INVESTIGATIONS OF THE COOLING BEHAVIOR OF TWO HIGH ACTIVE WASTE GLASS CANISTERS BY TEMPERATURE DISTRIBUTION MEASUREMENTS

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ABSTRACT

For the first time the temperature distribution in two canisters containing radioactive glass product composed of glass spiked with radioactive Cesium and Strontium, with loadings equivalent to commercial HLLW, have been measured. The temperature distribution was monitored during cooling from the melting point down to the thermal equilibrium under varying cooling conditions. The canisters with 30 cm diameter and 110 cm height contained 60 liters of glass product with a specific heat loading of 17.7 and 20.4 watts/liter respectively. Each canister was instrumented with 30 thermocouples.

Fourteen cooling experiments have been carried out to obtain information as to whether the equilibrium temperature distribution was effected differently by fast or slow cooling of the canisters which causes in turn a different type of fragmentation of the glass product by thermal stresses. The experiments with only two canisters were conducted by remelting the glass product within the canisters using a specially designed furnace.

INTRODUCTION

In the FRG canisters with vitrified high-level liquid waste (HLLW) are going to be disposed of in the Gorleben salt dome or in an equivalent geologic repository. The temperatures at the surface and in the center-line of the canisters shall not exceed 200 and 450 degrees Centigrade respectively. These temperatures depend upon the mass, the heat loading and the thermal conductivity of the glass product and particularly on the diameter of the canisters. The heat loading of the glass product to be disposed of in the salt dome should be a maximum of 17 watts/liter or a total of 2,500 Watts in a canister with 43 cm diameter and 120 cm height and a volume of 150 liters of glass product.

Up to now, information about the temperature distribution in canisters was available by calculations only (1). Computer codes for the calculations are finite element codes: For example, ADINAT and TEMPEST. These calculations had to be checked by experiments with high-level radioactive canisters. These experiments could be planned in 1985 when US-DOE and the FRG-BMFT contracted the fabrication of 32 radioactive heat sources at PNL for a heat and radiation experiment in the Asse salt dome. Two of the canisters have been instrumented with 30 thermocouples in three levels before casting the melted glass product.

EXPERIMENTAL

In March 1987, the two instrumented canisters were prepared and were filled with the radioactive glass product

Fig. 1. View Into Canister #13 with Al203 Insulation. Thermocouples are Guided by Means of Inconel Tubes. In the Vertical Position one can Observe the Arrangement of the Acoustic Emission Wave Guides.

in April 1987. Figure 1 shows an empty canister with the mounted thermocouple array. A group of five double thermocouples, each in three different levels, had been designed so that the temperature could be measured in equal steps of about 3.5 cm starting at the center of the canister wall (canister #19, thermocouple [TC] numbers 1-29, canister #13, thermocouple numbers 30-60). Rectangular to the thermocouples the wave guides for the acoustic emission analysis (AEA) are arranged. The AEA is the topic of the following paper (2). One of the canister's interior was lined with a layer of 3 mm A1203 as a thermal insulation and with the aim to avoid stresses between the glass and the steel wall. The second canister was prepared without such an insulation.

The glass product has been produced in the remotely operated radioactive liquid-fed ceramic melter (RLFCM) at PNL. Since no real HLLW was available, the simulated HLLW had to be spiked with the concentrations of Cs-137 and Sr-90 equivalent to the radiation and heat loading of commercial high-level waste glass products.

The size of the canisters are different from the canisters for the Gorleben salt dome because it was not possible to fill the much larger commercial so-called Cogema canisters, since only limited Cs-137 and Sr-90 was available at PNL. However, the measured temperature distributions give us a basic data set for more exact calculations of the temperature distribution in canisters with other sizes, e.g. the Cogema canisters with a 43 cm diameter and 120 cm height.

TABLE I

Specification of the Canister #13 and #19

Activity (KCi)	Dose (KRad/h)	Loading	Volume (Watts)	Volume (Liters)	Heat/Volume (Watts/ Liters)
Canister No. 13	197.1	174	1,063	60	17.7
Canister No. 19	224.1	183	1,227	60	20.4

Fourteen experiments were carried out with the two canisters; six with canister #13 and eight with canister #19. Each experiment included the remelting of the glass product within the canister in the annealing furnace at 950 Centigrade and cooling down in three different modes:

First Mode, CNTL

After controlled cooling in the annealing furnace, there is a transfer to the insulated cooling pot (ICP). After an adjustment of the thermal equilibrium, there is transfer to the air-cooling frame (ACF) for adjustment of a second temperature equilibrium in air.

