters must have been systematically leached by superficial fluids. The uranium in solution, descending through the deposit, was then susceptible to be retrapped by organic matter, if this organic matter was still fresh and at the optimum point of maturation. This explanation justifies the condition that the critical condition was only realized at the arrival near the surface, thanks to the trapping of new uranium. It also allows us to

understand the exceptional levels of non-unmixed uranium, because this had never known high temperatures.

### 3.4 ORIGINAL ASPECTS OF FUNCTIONING

In this hypothesis, the functioning of reactors in sector 7-9 then was rather different in several respects. In the first place, it was an inward transport of supplementary uranium in a medium already stabilized, and not new fracturing that provoked the chain reactions. This manner of looking at things agrees rather well with the observations. We noted in effect (figure 18 of chapter 1) that these zones consisted of a string of very small scraps separated from each other, contrasting with the continuous extent of four large groups in sector 1-6. While we envisaged for the latter a systematic propagation from a small number of ignition points, there would have been many more here, which is in agreement with the arrival of uranium being emplaced a little bit everywhere. Entry of water was not, on the contrary, as necessary, because hydrogen was already largely present in the heart of the reactors (thanks to the organic matter not yet degraded), and that the levels of uranium were not so high. The starting of the reactions could then occur without new tectonic actions. As a corollary, the phenomena of propagation would not have played such an important role as a result of the limited nature and the slowness of desilicification.

As we already noted, the control of the reactions was undoubtedly rather different and much more complicated than for the other zones. The hydrogen initially present must have been rather rapidly eliminated by degradation or mobilization of organic matter (of which a part has migrated and is found outside the reactors). It is possible, despite this, that uranium continued to be emplaced after the start of the reactions. (We found an isotopic depletion in the organic matter in the core that was a little less than in the neighboring argillites). The departure of hydrogen was compensated for by the entry of water accompanying desilicification, but as this was done slowly (in this range of temperatures) compactions, even very imperfect, had enough time to occur. There was not, of course, too much water, considering the mediocre neutron balance, and that explains why for most of the time the temperatures did not rise very much. In addition, even when there was a lot of water, it was perhaps necessary to call upon another peculiarity of this sector. The levels of uranium are in general very low (the maximum often being less than 25%), meaning that the environment could become in certain cases *overmoderated*.

In a condition of overmoderation, the control of the reactors is opposite to that which was analysed in chapter 9. When the porosity increases, not only are the temperatures not induced to rise, but the reactor can be extinguished by an excess of water. Conversely, it can restart when the terrain subsides more. One can reach in this situation the effect of coupling. In effect, let us suppose that at a certain moment the environment was near the optimum, but still undermoderated. If the volume available to water increased, normally the temperature increased and the density of water decreased to maintain the amount of water necessary for criticality. However, as we have shown, the increase in temperature could be frustrated by the environment (if the disequilibrium is only local). The amount of water could then

increase beyond the optimum. If at that moment the environment reacted in phase, the balance found equilibrium again, but on the other side. The control was then "to the opposite". This situation could not have existed everywhere at the same time, and it was not produced when the ore was very rich (the medium remaining always undermoderated). But this condition could contribute in this context to holding the temperatures down.

Of course, when there was at the same time relatively rich ore and a more pronounced delay in compaction, we find ourselves in the conditions analysed for zones 3 to 5, which is with the need to lower the water density, which given the pressure translates into local boiling. We understand then that the very low average temperatures (not only in absolute value but also in  $\Delta T$ ) are not incompatible with vaporization on the scale envisaged. This only occurred in a significant manner in a small number of cases.

While the reactions and associated desilicification occurred a little everywhere, because the compaction was never very great, there came a time when the equilibrium of the terrain was broken and there was a grand scale rockfall. This could moreover be favored by the condensation of vapor to the extent that it could cause a very localized desilicification. The collapse caused an unrecoverable stop to the reactions, giving a very low average level of reaction and a final situation in which criticality could not be justified.

Finally, we saw that, even if many problems exist for zones 7 to 9, our hypothesis allows explanation of nearly all the peculiarities of this sector. These were absolutely significant because they resulted from a situation profoundly different from that of the northern sector.

## CONCLUSIONS

In this work we have presented a study of the reactors of Oklo, essentially founded on the physics and more particularly on the neutronics, but also in which we have equally integrated all the information that has been acquired on the subject. Our ambition was to reach a better understanding of the phenomenon. In each of the chapters, we have explored in depth one aspect of the problem and analysed its many sides, regrouping the observations, measurement results, and interpretive calculations. It is, then, not a question of summarizing this work here; we could only give a poor picture of the phenomenon, while we have searched, on the contrary, to bring out all the complexity.

A peculiarity of this study has been to call upon the very elaborate calculation methods of reactors. That we were able to use the tools of calculation developed for industrial reactors to analyze events that took place in a uranium deposit 2 billion years ago is in itself extremely remarkable. It is this aspect of the study that we have chosen to comment on in this conclusion. Moreover, to emphasize that this approach has been fruitful, will give us the chance to recall a number of important results. We will end with some remarks on what has been of originality in this research.

