# GAMMA IRRADIATION EXPERIMENTS IN NATURAL AND SYNTHETIC HALITE

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## ABSTRACT

A description is given of the methods, instruments and samples used in gamma-irradiation experiments on salt samples performed in the storage pool of the High Flux Reactor (HFR) at Petten, The Netherlands.

Pre- and post- irradiation analyses and mechanical handling of the samples were planned and performed besides the irradiation experiments themselves and, prior to the irradiation experiments, the instruments needed were designed and constructed.

The developed irradiation instrumentation essentially consists of blocks both impermeable and isothermal which contain eight sample assemblages each. These blocks allow irradiation to take place at constant and homogeneous temperature. They were placed in racks among spent fuel elements in the storage pool of the HFR. The spent fuel elements were used as radiation source. Different combinations of spent fuel elements produce different dose rates due to the different decay time of the fission products contained in them.

The pressure conditions of the experiments were fixed independently for each of the eight sample assemblages contained in a block, and therefore samples irradiated simultaneously could be subject to different pressure conditions. The racks with the spent fuel elements are here called Gamma Irradiation Facility (GIF). Two different types of experiments named GIF A and GIF B were performed. GIF A experiments were performed at high and variable dose rates and non-enhanced pressure. GIF B experiments were performed at low and (quasi) constant dose rate and on two non-pressurized and six pressurized sample assemblages simultaneously. In this way each of the following factors could be varied independently: total dose, dose rate, pressure, and chemical composition and microstructure of the sample.

## 1. INTRODUCTION

The theories on the radiation damage of rock salt which underlie the models on which nuclear waste repository safety previsions are based, needed revision and countercheck. The purpose of our research was testing some of these theories against the experimental results of irradiation of samples in conditions as near as possible to those of a repository.

Our research was complicated by the existence of two different groups of variables. One group of variables are the physical conditions of the irradiation. The other group of variables are the different sample characteristics.

This required many irradiation experiments to independently vary the intrinsic irradiation parameters, and many samples in each irradiation experiment to vary the sample characteristics independently.

Two different types of experiments have been carried out in the Gamma Irradiation Facility (GIF), GIF A and GIF B experiments.

In chapter 2 we describe the GIF characteristics and the requirements of our experiments. In the subchapters 2.1 and 2.2 the instruments and procedures developed in order to use the GIF to fulfill the experimental requirements are described. The general methodological description of chapter 2 is followed by two chapters (3 and 4) which respectively describe the methodological particularities of GIF A and GIF B types of experiments independently. At the end of this article a chapter (5) of results summarizes the amount of samples of each type which have been irradiated under each set of conditions. The results of the analyses of the samples are not described in this article but can be found in the data report and are worked out and interpreted in different articles of this volume [De las Cuevas and Miralles, 1995; Donker and García Celma 1995 a and 1995 b; García Celma and Donker, 1995 and García Celma et al. 1995 ( articles. Nos 15, 17,18, 21 and 20 respectively, in this volume )]



## 2. GENERAL CHARACTERISTICS OF GIF EXPERIMENTS

The above mentioned Gamma Irradiation Facility (GIF) consists of spent fuel elements in compact storage racks (where they are placed to cool) in the pool of the High Flux Reactor (HFR) at Petten. Figure 1 gives a view of the GIF.

When the spent fuel elements are fresh, (just retrieved from the reactor) their emitted gamma dose rate and heat output are high. The intensity of both emissions decreases with time as shown in Fig. 2 a and 2 b. If the spent fuel elements are old, i.e. have already spent some months in the cooling pool, their emitted gamma dose and temperature are low and quasi-constant.

Our irradiation experiments required three main additional procedural conditions from the irradiation facility which had to be taken into account when building the instrumentation:

a) possibility to fix and control the temperature of irradiation (at 100°C)
b) possibility to irradiate simultaneously different samples to be able to guarantee that the samples had been subject to the same dose rate and temperature, and
c) possibility to identically irradiate identical samples but under different pressure conditions.