Second Mode, ICP

After remelting a direct transfer of the canister from the furnace into the ICP takes place. The ICP was a cylindrical steel container insulated with a layer of 2.54 cm Kao Wool. It is similar to the "cooling station" of the PAMELA vitrification plant at Mol, Belgium. After an adjustment of the thermal equilibrium in the ICP, the canister was transferred to the ACF for achieving temperature equilibrium in air.

Third Mode, ACF:

After the remelting, there is a direct transfer of the canister to the ACF. Cooling down to the thermal equilibrium in air is accomplished.

The aim of these procedures was to get information about the dependence of the canister centerline temperature from the cooling strategy. Slow cooling of the canister should lead to a minimum of cracks inside the glass product. Cracks can act as a barrier for the heat flow leading to an enhanced centerline temperature. Fast cooling in air should lead to more cracks inside the glass product and, therefore, a higher probable centerline temperature.

Figure 2 gives a schematic overview regarding all three types of cooling experiments and the test facility. To reduce the vertical heat flow from the canisters, they have been provided with insulation at their top and bottom.

RESULTS

The principle differences in the cooling behavior of nonradioactive versus radioactive (self-heating) canisters is displayed in Fig. 3. It shows the temperature dependence of the middle thermocouple level versus time during cooling of a nonradioactive canister versus the highly radioactive

TABLE II

Schedule of Experiments

Experiment Number	Canister Number	Experiment Mode	Starting Date	Ending Date
1	19	ACF	42787	50187
2	13	ICP	429	504
3	19	ACF	504	507
4	13	ICP	505	515
5	19	CNTL	507	515
6	19	CNTL	525	601
7	19	ICP	601	609
8	13	CNTL	602	618
9	19	CNTL	612	627
10	13	CNTL	622	709
11	19	ICP	706	710
12	13	CNTL	709	723
13	19	CNTL	716	730
14	13	ACF	723	727

