INVESTIGATIONS OF THE COOLING OF TWO HIGH ACTIVE GLASS CANISTERS BY ACOUSTIC EMISSION ANALYSIS (AEA)

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ABSTRACT

The cooling of high level waste (HLW) glass containing canisters was investigated with acoustic emission analysis (AEA). The suitability of AEA to detect crack formation during cooling was proven in earlier experiments with nonradioactive glasses. The acoustic emission (AE) was measured with a commercial AEA monitor system. Two different HLW glass containing canisters were used for the cooling experiments, one contained a ceramic (Al_2O_3) incan lining to avoid the direct contact of the glass with the canister wall. 14 experiments with three different cooling conditions were performed. The fastest cooling was in air, a slower cooling was achieved by applying a thermal insulated cooling pot and the slowest cooling was performed under controlled conditions in a furnace. After the solidification of the glass the AE increases with decreasing temperatures. When cooling in air a strong increase of AE was observed (about 500°C) which was caused by stress induced cracks due to the large thermal gradient. A significant reduction of AE was observed for the canister with Al_2O_3-lining. The main difference beween experiments with radioactive and nonradioactive glasses was the crack formation caused by the residual thermal gradient, which was due to the internal heat production of the radioactive glass.

INTRODUCTION

One concept for final disposal of high level waste (HLW) in the Federal Republic of Germany (FRG) is to incorporate the HLW in a borosilicate glass and to fill the glass melt in steel canisters. The glass melt is solidified below the transformation temperature of the glass (about 500°C). During the further cooling cracks may be formed which increase the glass surface area and influence the transport of the decay heat. The increase of the surface area will accelerate the material release in case of leaching.

The aims of these investigations were to check if the results obtained in experiments with nonradioactive glasses are valid for the cooling of radioactive glasses and especially to confirm the suitability of acoustic emission analysis (AEA) for detecting the formation of cracks during the cooling. The experiments with non-radioactive glasses were performed by the Kernforschungszentrum Karlsruhe (FRG), in cooperation with the Fraunhofer-Institut fuer Silicatforschung, Wuerzburg (FRG). The experiments showed, that during the cooling in air or in a thermal insulated cooling equipment cracks occurred, which led to an increase of the surface by a factor of up to 25. By controlled cooling the formation of cracks was minimized. An optimal cooling program could be realized with the help of the AEA. In temperature regions with high acoustic emission lower cooling rates were applied.

EXPERIMENTAL

Acoustic emission (AE) is caused by the formation and the growth of cracks in a solid material. An overview about the AEA for non-metallic materials is given by (1.). For acoustic emission analysis the ultrasonic frequency region from 50 khz until 1 Mhz is normally used to avoid the influence of the environmental noise. A sensor (piezoceramic material) transforms the mechanical signals to electrical signals. In Fig. 1 a cross-section of the instrumented canisters is shown. For measuring the thermal distribution inside the glass 15 thermocouple pairs were located in the canister. In three height levels, the radial temperature gradient was measured with 5 thermocouple pairs from the centre to the skin of the glass. For the evaluation of the AEA-data the temperatures in the centre (T2) and at the skin (T1) were used. The two sets of stainless steel waveguides were mounted acousticly decoupled from the canister wall at a distance of 5 cm to the wall. One waveguide formed a loop, the other ended in the glass. The waveguides had a diameter of 1.6 mm and lengths up to 8.7 m. The sensors were directly adhered to the ends of the waveguides steel waveguides and thermocouples and were located outside the canisters. The temperature and the chemical stability of the waveguides in glass melts were excellent as tested. The damping of the acoustic signals was 1.5 dB/m and was up to 800°C independent of the temperature. The electrical signals from the sensors were detected