### THE VERY DETAILED INTERPRETATIONS

If it has been possible to employ usefully the calculations for reactions, it is evidently because we encountered exceptionally favorable conditions. Not only the state of preservation of the reactors, which kept a prodigious amount of information written in the terrain, but also the peculiar conditions in which the reactions "operated", which allowed that the information was intelligible in terms of neutron physics. It is this second point that it is interesting to examine more closely. Indeed, it was not achieved at the beginning of the studies, and we realized later that it was not achieved everywhere.

It is very fortunate that the circumstances of discovery led us to concentrate the initial analytical studies on a small portion of zone 2, because it proved to be that this was the only place where there was at the same time an excellent representativeness of the measurements and a functioning sufficiently simple. (We will explain later what we mean by that). It is probable that if we had begun with one of the zones 5 to 9 for example, this work would not have been done. This is because to the extent that during the advancement of the studies the understandability of the results was confirmed, we were encouraged to multiply the isotopic analyses, perfect the calculational tools, and push further the interpretations.

It is advisable to emphasise that the calculational codes used in this study do not boil down to approximate formulas, but are rather "sophisticated". They have certainly been adapted to conditions particular to the problem at hand with certain simplifications, but we verified that they did not cause any loss of precision as compared to ways that are known to better treat this kind of neutron multiplier. As we explained in the introduction to chapter 5, despite all the information we lack concerning the conditions of functioning of the reactors at the start, this concern for exactness is a necessity. To maintain a chain reaction very precise constraints must apply, and it is only to the extent where we take into account these constraints that we can correctly exploit the experimental results, and in particular to adjust without ambiguity the unknown parameter values through the results of the analysis.

A good example in this respect is the calculation of the amounts of water present at the time of the reactions, of which the knowledge is fundamentally important. The ratio of water to uranium can be calculated from the "spectral index" (itself deduced from the isotopic analyses of two fission products: neodymium and samarium), but the calculation requires that we know how to link up in a rigorous manner that index to the structure of the neutron balance, and that we evaluate numerous elements of that balance (for example the plutonium that was present in transitory state). Good precision requires, on the other hand, that we treat simultaneously the ensemble of the distribution because of the importance of environmental effects on the spectrum, and it is necessary to equally take into account the history of the fission products, because it is these that are used. Moreover, it was necessary to do intercalibrations of separated isotopes in the reactor Triton and to interpret them using the same structure for the calculations.

In other words, to integrate correctly the experimental results into the reactor calculations, it is truly necessary to go into the details and use considerable rigor. One cannot avoid, however, making a certain number of hypotheses, notably on the evolution of the different quantities through time. Fortunately there are possibilities for cross-checking that allow verification of the validity of these hypotheses. Taking the same example, water plays a determining role in all the questions of criticality. Water controls the value of most of the neutronic parameters of which a certain number are accessible to measurement, and in addition the distribution of the level of reaction in the terrain depends very closely on the distribution of water. There is, then, a whole group of information of different kinds that we can combine. We constructed a framework, very fragile at the start, but which was reinforced as we incorporated new results, at least as it revealed itself to be coherent, which was the case in zone 2.

We know that we concentrated in a well-chosen drillhole in that zone, the SC36, a considerable number of measurements (some 215 isotopic analyses over a distance of 1.50 meters!). This abundance of results let us make very detailed interpretations, working out little contradictions in the measurements, and calculating numerous interesting results. We even went so far as to do a simulation of the operation of that portion of the reactor in space and time. The very remarkable coherence that we found in that attempt showed without possible doubt the applicability of the

calculations and at the same time confirmed the assumed conditions for functioning. This is the only case where we could go that far, but we were able to show that we could generalize the results to the whole of the sector. In particular, it was possible to explain the form of the distribution of the levels of reaction in other drill holes. It was equally satisfying to be able to justify with precision criticality in the final condition of the reactors in all that zone using parameters independently selected. It is all these interconnections that allowed us to think that the calculations rested on a solid foundation and that we could have confidence in the conclusions drawn from them.

### **A BETTER APPRECIATION OF THE GEOCHEMICAL REMOBILIZATIONS**

Our study allowed us to exploit to the maximum extent the measurements, but it led at the same time to a more objective appreciation of the representativeness and then to the geochemical remobilizations that were of a kind that compromised the measurements.

The isotopic chemistry could show good correlations between the indications from several elements, or on the contrary, show incoherences, but it could not lead to perfectly certain conclusions concerning their representivity, because of the lack of an indisputable reference point. It is because of this that for a long time we overestimated the stability of neodymium, which served as a reference for all the other fission products. Also, there has always been questioning about the integrity of conservation of uranium in the heart of the reactors, which is fundamental since it is this to which everything is related. We have shown that the constraints of the neutron calculations permit, in certain cases, to lift the ambiguities and even arrive at near-certain results.

Concerning uranium, that there are situations where there had been in the reactor core essentially no transport in or out or significant movement within the reactor after the start of the reactions. More particularly, the isotopic depletion of uranium has appeared in nearly all cases as the most certain information concerning the levels of reaction. But we have also shown that the behavior of this element was not the same everywhere, and that there were cases of significant mobilization. In addition, we explained the reason for that diversity of behavior, showing that it was tied to the timing of desilicification and nuclear reactions. The situations where uranium was stable throughout the reactions are explained by an early argillization that sufficiently blocked the circulations in a medium that remained reducing. In addition, the physics study confirmed that the decay of plutonium to <sup>235</sup>-uranium occurred entirely within the uranium that had formed it.