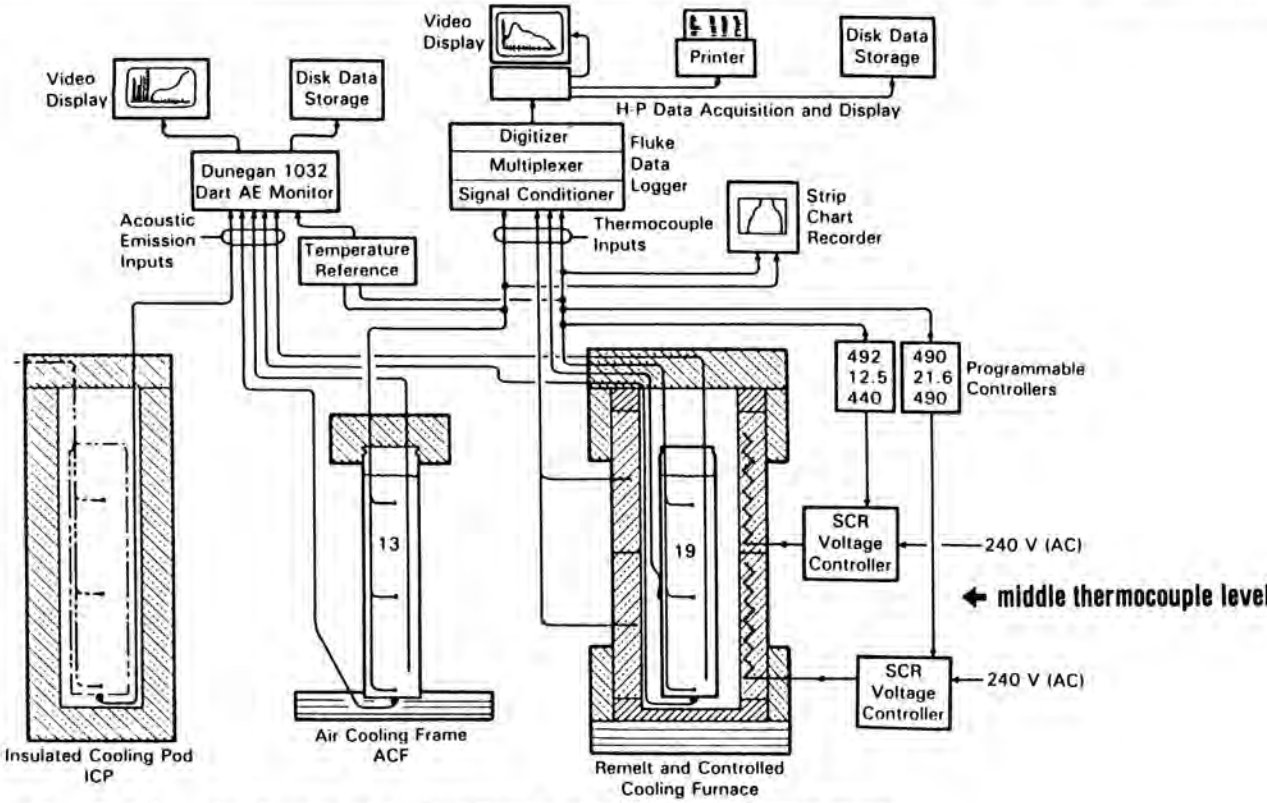

Fig. 2. Schematic Diagram of the Test Facility and the Mode of Experiments.

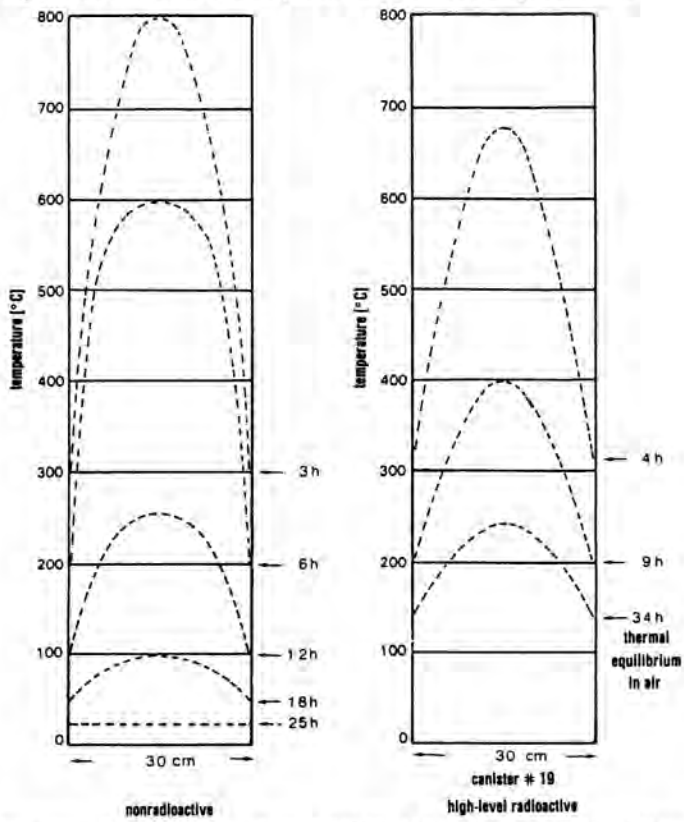

Fig. 3. Schematic Diagram of the Temperature Distribution Versus Cooling Time in Air in a Nonradioactive Canister and the High-Level Radioactive Canister #19.

canister #19. Apart from heat loading by radioactivity of canister #19, the experimental conditions have been similar; the size of the canisters, cooling in air, and no A1203-lining. After three and four hours, respectively, the temperature has reached 300 Centigrade on the inside of the canister wall and 800 and 675 Centigrade, respectively, at the centerline of the canister. Thus, there are temperature differences of 500 Centigrade in the nonradioactive glass block and only 375 Centigrade in the radioactive glass block.

However, after 25 hours, the temperature in the nonradioactive canister has reached room temperature and is constant over the diameter. On the other hand, only after 34 hours has the thermal equilibrium been reached in canister #19. At this time, the temperature is 140 Centigrade at the inside of the wall and 240 Centigrade at the centerline. Thus a T of 100 Centigrade remains constant at this level, if cooling conditions do not change.

The following temperatures were taken from the middle thermocouple level because a vertical heat flow in the upper and lower thermocouple level could not be excluded. Figure 4 shows the numbers of thermocouples in this middle level in both canisters.

It is possible to produce nonradioactive glass product in canisters with no or few cracks at temperature below the transition temperature of 500 Centigrade if the temperature differences are kept below 20 Centigrade (4). Because temperature differences are always larger in air-cooled canisters, Fig. 3 explains the why the glass product then came to have many cracks. The surface increase generated by cracks is about 25 times larger than the surface of a cylindrical monolithic glass block in the case of the nonradioactive canister. It must be expected that in the radioactive canister #19, where temperature differences of 100 Centigrade remain for very long times, the surface increase will be nearly the same.