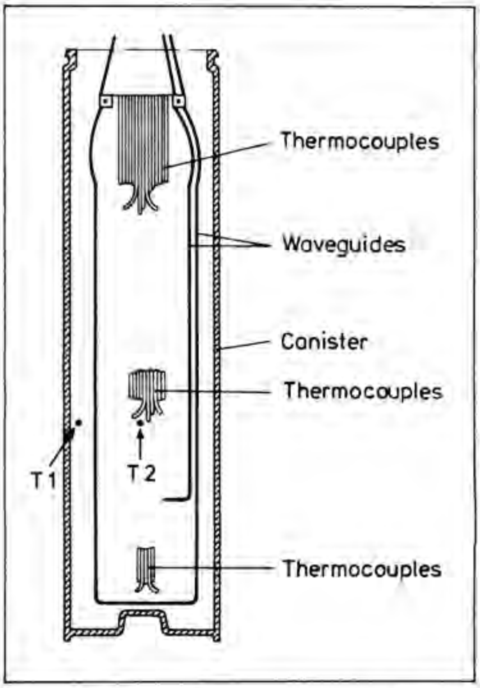

Fig. 1. Cross-Section of the Instrumented Canister With Stainless Steel Waveguides and Thermocouples.

by a commercial acoustic emission analysis equipment (Dunegan). The maximum amplitude, the duration, the rise time and the time of the signal arrival from the AEA-signals were determined and stored at diskettes for further evaluation. The equipment was designed to measure AE in a "hot cell" with a high gamma dosis.

Two instrumented canisters (no. 13 and 19) with a diameter of 30 cm and a height of 100 cm were used for the experiments. Canister no. 13 had a Al_2O_3-lining to avoid the direct contact of the glass with the canister wall and the additional stress caused by the shrinking of the canister on the glass (different thermal expansion of canister and glass). Both canisters were filled with a glass containing Cs-137 and Sr-90 as radioactive elements (radioactivity about 8 1 10^{15} Bq) which lead to a decay heat of about 1.0 and 1.2 kW (see Fig. 2.).

The canisters were heated in a furnace up to 950°C until the glass inside the canister was molten. Then the canisters were cooled down at first in air to 600°C at the outer glass region (T1) during all experiments. Three different cooling conditions were applied to the canisters below 600 °C:

- The fastest and easiest cooling was that in an air cooling frame (ACF).
- To reduce the cooling velocity and the thermal gradient in the glass a thermal insulated cooling pot (ICP) was used for the canisters.
- A further reduction of the thermal gradient was achieved by controlled cooling (CNTL) in a temperature regulated furnace.

In the latter case the final skin temperature (T1) was limited to about 380°C by the thermal insulation of the furnace and the decay heat. By additional forced air flow through the furnace the final temperature could be reduced to 250°C. When stopping the air flow in the furnace the temperature of the glass surface increased again to 340°C. A further reduction of the temperature could only be achieved by removing the canister from the furnace. An optimal cooling process consisted of three phases: controlled cooling in the furnace (CNTL: from 600°C down to 380°C), slow cooling in an insulated pot (ICP: down to 320°C) and the final cooling in air (ACF: down to 150°C). Also after direct cooling in the ICP a further cooling in the ACF was necessary. All three types of cooling experiments were pe formed with both canisters.

The AEA-data were evaluated by computer programs. The acoustic energy is the time integral of the squared signal amplitude and was calculated by the signal parameters. The calculated acoustic energy or the acoustic energy sum were evaluated as functions of time and of the temperatures T1 (on the glass skin) or T2 (in the centre). The acoustic energy was used for the evaluation, because this parameter characterizes the size of a AE-signal and was correlated with the crack size in experiments with nonradio- active glasses. Friction noise between glass and metall causes only low acoustic energy signals and can thus be distinguished from noises of crack formation.

RESULTS

Acoustic emission during cooling in the ACF and the ICP:

The cooling in air was the fastest cooling condition (ACF). About 17 h were needed to reach the final temperature distribution of 150°C at the skin (T1) and 250°C at the centre (T2; Fig. 2). At the beginning of the cooling the temperature gradient in-side the glass was 400 K (between T1 and T2) and decreased with cooling time. The acoustic energy sum (Fig. 3) increased strongly in the first 500 min and reached about 90 % of the final value.