We have shown on the other hand that the remobilizations of the rare earth elements were more significant than we believed at the start. They were not limited to perturbations at the borders, but had a very systematic character. This has not always removed their representative meaning because it consisted only of a spreading over short distance, that we have been able in one case to reconstitute in detail. The neutron fluence deduced from the isotopic analyses of neodymium are often rather

seriously imprecise, but the constraints put by the neutronics on the calculation of the conversion factors allows us by using the results from the uranium to reestablish the correct fluence curves. In the case of SC36, the differences are limited by the fact that the mineralization is very homogeneous, but they are not completely negligible. It is then remarkable that the analyses of thorium came to be placed afterwards exactly on the predicted curve. Concerning that element, it appears that thorium could have given very reliable results, although the divergences encountered with neodymium caused it to be suspect.

This better appreciation of the measurements and their integration into interpretive calculations allowed us to determine certain characteristics in a more correct or more precise way. This is the case, as we have just said, for the fluences and the factors of restitution for plutonium (recall that we have found in the cutting 2'P' a record fluence close to 2.5 n/kb, although no direct measurement has given values higher than 1.65 n/kb). The possibility to calculate exactly the level of rapid fission reactions when we know the ratio of water to uranium allowed us to draw from the isotopic analyses of ruthenium a more precise estimate of the duration of the reactions in the environment of SC36 (~850,000 years in "equivalent" conditions). It is also the combined interpretation of all the measurements carried out on that drillhole that allowed us to reduce considerably the margin of error on the dates of the reactions that had been for a long time the subject of controversy ( $1950 \pm 30$  my).

## **TO UNDERSTAND THE FUNCTIONING OF THE REACTORS**

Our study allowed us to understand how that little portion of zone 2, where most of the measurements were concentrated, functioned, and at the same time explain why the calculations of the reactor had been able to be used in good conditions.

Recall that a very remarkable result concerned the distribution of water. We found that the porosity of the terrain at the time not only was enormous (nearly 40%), but that it had been nearly uniform in all the reactor zone, and that additionally it had evolved little in the course of the reactions. Such an unusual value for the porosity clearly could only be explained in the light of the proposition of the geologists (who received thus a supplementary confirmation), that the quartz from the original sandstones had been totally dissolved by hot circulating fluids. It is then in the completely destroyed and imperfectly compacted terrains that the reactions took place. In particular, there no longer remains any trace of the initial fractures in the sandstones, which would have given a place to privileged circulations and that we would have been able to detect by the distribution of reactions. The results suggest, in addition, that we attained at the beginning of the reactions a situation of pseudoequilibrium in the compaction of desilicified terrains that, although they had not been completely finished, only evolved very slowly.

This precociousness was confirmed and elaborated on by another important result concerning the average distribution of silica during the reactions. The calculation of the conversion factors is indeed sensitive to the amount of this diluting element. It



shows that quartz was nearly totally absent from the core, although there were increasing amounts of it as one went to the exterior. It is, then, that the desilicification was practically finished in the core at the start of the reactions, but that it continued in the borders. This chronology is confirmed by the results of the study of thermal convection that showed that desilicification only proceeded in the borders when there was an impermeable "cork" at the site of the heat source. It is this circumstance that explains that the core was protected from fluid circulations, while one can see displacements of depleted uranium in the borders.

It remains, nevertheless, to explain why the reactions were installed each time in the terrains profoundly deformed but already stabilized, while the desilicification is itself a consequence of the nuclear reactions. We proposed a process to explain that situation, which consisted of assuming that the zone of reaction, at first limited to its upper part, extended progressively toward the base. In these conditions, immediately underneath the reactors, the water set in motion by convection was heated on rising and charged itself with silica to bypass the portions already turned to clay. The compaction of the terrains in the course of desilicification was done essentially without delay, because the overlying adjacent sandstones already detached on the uphill side exerted a significant pressure on that part of the terrain by the means of the pelites, a plastic medium, which came to settle by gliding downhill. On the other hand the reactions were temporarily retarded in their progression by the combined effects of neutron poisons and temperature, so that the medium in question was already practically stabilized when the neutron flux became significant there. This scenario conforms to the observations of the terrains (sliding of pelites), and we verified the compatibility of the kinetics that it implies.

Obviously one could only suspect at the start of the studies the appearance of perfect stability, which made one believe that the "fossil" reactors were in their original state, was in reality the result of rather considerable geometric upheavals. Furthermore, the stability of the reactor medium had not been as perfect as we had believed. The medium had still evolved at the start, and it is not excluded that the sliding and lateral deformations continued during the reactions (with happily little consequences on the interpretation of the measurements). It must be emphasized that the regularity of the progression, which gave rise to these very favorable conditions, was only possible through a combination of very favorable circumstances (starting at the top of the zone, sufficient dip but not too great, homogeneous mineralization, optimal thickness of the pelite bed, adjustment of the kinetics, ...) that were only fully united in the central part of that zone, which happened to be at the outcrop surface at the moment of the discovery of the phenomenon and that, therefore, was given priority in the studies. It is that luck that we mentioned just above.