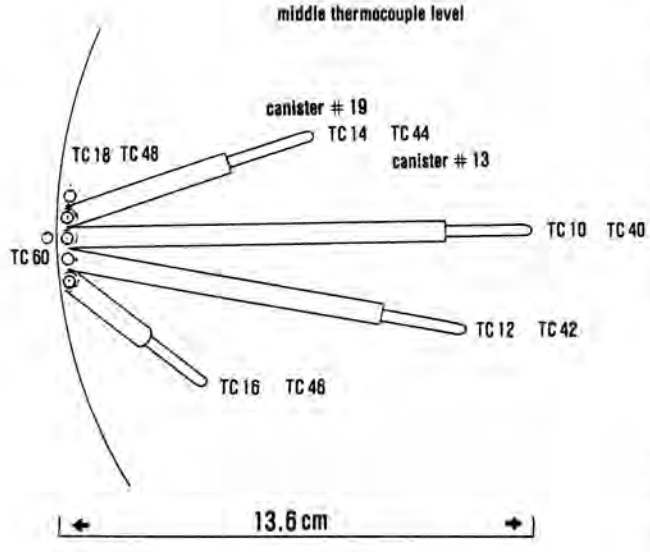

Fig. 4. Schematic Diagram of the Thermocouple Numbers in the Middle Thermocouple Level of Canister #19 and Canister #13.

Figures 5-7 give three examples of the 14 experiments performed, which include that of the CNTL, ICP and ACF, for canister #13. They are representative of all the experiments. In the case of the controlled cooling in the annealing furnace, it took too long to reach the thermal equilibrium. For this reason, the cooling of the canisters was accelerated in two ways. Firstly, lifting the canister from the furnace until the temperature inside the canister wall had dropped near to the transition temperature. Secondly, air blowing near the bottom of the furnace. At temperatures above the transition temperature of about 500 Centigrade, the glass product is still plastic. Cracks can therefore not be generated. Below 500 Centigrade the cooling rate was kept at 6 Centigrade per hour. Experiments with nonradioactive canisters have shown that at this rate, it was possible to get a glass product with few cracks.

The active canisters show persistent temperature differences between centerline and wall of 60-70 Centigrade for canister #13 and about 100 Centigrade for canister #19. This temperature distribution is fairly independent from the cooling mode in the CNTL, ICP and ACF cases. The temperature difference changes only when the canisters are moved from a higher thermal equilibrium to a lower one. Then the temperature difference rose from 70 to 110 Centigrade in canister #13 for a short transition period, during which the wall temperature fell more rapidly than the centerline temperature.

Figures 8-11 show plots of the temperature distributions from the centerline to the inner surface of the wall and to the surface of the canister after thermal equilibrium had been reached. In canister #19, a number of thermocouples failed in subsequent runs. They are marked with open symbols. From the inner surface of the wall to the surface of the canister, the temperature drop is 30 Centigrade in the case of canister #13 and only 10 Centigrade in the case of canister #19. This difference is due to the insulating A1203 layer at the inner surface of canister #13. The thickness of the canister wall is 8 millimeters and consists of stainless steel.

CONCLUSIONS

In conclusion, the authors believe that this unique opportunity to compare high-level waste canisters and their cooling behavior under varying cooling conditions with similarly prepared nonradioactive canisters gives verification of the results of computer codes already existing in USA and FRG. This should be possible for the calculation of different sized HLW canisters and different cooling modes.

Since monolithic glass blocks cannot be produced without cracks, due to remaining temperature differences which must in contrast to nonradioactive simulates lead to

