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with 9 % waste content for the  
vitrification of high-level waste from  
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CONTENT FOR THE VITRIFICATION OF HIGH-LEVEL  
WASTE FROM LWR NUCLEAR REACTORS

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DEVELOPMENT OF GLASS COMPOSITIONS WITH 9 % WASTE CONTENT FOR THE  
VITRIFICATION OF HIGH-LEVEL WASTE FROM LWR NUCLEAR REACTORS

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### Introduction

This work was sponsored by the Nuclear Fuel Safety Project (KBS), a joint organization of Sweden's nuclear power plant owners, and was carried out during the period September 1977 - September 1979. Most of the systematic work was done between October 1978 and September 1979. The preliminary investigations performed during the first year will not be reported in detail, but rather only when their results have a bearing on the subsequent work.

Since the Swedish project for the final isolation of reprocessed and vitrified nuclear waste is based on final deposition of the fuel in a crystalline bedrock repository approximately 40 years after its discharge from the reactor, it is necessary to limit the heat evolution from the waste bodies in order to eliminate the risk of cracking of the surrounding rock. This limitation has necessitated in a reduction of the waste content of the glass to 9 % by weight.

Comparison of a waste glass with 25 % waste content with the basic glass containing no waste has shown that the leaching value of the wastefree composition is higher by a factor of 10. Various published studies concerning high waste contents show that the leaching resistance of the glasses improves up to a waste content of 25 - 30 %, but with higher waste contents the concentrations of the vitrifying constituents, mainly the silica content, are too low and leaching resistance decreases. Hence, reducing the waste content from 20 - 25 % to 9 % causes a decrease of the leaching resistance of the glass, and a modification of the basic glass composition is necessary to compensate for this effect. Part of the development work described here was therefore aimed at modifying the sodium borosilicate glass types in order to make use of the difference between 21 % and 9 % waste content as a new degree of freedom. In parallel,

another type of waste bodies, based on a composition of the "glass ceramic" type, has been tested. Glass ceramics are materials that are produced by melting a glass of suitable composition while adding crystal-nucleating agents, cooling the glass to below the transformation point and transforming the glass into a crystalline product by means of a controlled reheating process. The nucleating agents induce a homogeneous crystallization in the glass mass, producing a fine-grained crystalline product.

The advantages of glass ceramics are: the final product is in a thermodynamically stable form, the radionuclides can be incorporated in the crystals for increased leaching resistance and glass ceramics have better physical properties, such as higher strength, higher heat conductivity and, in certain cases, a very low coefficient of thermal expansion, which can eliminate the risk of cracking due to thermal shock.

Glass ceramics contain varying amounts of residual glassy phase, depending on composition, but it must be pointed out that ceramic waste bodies also contain glassy phase, a fact that often seems to be forgotten by advocates of ceramic waste bodies!

A special advantage of glass ceramic waste bodies is that they are compatible with existing vitrification processes and do not require development of a ceramic sintering technology, which can be quite problematical. The use of a vitrification process to obtain a crystalline product has yet another advantage, namely that a homogeneous distribution of the waste oxides is much easier to achieve in a vitrification process than in a ceramic sintering process. With sintering, there is always a risk that some of the waste oxides will remain unreacted in the ceramic matrix.

#### Experimental methods

All the glasses were prepared with unradioactive, simulated waste composition. Batches of glass components and simulated waste mixture giving 400 g of glass were melted in platinum-rhodium crucibles, with the exception of a few compositions containing components that are incompatible with platinum metals, in which case pure nickel crucibles

were used. The melting temperature was 1200 °C and the melting time about 6 hours. The molten glass was poured out onto a steel plate and cooled rapidly. 85 g of the glass were used for viscosity measurement. The viscometer is of the rotating spindle type and the glass is very thoroughly homogenized during viscosity measurement. After viscosity measurement, the molten glass was poured into a steel mould and immediately placed in an annealing furnace. The homogeneous and annealed glass specimens were used for leaching tests.

The leaching resistance of the glasses was determined by a method that was specially developed with a view towards the analytical resources of our laboratory.

Leaching resistance was determined on 0.315 - 0.500 mm glass grains prepared in accordance with international standards.

10 grams of glass grains and 50 ml of distilled water were autoclaved at 121 °C for 2 hours. Leached quantities of Cs and Sr were determined in the leaching solution, Cs by flame emission spectroscopy and Sr by atomic absorption. The leaching values are expressed as ppm Cs and Sr in the solution. Weight loss of the grains was also determined and expressed as weight percentages. Consequently, our leaching values are not comparable with the values obtained by other laboratories, and determination of leaching rate in accordance with international standards is planned for a later stage of the development program for some of the most promising compositions.

Leaching values of crystallized glasses were determined after heat treatment. For the alkali-borosilicate basic glass compositions, the heat treatment was 800 °C for 14 days, and for the glass ceramic types 900 °C for 65 hours. The heat-treated samples were not homogenized in the viscometer, which may explain a larger spread in the values.

In the case of the glass ceramic types, the coefficients of thermal expansion were determined for the glassy and crystallized states.

#### Development of alkali-borosilicate glass types (ABS series)

The starting composition for the ABS series was a French glass, SON 64 A,

originally containing 21 % waste. When the level of waste oxides was reduced to 9 %, the leaching resistance of the glass decreased significantly. The levels of  $\text{UO}_2$  and  $\text{Fe}_2\text{O}_3$  were reduced in proportion to the total waste content. The glass compositions and leaching values expressed as ppm Cs and Sr in the leaching solution are shown in Table 1. The viscosities are given as the temperature corresponding to 400 poise viscosity, which is limited to 1100 °C in the vitrification process developed by CEA, Marcoule.