A slower cooling as in air was achieved by applying the ICP. At the beginning of the cooling a thermal gradient of about 150 K was measured (Fig. 4). At the end of the cooling the thermal gradient was reduced to 100 K. A steady state temperature distribution was reached after 30 - 35 h, with a centre temperature of 400°C and a skin temperature of 300°C. The corresponding acous- tic energy sum is shown in Fig. 5. During the first 500 min (T1 above 500°C) no AE was measured. After 1000 min the acoustic energy sum

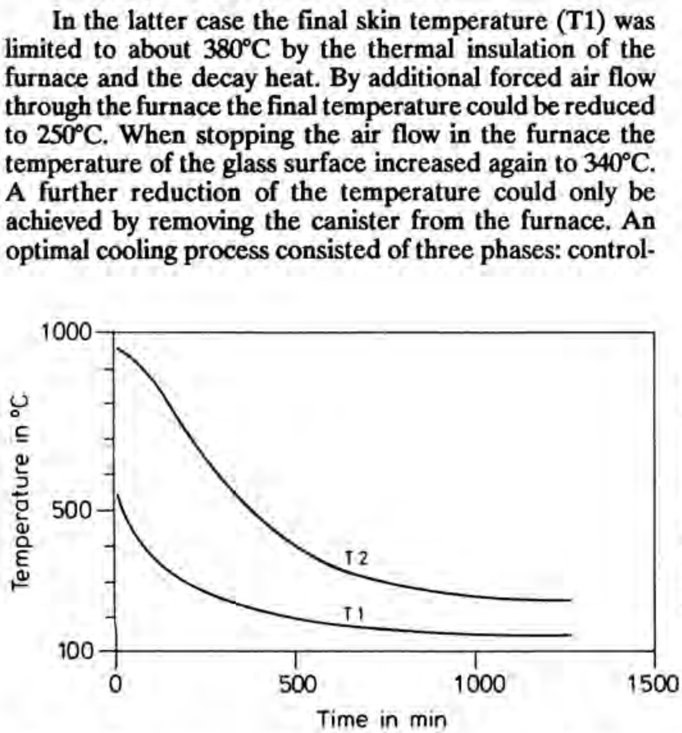

Fig. 2. Temperatures T1 and T2 as Function of Cooling Time for Canister No. 19 During Cooling in Air (ACF).

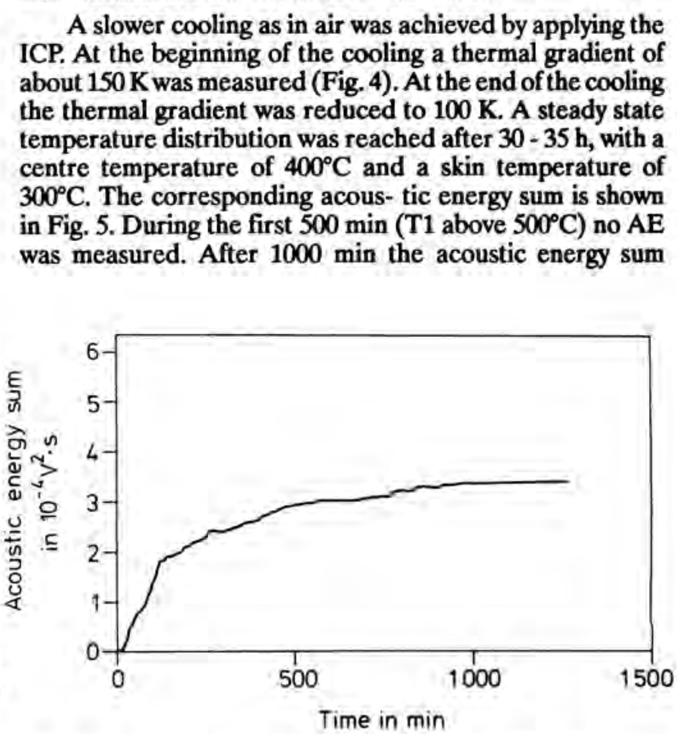

Fig. 3. Acoustic Energy Sum as Function of the Cooling Time for Canister No. 19 During Cooling in Air (ACF).