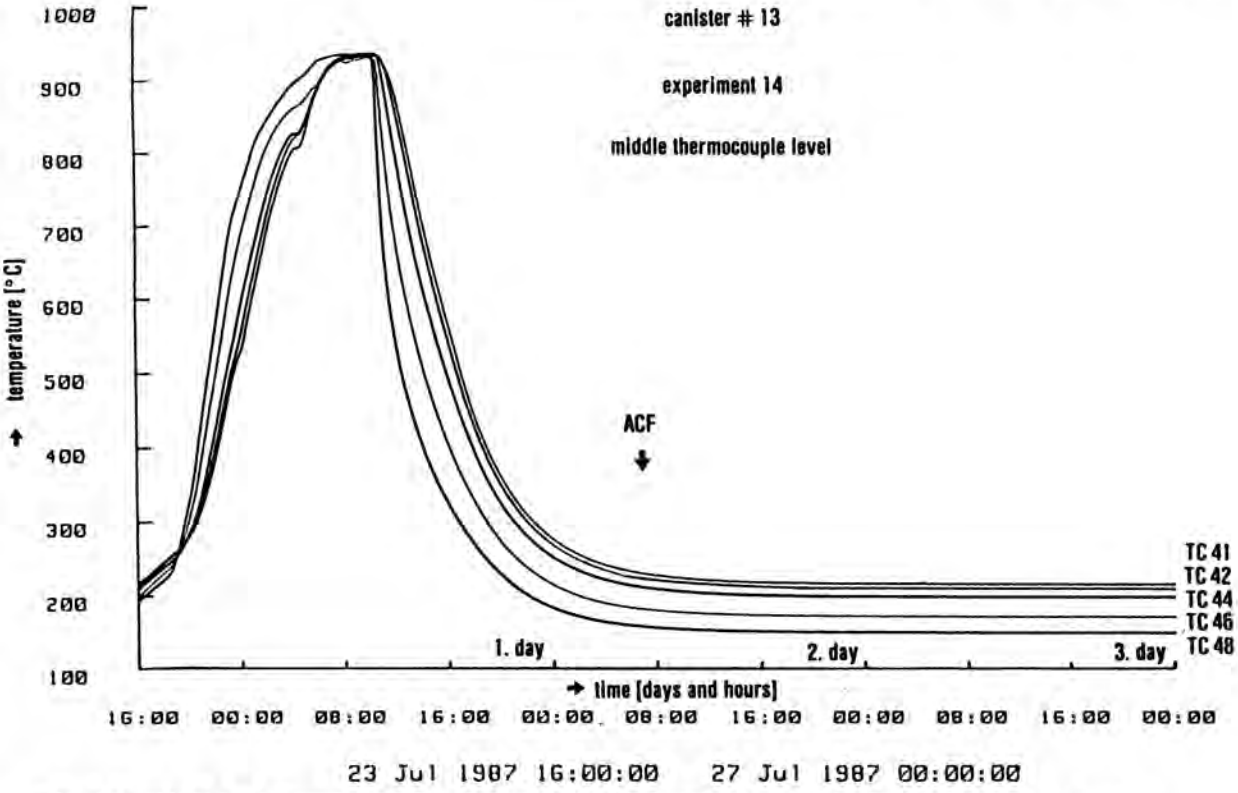

Fig. 5. Canister #13, Temperature in the Middle Thermocouple Level Versus Cooling Time in Air.