The compositions ABS 1 - 5 have different  $\text{Na}_2\text{O}/\text{B}_2\text{O}_3$  ratios and the leaching results show that the composition ABS 1, which has the same  $\text{Na}_2\text{O}/\text{B}_2\text{O}_3$  ratio as SON 64 A glass, is optimal, even at 9 % waste content. This optimal ratio for  $\text{Na}_2\text{O}/\text{B}_2\text{O}_3$  has been maintained in the compositions with some exceptions, where part of the  $\text{Na}_2\text{O}$  or  $\text{B}_2\text{O}_3$  has been replaced by  $\text{Li}_2\text{O}$ .

The glasses ABS 6 - 16 are compositions in which  $\text{Na}_2\text{O}$  and  $\text{B}_2\text{O}_3$  have been replaced by different oxides, which are known from the literature and from tests performed in our laboratory to increase leaching resistance. These oxides were:  $\text{TiO}_2$ ,  $\text{MgO}$ ,  $\text{ZnO}$ ,  $\text{ZrO}_2$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{SnO}_2$  and  $\text{Fe}_2\text{O}_3$ .  $\text{Fe}_2\text{O}_3$  can be a major constituent in certain fuel element constructions.

The best improvements were achieved by  $\text{TiO}_2$  and  $\text{ZnO}$  in glassy form, but after heat treatment at 800 °C for 14 days, the leaching resistance of the glasses containing  $\text{TiO}_2$  (ABS 7) declined. The glass composition containing 6 %  $\text{ZnO}$  (ABS 11) exhibited an insignificant increase in leaching.  $\text{Fe}_2\text{O}_3$  up to 8 % has no detrimental effect.  $\text{ZrO}_2$ ,  $\text{Cr}_2\text{O}_3$  and  $\text{SnO}$  improve leaching resistance, but have a pronounced viscosity-increasing effect, which limits their use at higher concentrations.

ABS 17 and 18 are experimental compositions with combinations of  $\text{TiO}_2$ ,  $\text{ZnO}$  and  $\text{Fe}_2\text{O}_3$ . Since these components were added as substitutes for  $\text{Na}_2\text{O}$  and  $\text{B}_2\text{O}_3$ , the viscosities were rather high, which could be compensated for by the addition of a few percent  $\text{Li}_2\text{O}$ . ABS 17 with the addition of 5 %  $\text{TiO}_2$  + 6 %  $\text{ZnO}$  shows good leaching resistance in the glassy state, but a deterioration after heat treatment and crystallization. This and the results from ABS 7 with 10 %  $\text{TiO}_2$  content show that  $\text{TiO}_2$  addition is only effective in uncrystallized glasses, and since the possibility of

crystallization cannot be ruled out, the use of  $\text{TiO}_2$  in alkali-boro-silicate glasses cannot be recommended.  $\text{TiO}_2$  is a component in some glass compositions developed in Western Germany, but no tests of the effects of crystallization have been reported in the literature.

ABS 19 and 20 are experimental compositions to which  $\text{Fe}_2\text{O}_3$  has been added. Here again, the leaching values are lower than for ABS 1, but the improvement is rather limited. ABS 21 - 24 are compositions aimed at examining the effect of  $\text{Li}_2\text{O}$ . Replacing  $\text{Na}_2\text{O}$  with  $\text{Li}_2\text{O}$  in an amount that should give the same viscosity has no beneficial effect on leaching resistance. The usefulness of  $\text{Li}_2\text{O}$  addition is in reducing viscosity, when desirable.

ABS 25 is a composition with 14 % waste content which was presented by COGEMA at a meeting with Base-Load-Customers. This composition exhibits only slightly lower leaching values than ABS 1, our reference glass.

### Conclusions

At this stage of the investigations, the results show that the leaching values of a sodium-borosilicate glass can be reduced by a factor of approximately 10 by the addition of  $\text{ZnO}$ . This improvement is stable even after heat treatment for 14 days at  $800^\circ\text{C}$ . Additions of  $\text{Fe}_2\text{O}_3$ , which can be unavoidable in certain cases, have no detrimental effect. Additions of  $\text{Li}_2\text{O}$  of 1 - 2 % can reduce viscosity significantly.

### Development of glass ceramic waste bodies (SAC series)

The general considerations for the development of a glass ceramic type of waste body are discussed in the introduction to this report.

It is known that aluminosilicate minerals generally possess better resistance to aqueous attack than pure silicates. This consideration has led us to develop a composition based on the  $\text{SiO}_2$ - $\text{Al}_2\text{O}_3$ - $\text{CaO}$  system (SAC series), which has an eutectic point at  $1170^\circ\text{C}$  (melting point) and contains approximately 62 %  $\text{SiO}_2$ , 14 %  $\text{Al}_2\text{O}_3$  and 24 %  $\text{CaO}$ .

A well known commercial glass has been developed from this composition: E-glass for continuous glass fiber production, which is known for its good chemical resistance. A typical E-glass composition is:

SiO <sub>2</sub>	54.5 %
Al <sub>2</sub> O <sub>3</sub>	14.5 %
CaO + MgO	23.0 %
B <sub>2</sub> O <sub>3</sub>	7.0 %
Alkalies	1.0 %

7 % of the SiO<sub>2</sub> content has been replaced by B<sub>2</sub>O<sub>3</sub> in order to reduce viscosity and increase stability against crystallization. The temperature of E-glass at 400 poise viscosity is close to 1270 °C, which is much too high for radioactive waste vitrification.