Because of that relative geometric and geochemical stability, the control of the nuclear reactions in this zone was rather simple. The two principle mechanisms were on the one hand the destruction of the neutron poisons (boron, samarium, gadolinium) that was faster than the destruction of the fissile nuclei, and on the other hand the rise in temperature that, principally through the change in the density of water, had an unfavorable effect on the neutron balance. In these conditions, the

destruction of the poisons began by liberating reactivity, which caused the temperature to rise. This passed through a maximum and then decreased, the poisons in large part destroyed, the decrease in fissile nuclei became preponderant. Finally the reactions stopped when the temperature had returned to the ambient level and there had been no reserve of reactivity. In practice, things were obviously a little more complicated, but we have good reason to think that this scenario that is amenable to calculation (what we have called the "reference model") was largely predominant in that part of zone 2. Because of the enormous thermal inertia in the terrain, and since their compaction barely evolved further, all the evolution was slow and smooth. In this model, the maximum temperature reached during functioning is rather closely correlated with the reserve of reactivity and thus the final level of irradiation.

An important peculiarity of the Oklo reactors is their shape as a very flattened "cake", whose thickness is relatively small as compared with the migration length for neutrons while the other dimensions are large. As a result, the coupling of the neutronics and thermal effects are very tight in the small dimension where everything functioned in phase, while it is very loose in the others, where it could cause stepwise functioning. In addition, the sedimentary origin of the ore deposit meant that the gradients in the uranium levels (especially after compaction) are much larger in the direction perpendicular to the layering than laterally. These are the two properties that justify the fundamental nature of our approach, consisting of studying systematically the distribution of levels of reaction in the small dimension. This is indeed by far the most significant. It depends relatively little on the environment that, in general, is not well known. (And also to the lateral sliding of the terrain, but we did not know of this until later.)

But this character of tight coupling in the small dimension had another consequence here, which is apparent when we made the simulation of operation in the framework of the "reference model". We noted that, except at the beginning, the flux curve changes rather little with time (which means that the exchange of neutrons in that direction remains largely superior to the differences in evolution of the local balance). The result from this is that we get a rather good approximation when we study the evolution in time using the averages in space, or when we study the distribution in space using the averages over time. It is this that explains, although we used only the results of measurements resulting from quantities integrated over the duration of functioning, that we could (by making only small corrections drawn from the model) profitably use the global neutron calculations to interpret the measurements. This is another aspect of the "simplicity" to which we alluded. It must be noted that this is only true if the distribution of uranium is sufficiently compact (which was the case in SC36). When it is more spread out or more irregular, and especially if there is a rich secondary layer, we have shown that the coupling in the small dimension is already more complicated. We see that we have benefitted from a whole ensemble of favorable circumstances.

## DIVERSITY OF CONDITIONS FOR FUNCTIONING

If zone 1 and the upper part of zone 6 have "functioned" in a manner comparable to zone 2 (with however, peculiarities), it was not at all like that for the other zones (3 to 5 and 7 to 9). It is equally a contribution of our study to have described this diversity and to have explained it. We pointed out, notably, that it is not possible to explain criticality or the distribution of water under the same conditions as for zone 2. We were led to think that the reactions took place in a medium not yet stabilized and that the compaction of the desilicified terrain was accomplished in an irregular manner.

The characteristic of these zones is essentially that the fractures that served to drain the circulations in the phase of super-concentration of uranium here affected only part of the mineralized bed. The rich zone is, therefore, more narrow, and intact  $C_1$  sandstones remain from place to place in the reactor. These rigid bridges transmitted the pressure of the overlying terrain during compaction of the desilicified portions in a less immediate manner than the pelites. The geologic situation shows that they became detached as little blocs (on the scale of meters), provoking discontinuities in the evolution of the porosity.

We showed that in these conditions, the reactions could propagate in the incompletely desilicified terrains, in an irregular manner and more or less in all directions. Because conversion to clay was not reached at the start of the reactions, the geochemical remobilizations and in particular the displacement of uranium has been more significant. On the other hand, the control of the reactions has been much less simple, notably because the volume left for water could vary greatly and randomly. When there was a delay in the rupture of the bridge-blocks, the porosity became larger and that would have caused the temperatures to rise. Conversely, a "catching up" of the compaction translated into a prolonged lowering of the temperature or even a premature stop of the reactions. We simulated through calculations these different possibilities in order to bound the plausible regions. In this case there is no longer a correlation between the maximum temperature and the level of final radiation. That is to say, the "simple" conditions encountered in zone 2 were far from being realized everywhere.

The differences are even more important for zones 7 to 9. These are characterized, moreover, by a whole collection of peculiarities: abundance of organic matter, indications that the temperatures remained low, different conditions of desilicification, tectonic readjustments on a much larger scale. We showed that it is impossible to justify criticality in the major part of these zones, even taking into account supplementary parameters tied to the behavior of the sandy bridge-blocks. It is necessary to admit that these reactors are not in the geometry in which they functioned. As we will say later, we were led to assume that the reactions took place under a clearly smaller amount of overburden. The pressure exerted undoubtedly was not enough to compact the terrains to the extent that desilicification proceeded, and this is why there had been, in a postponed manner, collapses on a large scale.

Finally, it is the manner in which compaction of the terrain was accomplished following the departure of silica that appeared as the most determining factor. Following this criterion, there have been three distinct modes of functioning of the reactors.