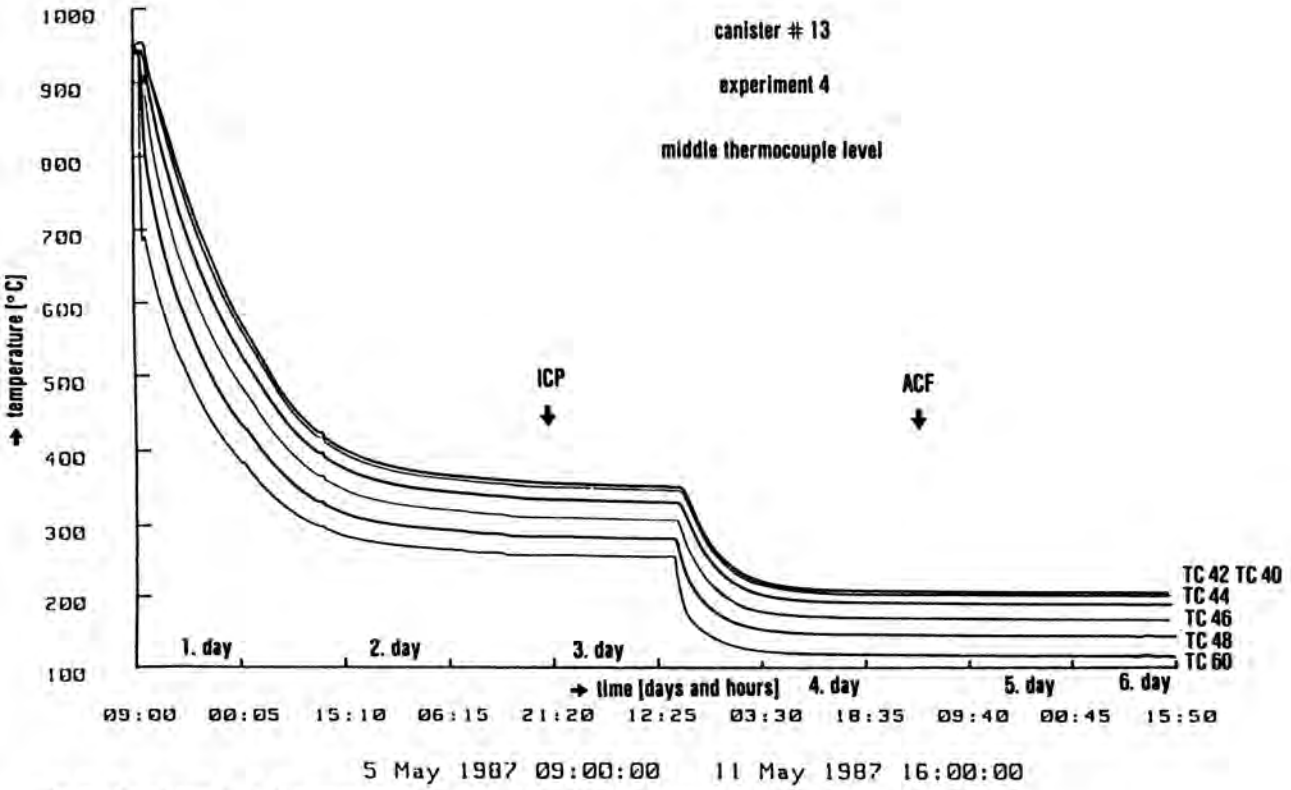

Fig. 6. Canister #13, Temperature in the Middle Thermocouple Level Versus Cooling Time in the ICP and in Air.

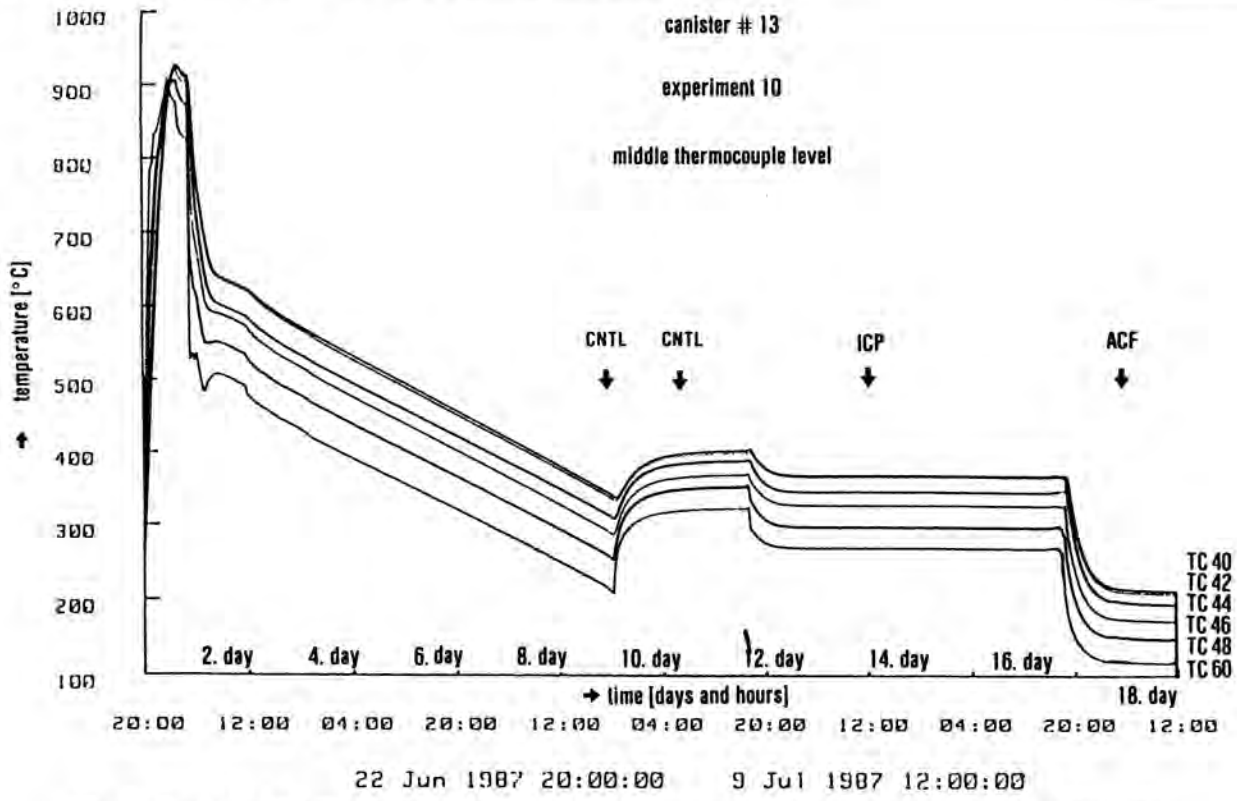

Fig. 7. Canister #13, Temperature in the Middle Thermocouple Level Versus Cooling Time in the Annealing Furnace, the ICP and in Air.