Increasing the B<sub>2</sub>O<sub>3</sub> content in order to reduce viscosity would be detrimental for chemical resistance and crystallization, and the most promising component to reduce viscosity is Li<sub>2</sub>O. Li<sub>2</sub>O is also necessary where a low coefficient of thermal expansion is desired. TiO<sub>2</sub> had to be added as nucleating agent, but TiO<sub>2</sub> also has a viscosity reducing effect and is beneficial for chemical resistance. An experimental melt without B<sub>2</sub>O<sub>3</sub> crystallized while it was being poured out. For this reason, a small amount (3.4 % B<sub>2</sub>O<sub>3</sub>) has been added to the glasses in most of the experiments. 4.0 % Na<sub>2</sub>O has been added to most of the compositions, since this corresponds to the amount of an alkaline cleaning solution from the reprocessing process which accompanies the waste at 9 % waste content. Although the elimination of Na<sub>2</sub>O does not seem to be practical, two glasses have been melted without the addition of Na<sub>2</sub>O in order to test the effect of Na<sub>2</sub>O in the glass.

The experimental glasses were tested for leaching of Cs and Sr, weight loss and coefficient of thermal expansion in the glassy and crystallized states. The aim of the experiments was to develop compositions with good leaching resistance in both the glassy and crystalline states and low coefficient of thermal expansion in the crystallized state in order to eliminate the risk of cracking due to thermal shock, which is probable in connection with decontamination and pool-cooling of the glass-containing canisters.

It has been observed that these compositions soften before crystallization, and even if the glass is initially cracked, it is healed by heat treatment and the final product is a crack-free homogeneous crystalline block.



The compositions, leaching values for Cs and Sr, weight losses, coefficients of thermal expansion for glassy and crystallized materials and temperatures at 400 poise viscosity for the glass melts are presented in Table 2.

SAC 1 - 6 were made with different  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios in order to find the optimal composition. The leaching values for Cs and Sr at 9 %  $\text{Al}_2\text{O}_3$  and higher levels exhibit small differences. The coefficients of thermal expansion decrease with increasing  $\text{Al}_2\text{O}_3$  content from  $\alpha = 8.5 \cdot 10^{-6}$  at 8 %  $\text{Al}_2\text{O}_3$  to  $\alpha = 1.3 \cdot 10^{-6}$  at 14.8 %  $\text{Al}_2\text{O}_3$  for the crystallized materials, but are practically unchanged in the glassy state. A decision has been made to continue with 14.8 %  $\text{Al}_2\text{O}_3$  content. In further experiments, the effect of replacing CaO with different oxides has been tested.

Surprisingly, SAC 7 and 7 A (7 A reproduction of 7) exhibited increased leaching values for Cs and Sr after crystallization, although this composition was made without the addition of  $\text{Na}_2\text{O}$ . But this glass also exhibited the lowest coefficient of thermal expansion, equivalent to the best low-expansion glass ceramics and of the same order as silica glass, which is completely shock resistant.

The addition of  $\text{MgO}$ ,  $\text{ZnO}$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{SnO}_2$  and  $\text{Fe}_2\text{O}_3$  have been tested in the following experiments (SAC 8 - 16). The results were similar to those with ABS glasses - the best results from the viewpoint of leaching resistance were obtained from 6 %  $\text{ZnO}$  addition. This glass exhibited low release of Cs and Sr in the glassy and crystallized states. However, the glass cracked during crystallization and the coefficient of thermal expansion increased to  $\alpha = 4.34 \cdot 10^{-6}$ . This increase of the coefficient of thermal expansion was unexpected, as  $\text{ZnO}$  is a common component in low expansion glass ceramics.

Additions of  $\text{Fe}_2\text{O}_3$  had no detrimental effect on leaching.

As our aim is to develop a composition with high leaching resistance in the glassy and crystallized states and a low coefficient of thermal expansion in the crystallized state without cracking or other mechanical damage, additional experiments were carried out. It was suspected that cracking is a consequence of stresses between the crystalline and residual

glassy phases, or between different crystalline phases. In order to reduce the glassy phase, 3 glasses were made where  $B_2O_3$  was replaced by BaO, which is known to reduce the rate of crystallization. These glasses are SAC 17, 18 and 19.

SAC 17 with 0 %  $B_2O_3$  content did not crack, SAC 17 with 1.4 %  $B_2O_3$  cracked to a small extent and SAC 18 with 2.0 %  $B_2O_3$  cracked more severely. Thus, the concentration of  $B_2O_3$  influences the tendency to crack, and future investigations will be concentrated on  $B_2O_3$ -free compositions.

The leaching values for SAC 17 are low for the glassy and crystallized states and the coefficient of thermal expansion is nearly the same as that for commercial "pyrex" glass, which is low, but not low enough to provide a full guarantee against cracking in the case of severe thermal shock. The viscosity of the glass is below the stated limit, but it would be an advantage to reduce it further, which can be achieved by adding more  $Li_2O$ , which will probably reduce the coefficient of thermal expansion as well.

### Conclusions

The leaching resistances of the glass ceramic waste bodies are on the same level as the best values for the ABS glasses, but the crystallized glass ceramic materials have a significantly lower coefficient of thermal expansion than glassy waste bodies. Although there are no measurement results available at this time concerning mechanical strength, a subjective evaluation based on the crossing of glass grains indicates that the crystallized glass ceramics are much tougher than glasses - it is much more difficult to cross them. It is generally known that glass ceramics possess higher physical strength and better heat conductivity than glasses.

### Distribution of elements in crystallized glass ceramic waste bodies

Several studies were conducted in order to determine the distribution of the most important elements in the crystallized glass ceramic materials. The purpose of this investigation was to see whether the important nuclides Cs, Sr, Nd, La are concentrated in crystals or distributed evenly in the residual glassy phase.

In the first study, SAC 11 was examined by electron-microprobe and the

distribution of Cs, Nd and La was studied. Nd and La were clearly concentrated in crystals, but the distribution of Cs showed a diffuse picture with only slight deviation from an even distribution pattern. The content of SrO (0.26 %) was too low for any distribution pattern to be recognizable.