### 2.1. Instrumentation developed

The **HE**ated **I**rradiation of **SA**lt instrumentation (HEISA) was designed and constructed to simultaneously irradiate many samples at a constant temperature. A HEISA consists of an external metallic prism containing an inner solid metal block and connected to a computer, a gas system and a temperature regulator. The metallic prism is placed in the racks amongst the fuel elements. The gap between the metallic block and the prism is filled with a gas. Different gases can be used depending on their heat conduction characteristics and on whether cooling or protection against cooling is needed to maintain a constant temperature inside the HEISA. In the metallic block holes have been machined out to place in tight fitting eight sample holders, and slots have been machined as well to fit the heating elements. The heating elements are connected

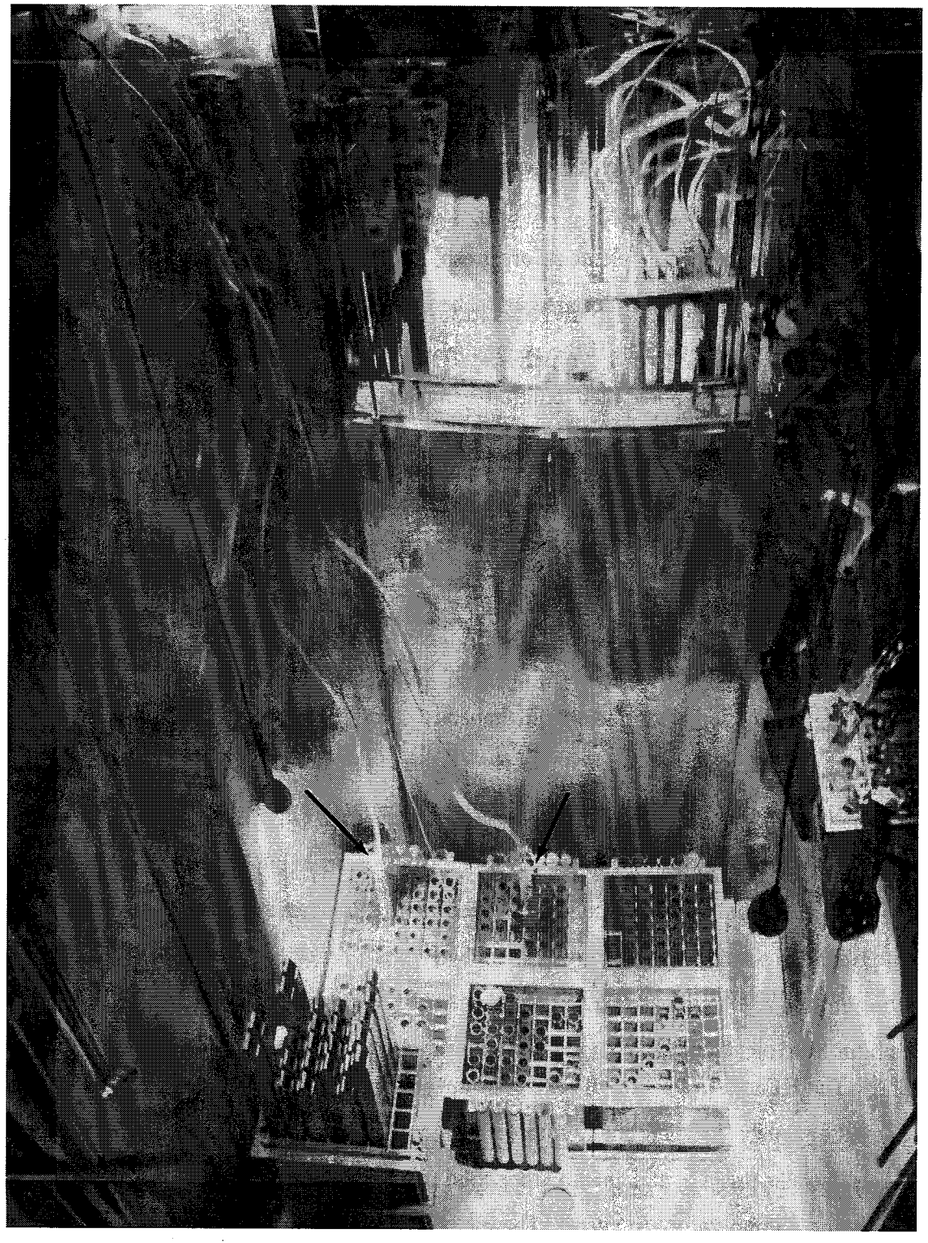

Figure 1 a. *A view of the compact storage racks with the spent fuel elements at the bottom of the pool where the Gamma Irradiation Facility (GIF) experiments are carried out. Arrows show the HEISA'S where experiments GIF A and GIF B take place.*