## TEMPERATURES AND PRESSURES

The physics study carried some important elements of appreciation concerning a controversial question, that of temperatures and pressures. We knew from the beginning, from the geology, that the reactions - at least for the sector of zones 1 to 6 - had taken place under a cover of some kilometers, so with a pressure of several hundreds of bars and an ambient temperature in the interval 100 to 200°C. But the indications drawn from the study of minerals were rather vague. Moreover, they could not give us information on the temperatures reached during the course of the phenomenon, because the minerals in the core of the reactors had been destroyed by radiation damage and have later recrystallized. The most important results have been furnished by study of relict fluid trapped in microscopic inclusions in quartz. But these can only give a relation between temperature and pressure, the interpretation of which is very delicate (above all if the trapping was done in the region of boiling) and in addition, quartz has been destroyed or altered in the immediate environment of the reactors. The results published in 1977 from the colloquium in Paris (pressure greater than 800 bars, temperatures in the course of the reactions of 450 to 600°C) have been subsequently challenged and one has gone as far as to deny any representativity of the fluid inclusions in the vicinity of the reactors. On the other hand, the isotopic analyses, notably that of lutetium, did not give precise results because of remobilization of that element and the lack of knowledge concerning effective cross-sections.

On the other hand, physics imposes unavoidable constraints on the temperatures. In the first place, it is clearly necessary that criticality be maintained. Because it is temperature that has controlled the reactions, it was necessary to have a sufficient margin of control, but also that it remain compatible at all times with the necessary reactivity. In the second place, in a chain reaction, the power is not determined from the outset; it adapts itself so that the temperatures have the level desired taking into consideration the conditions for removal of heat. We have indications concerning the neutron flux from the measures of the level of fission in plutonium and, thus, on the densities of power. These were very weak (a million times smaller than in industrial reactors!), which implies limitations on the elevation of temperature. The indications concerning pressure (which conditions the density of water) are only indirect, but there are, there also, constraints by reason of compatibility between the different results. Our study led, finally, to values of temperatures and pressures considerably more moderate than those that had been put forward in 1977, but which are compatible with the more recent geological studies.

We have shown that, as far as zones 1 to 6 are concerned, the temperatures have at certain moments (but only in the most rich part of zone 2 and some portions of zones 3 to 5) been able to reach the range 360-400°C (for an ambient temperature of the

order of 140 to 150°C) but have not probably passed these values. The average differences in temperature with the environment (mean in space and time) have been of the order of 100° for a large part of zone 2, but hardly passed 50° for the other zones.

In this sector, we showed that control was effected essentially in a single phase, which means a pressure of at least 150 bars, but we think that it was not above 300 bars, and there are arguments in favor of a lower pressure. This implies that conditions were at times in the vicinity of boiling and that, on the other hand, that the geothermal gradient was very high, which is plausible 2000 million years age, considering the decay of radioactive elements that supply the internal heat of the planet. One can note that our conclusions concerning the behavior of the terrain (very imperfect compaction at the time of stabilization in the case of encasement in pelites, and systematic delay for the breaking of bridge-blocks in the case of encasement in sandstones) argues in favor of a moderate pressure.

In the case of zones 7 to 9, there are a whole collection of facts that lead us to think that the overburden was considerably less at the time of the reactions. We propose a cover of the order of 400 to 500 meters (giving a pressure of 40 to 50 bars, and an ambient temperature of about 50 degrees). Under these conditions, the temperatures would have been limited by boiling at about 250 to 260°C. It seems clear that evaporation of water has effectively played an important role in that sector.

## **THE CURRENTS OF THERMAL CONVECTION**

Our study of thermal convection has allowed us to better define the characteristics and consequences of fluid movements induced in the terrain by elevation of the temperature. The currents played a minor role in the removal of heat, but a very important one in the area of geochemistry. We have specified in particular the conditions under which desilicification of the original sandstones was accomplished, and its effects on the progress of the reactions.

We verified that for plausible values of the permeability and furthermore for the temperatures proposed, we could effectively justify the total dissolution of quartz and its elimination during a time that was short relative to the duration of the reactions. We can equally justify the observations concerning the extent of desilicification in the border regions and the differences in this respect between the reaction zones. In the case of zones 7 to 9, a part of the effects are explained by the occurrence of boiling, the quartz being dissolved not only during heating of the water, but also during condensation of the vapor.

Another effect of thermal convection was the formation of large amounts of magnesian chlorites. We showed that most of the observations could be explained by a competition between the dissolution of quartz and the transformation of illite, taking into account the slowness of the inward transport of magnesium by the induced currents. The orders of magnitude of the circulation speeds before and after argillization also permitted us to justify the differences in behavior of uranium, as

well as the isotopic rehomogenizations observed, taking into account the nearly total destruction of the minerals in the core. There is definitely a good coherence among all the results.

On another topic, this study lets us understand why the Oklo reactors do not necessarily constitute a good "natural analog" for studies of the disposal of radioactive waste. Effectively, it was principally during the reactions themselves that the migrations took place. In that period, of which the duration is, nevertheless, rather long - on the order of a million years - the conditions were not to say the least those of a good confinement, and were not at all representative of a repository characterized by well defined parameters. The medium was completely torn apart; it lost 80 percent of its matter; the clay minerals were metamorphosed by radiation damage and transformed; the temperatures were relatively high and convection currents were important.