According to (1), the minimum concentration for Cs to form pollucite ( $\text{CsAlSi}_2\text{O}_6$ ), which is the desirable host phase for Cs, is 2 %. In the second study, the concentration of Cs was increased to 2.5 %  $\text{Cs}_2\text{O}$  and the concentration of Sr to 1.0 % SrO. A clear concentration of Cs could be observed in this specimen and Sr showed a more distinct pattern as well. However, this specimen showed a high degree of inhomogeneity, so a new specimen was prepared which was thoroughly homogenized during melting by mechanical stirring. This sample had the composition of SAC 17 with the  $\text{Cs}_2\text{O}$  content increased from 0.88 % to 2.5 % and the SrO from 0.26 % to 1.0 %. The concentrations of the investigated elements in oxide form were:

$\text{SiO}_2$	41.9 %	$\text{Na}_2\text{O}$	4.0 %
$\text{Al}_2\text{O}_3$	14.8 %	$\text{TiO}_2$	6.6 %
CaO	9.3 %	$\text{Cs}_2\text{O}$	2.5 %
BaO	3.4 %	$\text{UO}_2$	1.66 %
SrO	1.0 %	$\text{Nd}_2\text{O}_3$	1.21 %
ZnO	6.0 %	$\text{La}_2\text{O}_3$	0.71 %

In Fig. 1, the electron micrograph showing the morphological appearance of a polished surface is presented first, followed by radiographs showing the distribution of Si, Al, Ca, Ba, Sr, Zn, Na, Ti, Cs, U, Nd och La.

Examination of the electron micrograph reveals a large medium-gray crystal on the left side of the picture, which coincides with the Zn distribution. This crystal also contains large amounts of Ca and an indication of some enrichment of Sr, but no Ba. There is a clear deficiency of Al in this crystal. The only known mineral which can be suspected is hardystonite:  $\text{Ca}_2\text{ZnSi}_2\text{O}_7$ .

Some of the crystals contain large amounts of Ti and Ca, which indicates that these crystals are perovskite ( $\text{CaTiO}_3$ ), which could be a host phase

for Sr, but no sign of Sr enrichment can be detected. The Sr content is probably too low in this case. These perovskite crystals seem to be host crystals for Nd. La can be found concentrated in some smaller titanium-containing crystals, but not in the larger ones. These smaller titanium-containing crystals contain only Nd and La. These crystals are probably one of the lanthanum titanates  $\text{La}_2\text{O}_3 \cdot \text{TiO}_2$ ,  $\text{La}_2\text{O}_3 \cdot 2\text{TiO}_2$  or  $2\text{La}_2\text{O}_3 \cdot 9\text{TiO}_2$  reported by MacChesney and Sauer (2). In these lanthanum titanates, part of the lanthanum can be replaced by neodymium or other rare earth elements. Thus, rare earth elements have two different host phases: the lanthanum titanates and perovskite.

Cs is found in all areas where there is a deficiency of Ca and an elevated concentration of Al. The Si distribution is too indistinct, only a very slight indication can be discerned. There are signs, but no proof of the presence of pollucite:  $(\text{Cs}, \text{Na}) \cdot \text{Al Si}_2\text{O}_6$ .

The Ba distribution cannot be correlated with any other elements, but Al, Si, and Ba are probably in the form of h-celsian:  $\text{BaAl}_2\text{Si}_2\text{O}_8$ .

X-ray diffraction studies for identification of crystal forms are planned for the future, but it is uncertain whether it will be possible to separate different crystal forms if there are many types of crystals present in the same specimen.

#### Separation of molybdate phase

The problem of "molybdate phase" separation has been discussed extensively in the literature. It has been postulated that the solubility of  $\text{MoO}_3$  lies between 3 and 4 % in borosilicate glass. Nevertheless, in one of our experiments we found a full visible separation of molybdate phase from an alkali-borosilicate glass with 9 % waste content, which corresponds to a  $\text{MoO}_3$  content of 1.6 %, after heat treatment at 800 °C during 14 days. (Fig. 2)