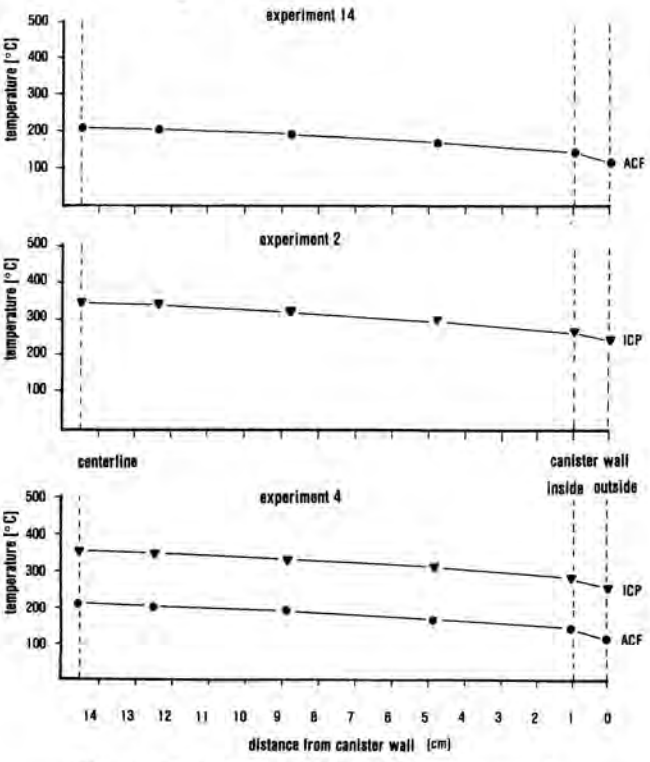

Fig. 8. Canister #13, Middle Thermocouple Level, Experiment #14, 2 and 4: Temperature Distribution Between Centerline and Canister Wall at Thermal Equilibrium.

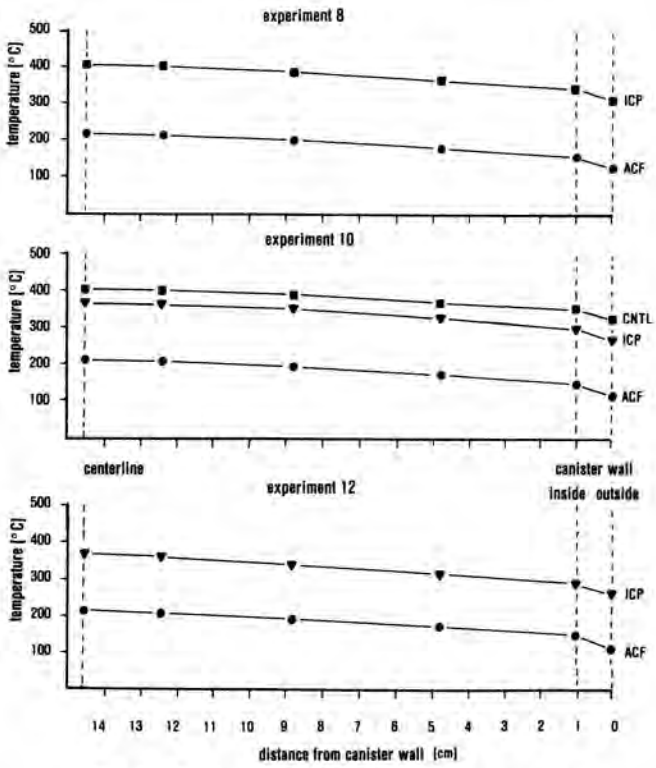

Fig. 9. Canister #13, Middle Thermocouple Level, Experiment # 8, 10 and 12: Temperature Distribution Between Centerline and Canister Wall at Thermal Equilibrium.