On this subject, the fact that we had enough information to do this study (essentially because of the retention of uranium in some privileged zones) should not delude us. Only a small number of elements, such as plutonium and radiogenic thorium, remained entirely in the uranium that was their source. Even if for some other elements that partially left (such as the rare earths and ruthenium) we can determine that they did not go far, for the most part the dispersion was much too large to allow us to study the modes of dispersion and do any kind of balance. These remarks do not exclude so much the interest in complementary studies on those fission products that remained in the environment of the reactors.

### **THE PROGRESS OF THE PHENOMENON TAKEN AS A WHOLE**

In the light of all of our results, we searched for a more global understanding of the progress of the phenomenon in its entirety, and the manner in which it fits into the regional geological history. This then, of course, consists more of speculations.

The start of the nuclear reactions is situated in an intense phase of tectonic activity on a great scale, and it is without doubt a consequence of that. The date of the reactions - 1950 million years - notably coincides with that of a period of metamorphism in the neighboring series of Ogooué, more to the west. F. Gauthier-Lafaye considers that there were events there that as an indirect consequence provoked a restructuring of the whole of the Francevillian. It was at this occasion in particular that the great fractures served as drains for the circulations of mineralizing fluids that formed by trapping the "superconcentrations" of uranium constituting the critical masses.

It is not sure, however, that it was the accumulation of uranium itself that was the determining factor in starting the reactions. We think it was rather - at least in the sector for zones 1 to 6 - the play of well identified transverse tectonic structures that, by opening, allowed sufficient water to enter into the already constituted uranium ores. It is possible that these two actions were more or less simultaneous, the ignition taking place "on the heels of" the emplacement of the uranium, but we cannot exclude a certain delay. In any case, the possible arrival of supplementary

uranium during the course of the reactions, notably by the effects of the induced convection currents, can only have been a minor phenomenon in our opinion.

The conditions necessary to start the chain reactions in the sandstones are very restrictive (very high level of uranium, significant entry of water) and they were undoubtedly only realized in some privileged locations. For zones 1 and 2 we think that the reactions started at the top, in the part that had already been mined at the time of the discovery of the phenomenon. As far as zones 3 to 6 are concerned, we identified three possible points of ignition, and it is not certain if there were others. It was essentially by propagation that the reactions were extended to the whole of these zones, and this is why they continue for distances that are very large compared to the migration distance for neutrons. We know that the "motor" was the desilicification of the sandstones that concentrated the uranium closer and closer, and we saw that the methods of propagation, and the compaction of the terrain that was a consequence of it, were not the same throughout; but they constituted in all cases a determining element in the control of the nuclear reactions.

Concerning zones 7 to 9, we have backed up the hypothesis that the reactions did not start at the same time as the others, but that they were a little later, at a time when the overburden had become noticeably less. We propose an explanation for this delay: the accumulations of uranium had not been sufficient at the time of starting of zones 1 to 6, but a new arrival of hydrocarbons allowed the later trapping of supplementary uranium. This idea rests notably on the fact that the rich sandstones of this sector contain much organic matter that has kept large amounts of uranium in the form of complexes. In this case, it is the emplacement of the supplementary uranium that started the reactions.

In this hypothesis one is led to think that the functioning of the reactors was rather different from those of zones 1 to 6, not only because of less overburden of which we have already recalled the effects (lower temperatures, more marked occurrence of boiling, less pressure exerted by the terrains lying above with rock falls of larger extent) but also from the fact of the different conditions of starting (uranium associated in part with the organic matter still active, less importance for the entry of water, equally less importance for the phenomena of propagation: the zones of reaction acting here like a series of laminae). Complimentary studies should, in our opinion, be conducted on this sector, notably exploiting the increased performance of the ion microscope. It would be interesting to examine in more detail the behavior of uranium and fission products in this particular environment.

We know that a whole series of other nuclear reactor zones have been identified. If we admit that the only two that have been studied functioned at different dates and under dissimilar conditions, we must obviously wonder what is the case with the others. Our hypothesis makes the second a sector of reactions in some way "failed" that were retrapped a little later, and we could claim that this was an exception. But we can also think the opposite, that it is the sector 1 to 6, because of having collected from the fact of its privileged position high in the ore deposit, an enormous mass of uranium during the episode of superconcentration, in a region moreover strongly

tectonized, should be distinguished by starting before the others. The exploitation of new zones and in particular that of the large sector of zone 10, which was begun in 1987 and should be continued for several years, could perhaps bring some answers to these questions. This sector appears particularly interesting, and it is desirable that one make here a minimum of investigations.

## **INTERMINGLING OF THE GEOLOGY AND THE PHYSICS**

The "natural nuclear reactors" reveal the same physics then as those we know. The laws have not changed over the two billion years [ref. 194] and the same methods of calculation apply. If there is any distinction between them, it is through the complicated history of the medium that they are part of, that one is obliged to take into consideration and which, itself, is within the realm of geology. One would have been able to note in this affair to what an extent the geology and the physics have been intermingled.