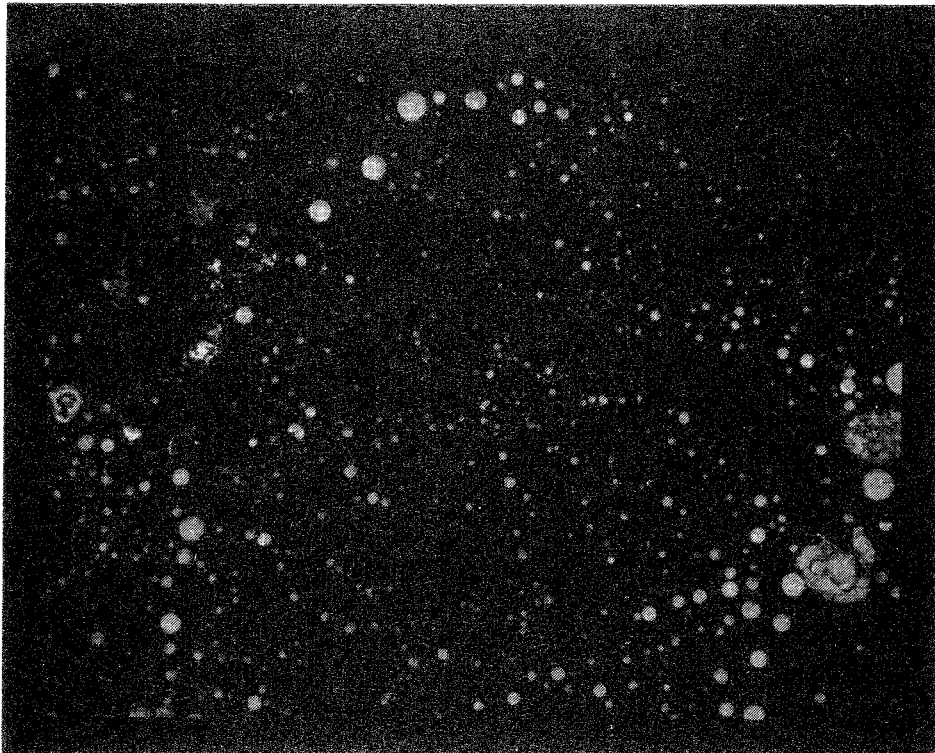


Fig. 2

This indicates that the separation of the molybdate phase is highly dependent upon the thermal history of the glass. This may explain the fact that under normal cooling, molybdate phase does not separate from laboratory scale melts, but in some cases from full scale melts, where the speed of cooling is much lower due to the large mass of the melt. Other factors must also have a bearing on the separation of molybdate phase, since in our own experiments no separation of molybdate phase could be seen from the same glass composition and heat treatment. The separated droplets were examined by electron-microprobe analysis and the phase contained approximately 70 % sodium molybdate, Cs was found at low concentration, Sr at rather high concentration, La at low concentration and Nd could not be detected.

In order to study the different parameters influencing the separation of the molybdate phase, experiments were conducted with the  $\text{MoO}_3$  content raised to 5 %.

In a sodium-borosilicate glass, the molybdate phase could not be seen in the molten glass immediately after pouring out into a steel mould, but an extensive phase separation was observed during the cooling period.

The addition of different reducing agents reduced the tendency towards phase separation; carbon black, graphite and elementary silicon were tested. The best effect was obtained by replacing 3 % of the  $\text{SiO}_2$  by an equivalent quantity of silicon. In an alkali-free SAC glass, no molybdate phase separation could be observed with a 5 %  $\text{MoO}_3$  content.

It is reported in the literature that the separation of molybdate phase can be counteracted by increasing the  $\text{B}_2\text{O}_3$  content. On the other hand, at higher  $\text{B}_2\text{O}_3$  content, the chemical resistance of the glass declines.

The author is of the opinion that the tendency towards molybdate phase separation is highly dependent upon the "alkalinity" of the glass, which explains the beneficial effect of  $\text{B}_2\text{O}_3$  addition and the fact that alkali-free glasses show no visible phase separation. Highly acidic phosphate glasses can also contain large amounts of molybdenum.

A series of experiments was conducted on two different glass types with normal  $\text{MoO}_3$  content (1.6 %), with and without the addition of reducing agents. The glass types were alkali-borosilicate glass modified with 6 %  $\text{ZnO}$  (ABS 11) and a glass ceramic type, SAC 11.

Each of these glasses were melted unmodified and with the addition of elementary silicon and silicon carbide replacing 3 % of the  $\text{SiO}_2$ .

The glasses were melted in nickel crucibles, since platinum is incompatible with silicon and silicon carbide. After melting, approximately half of the molten glass was poured into a stainless steel crucible. The weights in the nickel crucible and in the stainless steel crucible were measured. The crucibles were annealed, and after cooling, the surfaces of the glasses were washed with hot distilled water in order to dissolve any molybdates from the glass surfaces. The dissolved Mo content was determined analytically. The stainless steel crucibles were then heat-treated: the ABS 11 glasses at 800 °C for 14 days and the SAC 11 glasses at 900 °C for 65 hours. After heat treatment and slow cooling, the surfaces were washed again with hot distilled water and the Mo contents determined.

The quantities of dissolved Mo were calculated per 100 grams of glass, and the results of these experiments are shown in Table 3.

	Ni-crucible	Stainless steel crucible	Stainless steel crucible after heat treatment
ABS 11	1.22 $\mu\text{g}$	5.62 $\mu\text{g}$	1.14 mg
ABS 11 + Si	0.122 mg	4.11 $\mu\text{g}$	1.24 mg
ABS 11 + SiC	1.42 mg	4.29 $\mu\text{g}$	0.98 mg
SAC 11	2.95 $\mu\text{g}$	1.50 $\mu\text{g}$	16.91 $\mu\text{g}$
SAC 11 + Si	1.19 $\mu\text{g}$	1.50 $\mu\text{g}$	68.42 $\mu\text{g}$
SAC 11 + SiC	0.94 $\mu\text{g}$	1.13 $\mu\text{g}$	32.05 $\mu\text{g}$

Table 3. Mo dissolved from the glass surfaces per 100 g of glass.

No significant effect of the reducing agents could be observed. The alkali-borosilicate type ABS glasses show a wide variation from microgram to milligram quantities, which can be explained by a random distribution of larger or smaller droplets of molybdate phase. After heat treatment, all values were on the milligram level due to separation of molybdate phase on the surface during heat treatment.

All of the SAC glasses with low alkali content show lower levels of Mo separation, which never exceeds microgram levels. A significant increase occurs during heat treatment, but the levels are still of microgram order.

These results show that Mo phase separation occurs during heat treatment, but the total quantities of separated Mo are lower for low-alkali SAC type glasses by a factor of approximately 50.

The factors influencing molybdenum phase separation require further studies.