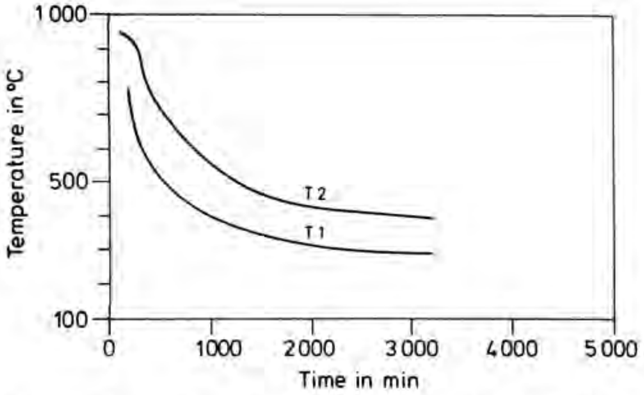

Fig. 4. Temperatures T1 and T2 as Function of Cooling Time for Canister No. 19 During Cooling in the ICP.

reached about 75 % of the final value (which was reached after 2000 min). During the further cooling only a very small increase in the acoustic energy sum was observed.

Acoustic emission during controlled cooling (CNTL): Fig. 6 shows the measured temperatures T1 and T2 during controlled cooling of canister no. 13 (with Al_2O_3-lining) as function of time. The time scale starts with the heating of the canister. The thermal gradient between the centre and the skin of the glass was about 90 K. After stopping the air flow through the furnace, the thermal gradient decreased sligthly. During the rapid cooling in the ICP or ACF the thermal gradient increased again.

The acoustic energy sum increased only slowly during the CNTL- cooling (cooling rate: 0.03 K/min) (Fig. 7). Most of the acoustic emission was measured during ICP- and ACF-cooling (about 75 % of the final value of the acoustic energy sum. The results for cani- ster no. 19 (without Al_2O_3-lining) are shown in Figs. 8 and 9. The cooling process was optimized in this case: the temperature increase due to the stopping of the air flow was avoided. The acoustic energy sum (Fig. 9) showed during the cooling in the furnace (CNTL) only a slight increase with the cooling time (and reached less than 10 % of its final value). Immediately after moving the canister to the ICP the acoustic energy sum increased drastically. The same happened when the canister was moved to the ACF. When the temperature distribution remained constant (from 9000 to 14000 min and after 17000 min) no further increase of the acoustic energy sum was observed. The main acoustic emission (more than 90 % of the acoustic energy sum) occurred during the ICP- and ACF-cooling.

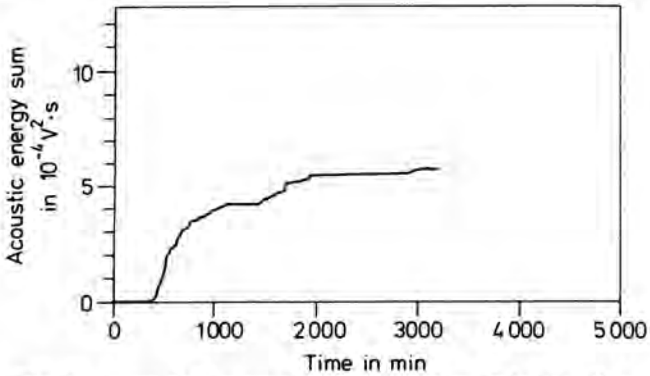

Fig. 5. Acoustic Energy Sum as Function of the Cooling Time for Canister No. 19 During Cooling in the ICP.