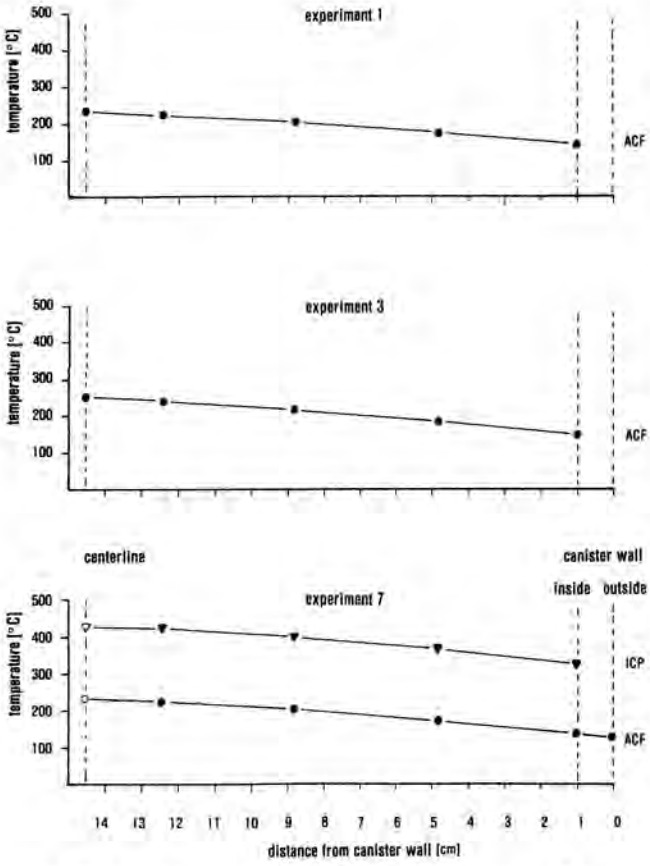

Fig. 10. Canister # 19, Middle Thermocouple Level, Experiment # 1, 3 and 7: Temperature Distribution Between Centerline and Canister Wall at Thermal Equilibrium.

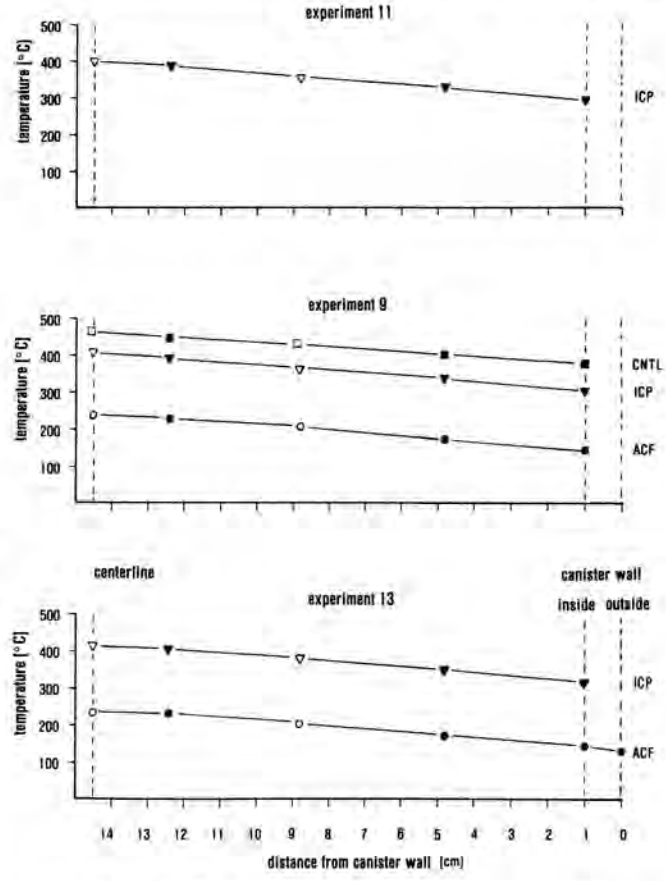

Fig. 11. Canister # 19, Middle Thermocouple Level, Experiment # 11, 9 and 13: Temperature Distribution Between Centerline and Canister Wall at Thermal Equilibrium.

cracks, the temperature distribution was not so well predictable.

During the experiments we also registered the acoustic emissions. We were intent to draw conclusions from the comparison between active and similar nonradioactive experiments. First results of this comparison are presented in the paper by W. Storch (2).

Ongoing research with nonradioactive simulation, computer code development and further comparison of data from these two canisters with Cesium and Strontium activity will enable us to predict conditions for commercially produced HLW canisters with much more accuracy than hitherto.

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