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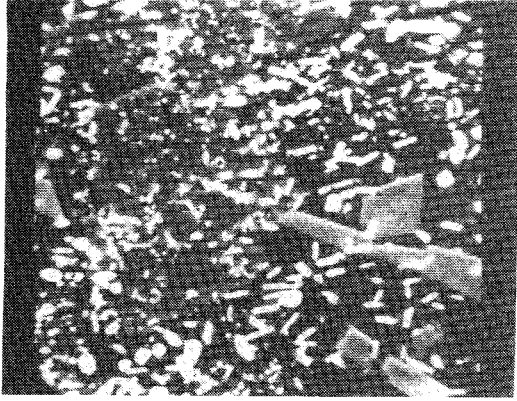
No.	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	B <sub>2</sub> O <sub>3</sub>	UO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	FP+ TRU	TiO <sub>2</sub>	MgO	ZnO	ZrO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	SnO <sub>2</sub>	Li <sub>2</sub> O	glass			400 poise viscosity °C	crystallized		
															Cs ppm	Sr ppm	w.loss %		Cs ppm	Sr ppm	w.loss %
1	50.0	2.1	13.6	23.1	1.66	0.6	9.0								6.35	0.28	1.27	1008			
2	50.0	2.1	16.7	20.0	1.66	0.6	9.0								10.95	0.27	1.36	978	3.70	0.61	1.22
3	50.0	2.1	11.5	25.2	1.66	0.6	9.0								11.25	0.43	1.70	1020			
4	50.0	2.1	12.5	24.2	1.66	0.6	9.0								10.77	0.37	1.47	1013			
5	50.0	2.1	14.5	22.2	1.66	0.6	9.0								9.72	0.26	1.27	993			
6	50.0	2.1	11.7	19.95	1.66	0.6	9.0	5.0							1.37	0.030	0.52	1025			
7	50.0	2.1	9.89	16.85	1.66	0.6	9.0	10.0							0.49	0.018	0.25	1050	2.03	0.065	0.66
8	50.0	2.1	12.49	21.21	1.66	0.6	9.0		3.0						3.03	0.335	0.765	1024			
9	50.0	2.1	11.38	19.32	1.66	0.6	9.0		6.0						3.67	0.405	0.68	1024			
10	50.0	2.1	12.49	21.21	1.66	0.6	9.0			3.0					1.53	0.080	0.635	1037			
11	50.0	2.1	11.38	19.32	1.66	0.6	9.0			6.0					0.69	0.029	0.25	1051	0.99	0.032	0.31
12	50.0	2.1	12.12	20.58	1.66	4.0	9.0								1.89	0.162	0.55	1018			
13	50.0	2.1	10.64	18.06	1.66	8.0	9.0				2.0				1.35	0.066	0.37	1025			
14	50.0	2.1	12.86	21.84	1.66	0.6	9.0					2.0			2.12	0.154	0.63	1062			
15	50.0	2.1	12.86	21.84	1.66	0.6	9.0					2.0			3.59	0.142	0.85	1053			
16	50.0	2.1	11.75	19.95	1.66	0.6	9.0						5.0		1.47	0.047	0.44	1083			
17	50.0	2.1	9.52	16.18	1.66	0.6	9.0	5.0		6.0					1.04	<0.02	0.074	1080	3.24	0.061	0.35
18	50.0	2.1	8.78	14.92	1.66	4.0	9.0	5.0		4.0					0.89	<0.02	0.050	1113			
19	48.05	2.01	13.07	22.20	1.66	4.0	9.0								4.06	0.202	1.114	1018			
20	45.80	1.92	12.46	21.16	1.66	8.0	9.0								3.92	0.114	0.919	1026			
21	51.60	2.2	9.1	23.9	1.66	0.6	9.0							1.8	3.67	0.27	1.456	989			
22	51.60	2.2	7.4	19.6	1.66	0.6	9.0			6.0				1.8	1.03	0.040	0.286	1047			
23	51.60	2.2	10.0	17.0	1.66	0.6	9.0			6.0				1.8	1.16	0.017	0.068	1012			
24	51.60	2.2	8.5	14.5	1.66	4.0	9.0			6.0				1.8	1.06	0.016	0.079	1043			
25	47.60	1.4	12.9	18.6	1.9	5.5	12.1								5.23	0.070	0.525	1016			
															3.98	0.052	0.391		per 9 % FP + TRU		

Table 1. ABS glasses: Compositions in weight percentages, leaching values for Cs and Sr as ppm in the leaching solution, weight losses in percentages, temperatures corresponding 400 poise viscosity level and some leaching values for heat treated glasses at 800 °C for 14 days.