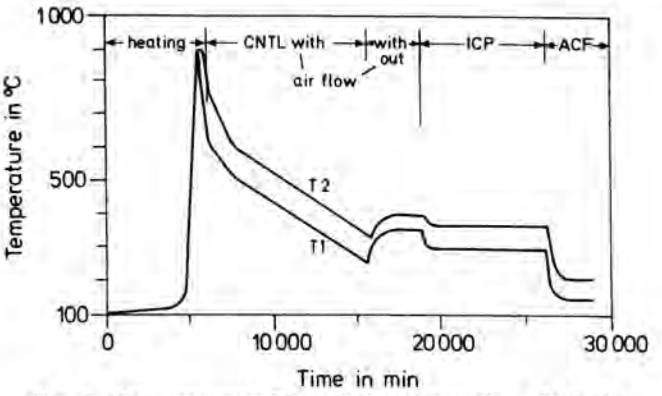

Fig. 6. Temperatures T1 and T2 as Function of Cooling Time for Canister No. 13 During Controlled Cooling.

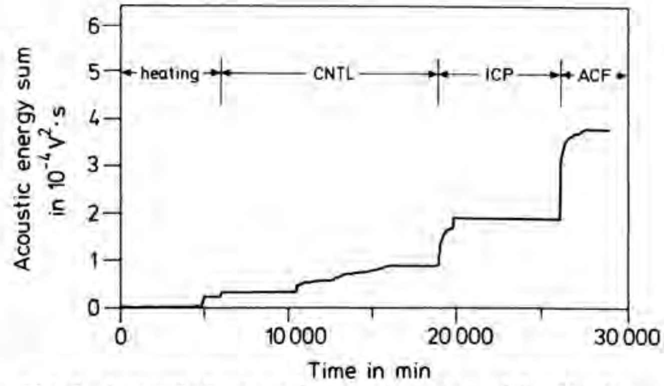

Fig. 7. Acoustic Energy Sum as Function of the Cooling Time for Canister No. 13 During Controlled Cooling.

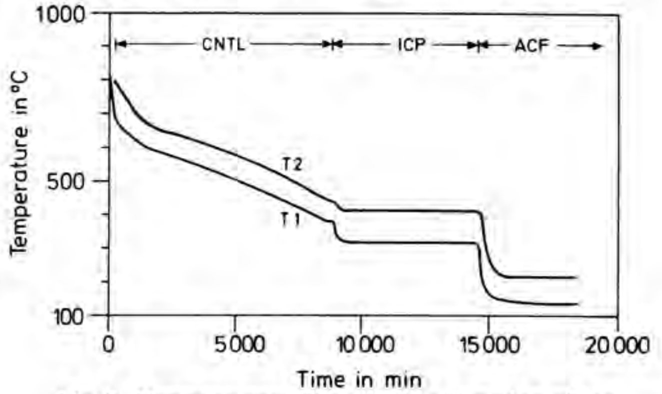

Fig. 8. Temperatures T1 and T2 as Function of the Cooling Time for Canister No. 19 During Controlled Cooling.

DISCUSSION

A direct comparison of the results of the AE measurements of radioactive and nonradioactive glasses was not

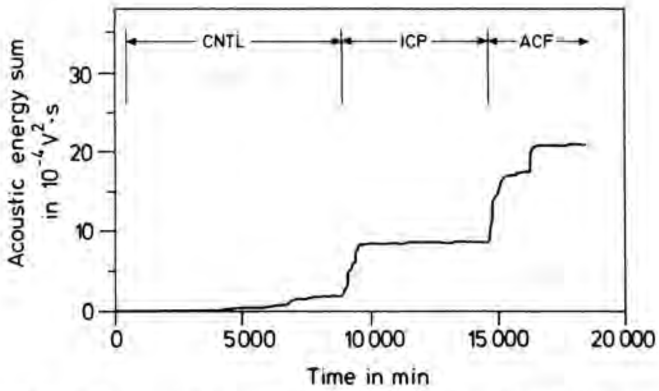

Fig. 9. Acoustic Energy Sum as Function of the Cooling Time for Canister No. 19 During Controlled Cooling.

possible, because of the different equipment and the different cooling conditions. Therefore it was not expected that the acoustic energy sums for both types of glasses were in the same order of magnitude. For a relative comparison the acoustic energy sums of all the experiments are listed in Table I.