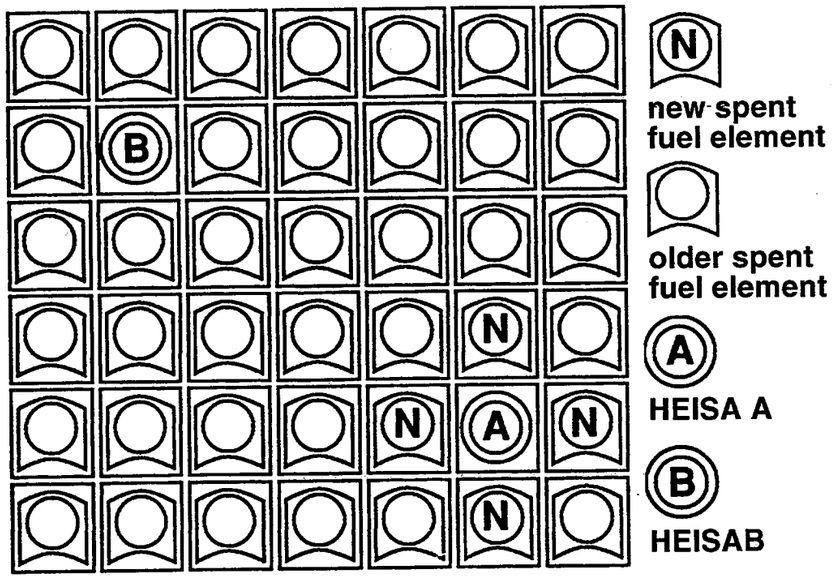


Figure 1 b: *Storage rack with two HEISAS containing the GIF experiments.*

to a proportional band temperature regulator (outside the pool). There is a minimum of two thermocouples in each HEISA, one produces the reading for the temperature controller and the readings of the other are directly registered by a computer (see Fig.3).

A sample holder consists of a stainless steel cylinder with only part of a basis. The holders are represented in figure 4. A plate of stainless steel is set loose within the holder and rests on the incomplete basis. Within the stainless steel cylinder a second stainless steel cylinder closely fitting the first one is introduced. The third cylinder is a gold jacket containing the samples. Gold jackets were chosen to reduce holder corrosion by the chlorine emitted by the NaCl during irradiation. There have been earlier designs which were used to carry out some of the earlier experiments and which did not use gold jackets by silver wrappings. The old set up of the samples within the various cylinders is described in [García Celma and Van Wees, 1992]