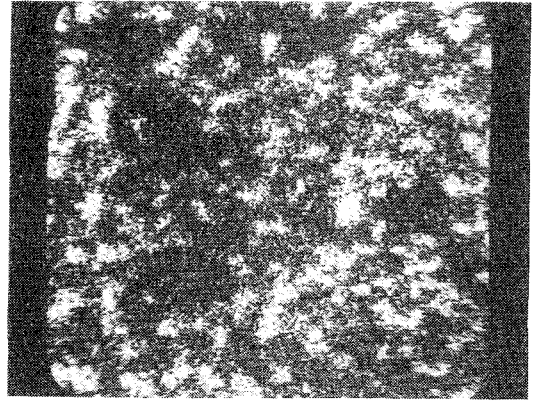


No.	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	B <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	CaO	Li <sub>2</sub> O	UO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	FP+ TRU	MgO	ZnO	ZrO <sub>2</sub>	TiO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	SnO <sub>2</sub>	BaO	glass				crystallized			α · 10 <sup>6</sup>		
																	Cs ppm	Sr	w. loss %	°C	Cs ppm	Sr	w. loss %	glass	cryst.	
1	44.8	11.9	3.4	4.0	15.3	3.8	1.66	0.6	9.0									1.00	0.18	0.20	1018	1.88	0.064	0.071	9.04	5.15
2	47.7	9.0	3.4	4.0	15.3	3.8	1.66	0.6	9.0				6.6					1.07	0.17	0.21	1022	1.55	0.109	0.071	9.23	7.75
3	41.9	14.8	3.4	4.0	15.3	3.8	1.66	0.6	9.0				6.6					1.08	0.20	0.06	1029	1.95	0.045	0.118	9.21	1.35
4	48.7	8.0	3.4	4.0	15.3	3.8	1.66	0.6	9.0				6.6					0.75	0.066	0.031	1003	2.22	0.099	0.102	9.25	8.39
5	49.7	7.0	3.4	4.0	15.3	3.8	1.66	0.6	9.0				6.6					0.56	0.067	0.053	993	2.18	0.140	0.072	10.17	8.50
6	40.7	16.0	3.4	4.0	15.3	3.8	1.66	0.6	9.0				6.6					0.87	0.096	0.083	1036	2.26	0.058	0.118	8.46	1.36
7	43.9	15.5	3.6	0.0	16.0	4.0	1.66	0.6	9.0				6.6					0.63	0.098	0.093	1063	3.41	0.359	0.086	7.89	0.63
7a	43.9	15.5	3.6	0.0	16.0	4.0	1.66	0.6	9.0				6.9					0.63	0.098	0.093	1063	3.41	0.359	0.086	7.89	0.63
8	41.9	14.8	3.4	4.0	10.3	3.8	1.66	0.6	9.0	5.0			6.6					0.99	0.065	0.076	1028	3.49	0.743	0.112		0.31
9	41.9	14.8	3.4	4.0	5.3	3.8	1.66	0.6	9.0	10.0			6.6					1.47	0.040	0.053	1071	2.20	0.034	0.090	8.96	5.61
10	41.9	14.8	3.4	4.0	12.3	3.8	1.66	0.6	9.0		3.0		6.6					0.76	0.075	0.093	1029	2.64	0.070	0.090	8.88	7.15
11	41.9	14.8	3.4	4.0	9.3	3.8	1.66	0.6	9.0		6.0		6.6					0.76	0.075	0.093	1029	0.80	0.126	0.061	8.92	8.49
12	41.9	14.8	3.4	4.0	11.3	3.8	1.66	4.0	9.0				6.6					0.74	0.062	0.040	1043	0.71	0.047	0.065	9.11	4.34
13	41.9	14.8	3.4	4.0	7.3	3.8	1.66	8.0	9.0				6.6					0.74	0.053	0.058	1035	1.07	0.055	0.076	9.08	9.87
14	41.9	14.8	3.4	4.0	15.3	3.8	1.66	0.6	9.0			2.0	4.6					0.74	0.057	0.086	1063	1.23	0.028	0.056	8.83	4.90
15	41.9	14.8	3.4	4.0	15.3	3.8	1.66	0.6	9.0				4.6					0.77	0.099	0.092	1037	1.76	0.119	0.069	9.15	5.90
16	41.9	14.8	3.4	4.0	10.3	3.8	1.66	0.6	9.0				4.6		2.0			0.88	0.110	0.074	1032	1.74	0.055	0.089	9.37	3.72
17	41.9	14.8	0.0	4.0	9.3	3.8	1.66	0.6	9.0		6.0		6.6			5.0		0.85	0.054	0.081	1074	1.14	0.090	0.083	8.76	5.48
18	41.9	14.8	2.0	4.0	11.3	3.8	1.66	0.6	9.0		6.0		6.6				3.4	0.78	0.070	0.064	1064	0.85	0.030	1.07	6.72	3.91
19	41.9	14.8	1.4	4.0	9.3	3.8	1.66	0.6	9.0		6.0		6.6				2.0	0.91	0.059	0.076	1039	0.72	0.035	0.064	8.64	5.26
											6.0		6.6					0.83	0.071	0.039	1051	0.62	0.037	0.025	6.34	6.36

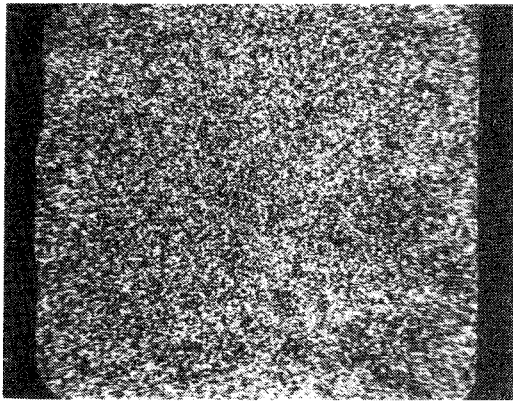
Table 2. SAC glasses: Compositions in weight percentages, leaching values for Cs and Sr as ppm in the leaching solutions, weight losses in percentages, temperatures corresponding 400 poise viscosity level, leaching values for crystallized samples, thermal expansion coefficients for glassy and crystallized samples.