The reproducibility was to a high degree affected by the experi- mental conditions. It is not possible to compare results obtained for totally different cooling processes. The reasons for this are the the decoupling of the waveguides and the damping of the acoustic signals due to fracture formation, which depend on the cooling history of the glass blocks. Even slight changes of the conditions caused large deviations. Therfore up to three experiments were performed for each condition, especially for controlled cooling.

Regarding nearly identical cooling conditions it was found that the Al_2O_3-lining reduced the acoustic energy sum significantly for both types of experiments (nonradioactive and radioactive glasses) because stresses due to the shrinking of the canister to the glass were avoided.

The influence of the cooling condition on the fracture formation cannot be discussed without visual inspection of the glass block, which was not possible for the radioactive glass. In the case of the nonradioactive glass it was proven by visual inspection that during controlled cooling with an Al_2O_3-lining a nearly fracture free glass block was obtained. The acoustic energy sum measured in that case was the lowest value obtained for nonradioactive glasses (0.610^{-4} V^2s).

An influence of the different cooling conditions was found with regard to the temperature range in which the strongest AE was measured. With slower cooling rates this temperature range was shifted to lower values because the thermal gradients and thus the stresses in the glass decreased.

In the case of radioactive glasses the temperature gradient can- not be minimized to zero because of the decay heat. Therefore it was not surprising that even in the case of low cooling rates a relatively high acoustic energy sum was measured.

In analogy with results obtained for nonradioactive glasses a strong formation of circumferential and radial cracks can be expected for high cooling rates when cracks are formed immediately after solidification. When the crack formation is shifted to lower temperatures fewer but larger cracks can be expected. In the case of nonradioactive glasses this led to a decrease of the surface area caused by fractures.

CONCLUSION

The acoustic emission analysis is a suitable method for measuring noises caused by the formation of cracks in cooling radioactive HLW glasses. Under nearly identical cooling conditions the measured acoustic energy sum seems

TABLE I

Acoustic Energy Sum in $10^{-4}V^2$s Measured During Cooling of Nonradioactive and Radioactive Glass Containing Canisters

	ACF		ICP		CNTL	
	without	with	without	with	without	with
	A1$_2$0$_3$-lining		A1$_2$0$_3$lining		Al_2O_3-lining	
nonradioactive:						
	-	9.2	5.6	4.3	18.0	3.6
	-		-	9.5	-	4.1
radioactive:canister number:	19	13	19	13	19	13
	3.4	2.5	3.6	1.4	9.2	1.4
	-	-	5.8	-	25.0	3.8
	-	-	5.8	-	21.0	3.7
ACF: air cooling frame, ICP: insulated cooling pot, CNTL: con trolled cooling						

to be correlated with the degree of the damage. This was proven for nonradioactive glasses by visual inspection. For radioactive glasses an analogous behaviour is assumed. Major differences between the cooling of radioactive and nonradioactive glasses are caused by the internal heat production of the radioactive glasses. This leads to a non vanishing radial thermal gradient causing mechanical stresses. Nevertheless the thermal gradient can be reduced by slow cooling. One experimental observation (the decrease of the thermal gradient by reheating, Fig. 6) suggests that a further reduction of the thermal gradient might be possible by an oscillating cooling and heating process in the temperature range of glass solidification.

ACKNOWLEDGEMENT

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