Of course, from the beginning we knew that the nuclear phenomenon was squeezed into a geologic episode, because it was necessary to concentrate the uranium and fracture the rocks to make water come in here, but we could hope that after that the reactions took place in a stable environment. The following should show that it was nothing like that and that the nuclear reactions themselves have contributed to completely modifying their environment. Still more astonishing, the desilicification of the original sandstones, a consequence of the reactions, permitted their extension, justifying in this way an idea so extraordinary that at the beginning we did not dare to formulate it: "the reactors made themselves gradually".

We could further be astonished that it took so long to identify that important upheaval. At that beginning we expected to find extraordinary effects, although the hydrothermal phenomena were ordinary. The environment of the reactors seemed "normal". It required minute observations to arrive at this conclusion and also much reflection and calculation to analyze all the consequences of it on the functioning of the reactors, and in particular to understand that the modes of compaction of the terrain at the end of desilicification had played a major role.

It is effectively the variation of the volume offered to water, a fundamental parameter for chain reaction, that for the most part conditioned the development of the reactors in space as well as time. The temperatures, whose level came at each moment to adjust in order to achieve compensation, also played a role by their distribution that could generate extension of the reactions, or favor desilicification. From this came the extreme complexity of the couplings that intervened to control and to propagate the reactions, and which arise at the same time from physics and earth sciences. If the calculations allowed us to clarify the mechanisms, only the numerous observations of the terrain could furnish all the keys.

During one of the first meetings that followed the discovery of the phenomenon, at a moment when we still had trouble believing in the reality of the chain reactions, and when we wondered how the devil the reactors had been able to be controlled, P.



Taranger, the director of Production at the time, had this word: "what if they had been controlled by tectonics?" Everyone looked at each other, taken aback, not daring to confess that they did not understand the meaning of that slightly impenetrable quip. He, himself, at that time could not have known how right he was.

## **AN ENRICHING COLLABORATION**

This interlayering of two disciplines that had not until this occasion been combined, which gave then all the originality to this research, has naturally required very close relations and constant discussions between reactor physicists and geologists, and this unusual collaboration merits perhaps some comments.

It is useful first to emphasize not only the quality of the work done by the different groups but also the friendly climate in which the discussions were held, especially sustained by the geologists in Strasbourg, a climate that allowed this collaboration to be passionate and fruitful. It is not enough to say that it was necessary to combine the competence of each other; it was necessary at the same time to be a specialist in reactors and geology, that each entered in a way the skin of the other to adopt their language and way of thinking.

This was not easy, because the intellectual starting points are different, and there is always a little lack of comprehension in the dialogue. Geology is not an exact science; a collection of observations, each one of which is debatable, does not necessarily constitute a completely convincing argument for a physicist. The latter is a little annoyed also by the mania for description itself in the natural sciences (what difference, he asks naively, is there between green, black, brown, or grey argillites?), but it is because the geologist notes everything, even nonuseful differences in appearance, that he discovers. From his side, the geologist is not left impressed by calculations that appear to him to rest on results that are a little arbitrary, and he had a hard time to admit that these could modify his own conclusions. It is not evident, when one is not in the field, to appreciate how restricting it is to maintain a fission chain reaction.

They each do not have the same scales of time and space. The specialist in reactors does not have the habit of thinking in millions of years. The geologist, who reasons on the scale of the Francevillian, reproaches him for keeping his eyes too tightly fixed on these few cubic meters where the neutrons conducted their dance, and thus to lack perspective. But the geologist himself, if he understands well hydrothermal phenomena, recognizes that he isn't used to studying their effects for a heat source of this dimension. Each must do some adaptation.

One of the grand moments of these discussions, sometimes rather lively, but always enriching, was naturally the "brain storming" that accompanied the discovery of desilicification. We explained in chapter 4 that this discovery was gradual, preceded by a long maturation. The geologists hesitated to go to the end of their intuition, in part because of the physicists, who were not groundless. The hypothesis final was imposed when it became clear that even if it raised some problems, it also brought a

lot of answers to very important questions. But everything wasn't settled by this. The necessity to integrate all the consequences into the evolution of the nuclear phenomenon required detailed examination of a whole range of questions, and this gave rise to many controversies.

Another great theme of the discussions - more diffuse, but more durable - appeared with the discovery of zones 7 to 9, which quickly revealed themselves to be "troublesome". The principal question is to know if the differences are really significant. The geologists, because they saw in the whole ore deposit a large variety of sandstones, organic matter, and behavior of the terrain, tended to think that they were not fundamental, and to judge that too much attention was paid to this "pocket handkerchief". But the fact that these contained nuclear reactors that posed absolutely constraining problems, and that could not be explained in the same manner as those that were studied further north, is for the physicist a sufficient reason to take an interest in what distinguishes them, and leads one to think that their history has not been exactly the same. On this subject, as on many others, the discussions are not finished and continue to feed friendly controversies.

## **A PASSIONATE INTELLECTUAL ADVENTURE**

As we have been able to realize, the study of the Oklo phenomenon has known numerous new developments. It is with a brief recalling of its events that we would like to finish.