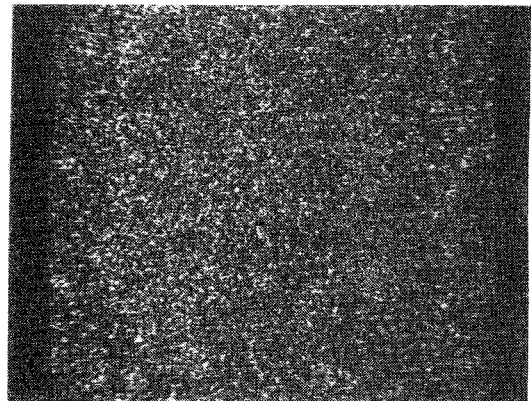
Electron picture



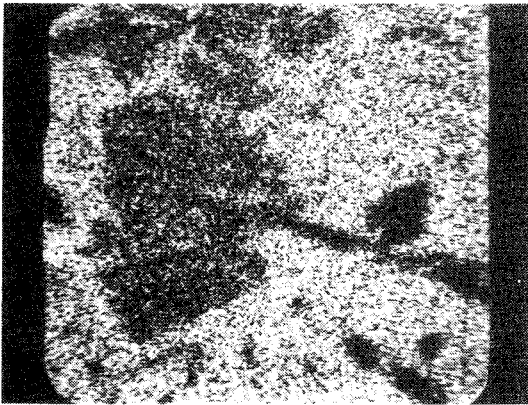
Ba



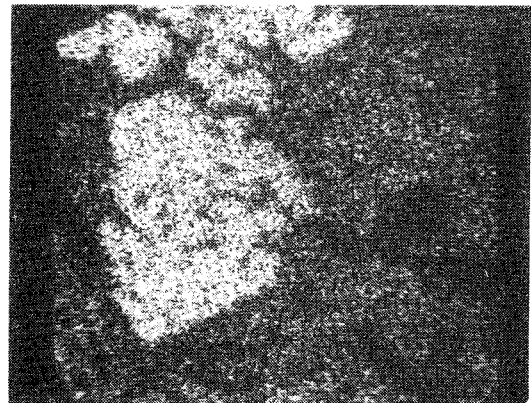
Si



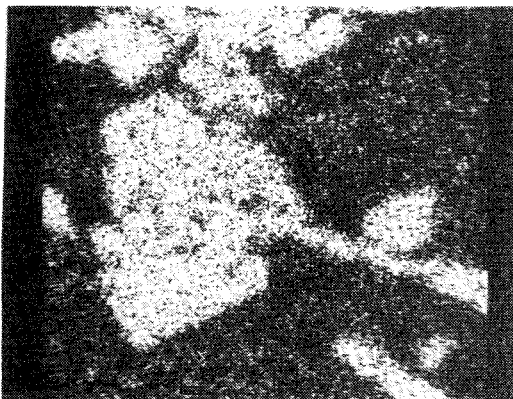
Sr



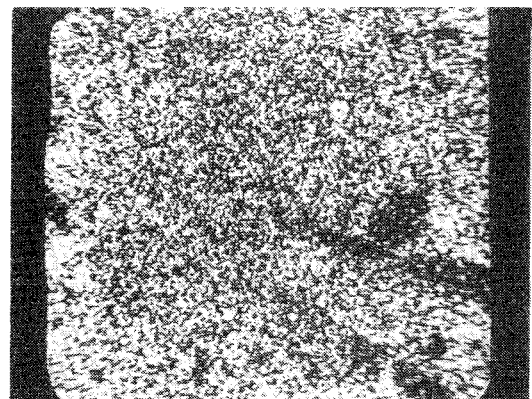
Al



Zn

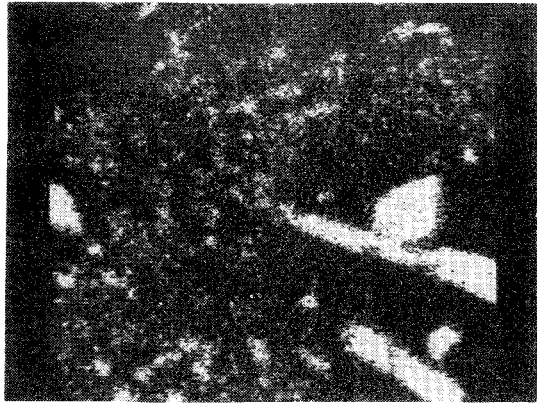


Ca

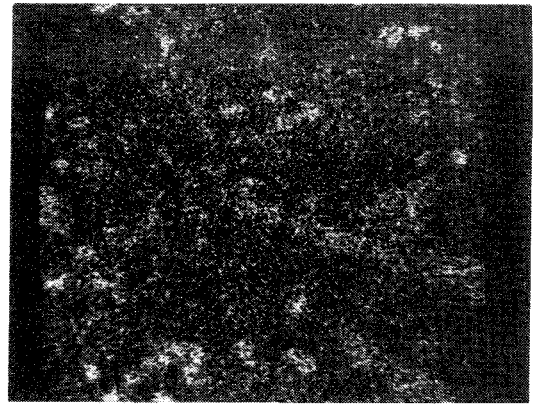


Na

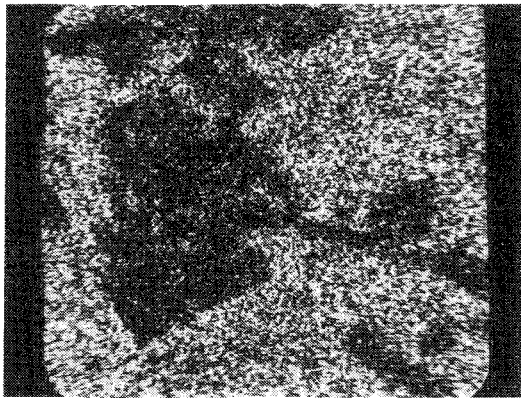
Fig 1a. Element distribution in crystallized SAC waste form



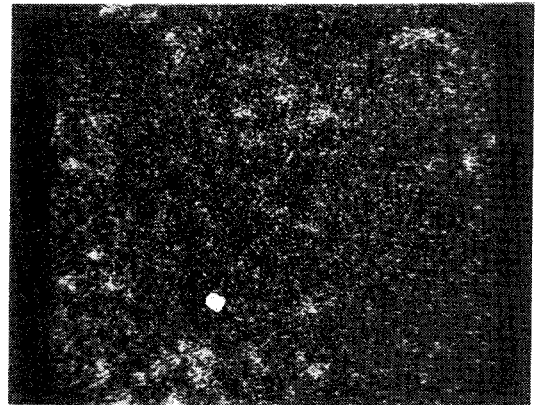
Tl



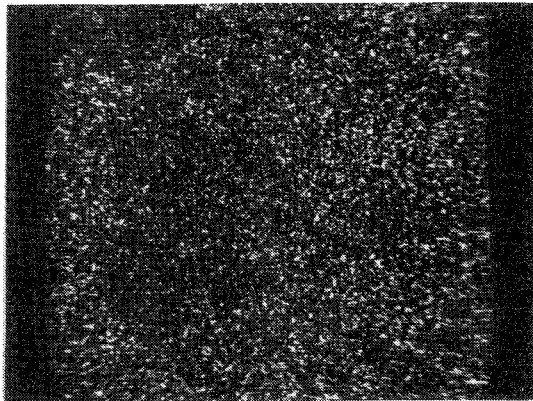
Nd



Cs



La



U

Fig 1b. Element distribution in crystallized SAC waste form

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