In its discovery, this phenomenon created a vague astonishment and very lively curiosity. Those who lived through the first research projects, in particular the identification of fission, are not about to forget the moments charged with enthusiasm and sometimes intense emotion. After a long association with the subject, we are no longer astonished that a chain reaction spontaneously appeared in a uranium ore body and was controlled during a rather long time, but at the time, this seemed hard to believe, nearly magical! The reconnaissance of the reactors in the terrain in which they were found as so-called fossils, was itself full of unforeseen events. We discovered without ceasing the prolongations with the new zones, each time different and often disconcerting.

Very quickly the problem of representivity of the measurements arose. The beautiful enthusiasm of the beginning had to give way to circumspection. If many of the results were encouraging, others were disconcerting, as these "anticorrelations" that we saw in the distributions, tied as we showed later to the partitioning of water, but which could have been the result of mixing of uranium. Hardly had we put together enough arguments to convince ourselves about the stability of the reactors, than we found the remobilizations in the border regions. Didn't this put everything in question? We convinced ourselves in the beginning that it was only recent alteration (which was false, of course), and we were not yet too disturbed at the time of the Libreville colloquium, where the delivery of the results was not free of a certain flavor of triumph. "Isn't it marvelous, one said, that the isotopic analyses allow us to

localize, to nearly the centimeter, the manner in which water circulated two billion years ago!" We thought at that time that we understood nearly everything.

But the continuation of the work, and in particular the geologic study of the exploitation of the reactor zones, led to a rapid cooling of that optimism. Everything revealed itself to be more complicated than we believed: the importance of tectonic effects, the existence of relatively high temperatures (that could have induced convection currents), the evidence for significant geochemical movements, making us doubt more and more the stability of the reactors. The results from studying the new reactor zones restarted the so-called "Oklo mystery", and made there seem to be mechanisms of propagation that seemed impossible to explain. The demonstration of total desilicification of the C<sub>1</sub> sandstones delivered the final blow. We had to admit that in reality nothing had been understood and that it was necessary to start again from the beginning. At the time of the Paris symposium, that we had organized with the ambition of presenting a final synthesis of all the studies, the crisis was just about overcome, but it was necessary to admit that even if the harvest of results was impressive, the objective had not been reached and that much remained to do.

It is interesting, with the passage of time, to cite a passage from the last paper from that symposium [ref. 172], in which the author of this book [R. Naudet] concluded with these words: "I spoke a little while ago about a "suspense novel". The Oklo phenomenon was not sparing with its surprises to us. Who would have thought, only six months ago, that nuclear reactions could have restructured a uranium ore deposit. As we have progressed in this study, new problems keep coming up, and I think I can say that in five years, our curiosity has not had the time to be blunted." Certainly! Far from being discouraging, the progressive discovery of this complexity was stimulating, and reinforced that interest in a study that without that would have appeared "too pretty to be true". But that came to last longer than five years!

Only three months, in fact, after those lines were written, zone 7 was discovered ... and an avalanche of new problems with it. One of the disturbing subjects was that we absolutely could not justify criticality. A different analog appeared for zones 3 to 5, before we discovered desilicification and the possibility of a start-up in incompletely transformed sandstones. Was this not some new thing that had been completely missed and that could once again put everything in question? We have to admit in any case that these reactors that had perhaps functioned in the heart of a still active petroleum deposit, were rather astonishing. One could jokingly say that nearly all the types of industrial reactors were present at Oklo. After the water reactors - pressurized, boiling, or subcritical - then we discovered reactors moderated by liquid organics or graphite!

In parallel, while we had proof of significant upheavals, the neutronic study of SC36 continued unperturbed to bring arguments in favor of an environment that was nearly perfectly stabilized, and we launched studies of propagation of the reactions to try to explain this paradox. We showed that this could be explained by a very slow progression toward the top. We exposed in chapter 13 those arguments that came, one after the other, to make that solution unacceptable. It is not without some

reticence that we finally recognized that the good solution was undoubtedly 180° from that - that is to say that we must rather envisage a relatively rapid propagation towards the bottom! Let us not dwell too long either on the difficult moments that accompanied that thermal convection study for a computer novice who had the audacity to attempt it.

During this time, the results of the geological study of the Gabon uranium ore bodies, that was undertaken at Strasbourg in the form of a whole series of theses, periodically came to restart the debate on the different phases of emplacement of the uranium, on the manner in which the nuclear phenomenon was inserted there, and on the chronology of the ensemble. As we said, the discussions begun fifteen years ago have never ended, and they will not be closed. The new developments, moreover, are undoubtedly not finished, because one continues to find new reactors. It is not excluded that certain of these will have functioned under still different conditions. The sectors recently put into production could bring new surprises.

This research was definitely a passionate intellectual adventure. And this even though - or perhaps because - it was totally free. It must be recognized that, if this mysterious phenomenon had everything to excite our curiosity, it was in the universe, only an insignificant event. Even with much extrapolation, it concerned, in a past that is beyond our scale, only disturbed portions of our planet, with derisory [very small] consequences. On the other hand, we knew from the beginning that this study could not teach us anything fundamental and that it would lead to no concrete application. Was it not that the passionate aspect of these strange "fossils" was the testimony of the tireless thirst for knowledge that is part of mankind, and its capacity for wonder.

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<sup>1</sup> AECL, Whiteshell Laboratories, Pinawa, Manitoba, Canada

<sup>2</sup> University of Göteborg, Department of General and Marine Microbiology, Göteborg, Sweden

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<sup>1</sup> Conterra AB

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