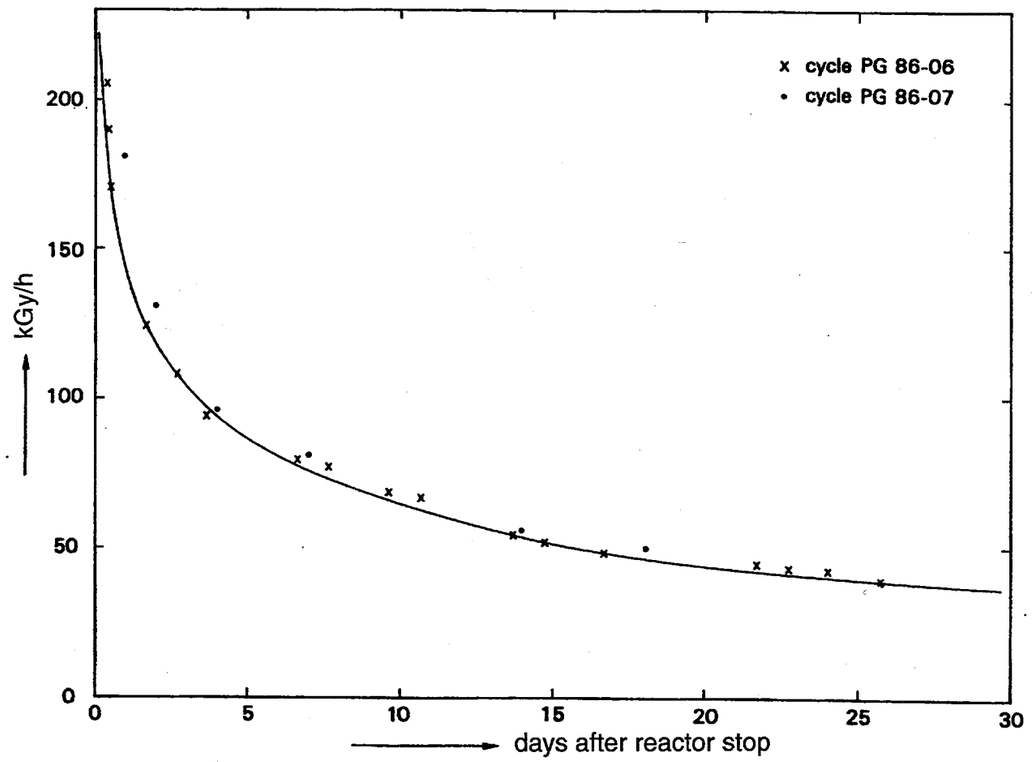


Figure 2 a.: *Dose rate in the target line of a GIF.*

The top of the stainless steel cylinder can be closed in two ways. One way of closing the top of the stainless steel cylinder is a lid in contact with the top of the gold jacket and welded to a gas bellows. The top cover of the gas bellows is, in its turn, welded to the top of the stainless steel cylinder. The gas bellows can be filled in with gas at a high pressure. This holder exerts a pressure of about 200 bar on the inner (gold) cylinder (Fig. 4) [García Celma and Van Wees, 1992, and García Celma, 1992]. This is a *pressurizing sample holder*.

The other way of closing the samples holder is by a lid fixed to the holder by a bayonet fitting and having a spring in the inside with an end piece which contacts the top of the gold jacket keeping it into place. This spring is assumed not to subject the samples to additional pressure this



type of holder is called a *non-pressurizing sample holder*.

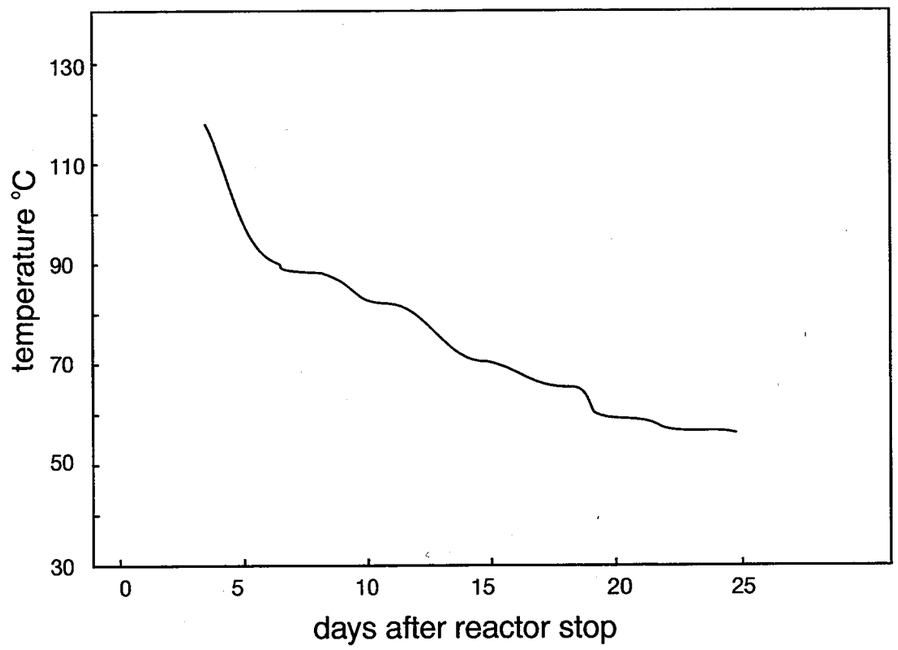


Figure 2 b: *Temperature evolution as registered by a thermocouple in a sample container in the irradiation facility in absence of the HEISA instrumentation.*

There are two types of gold jackets, one of about 52 mm length with an inner diameter of 24.26 mm and one of about 12 mm length and with an inner diameter of 23.76 mm. The holders are assembled with the samples in the following manner. Inside the long jacket a long sample is introduced, on top of it the shortest gold jacket is introduced in the longest. Then a tablet shaped sample is introduced in the shortest jacket and a gold lid is placed on top of it. The lid and the two top ends of the jackets are then welded together. In this way the samples are gastightly packed during irradiation. The gold jackets used in pressurizing sample holders contain as well a ceramic plate of the same diameter as the samples on the top of each of the two samples. This is done to avoid sample fracturing, since the pressure is transmitted by the lower surface of the bellows which is smaller than that of the sample (see Figs. 3 and 4).

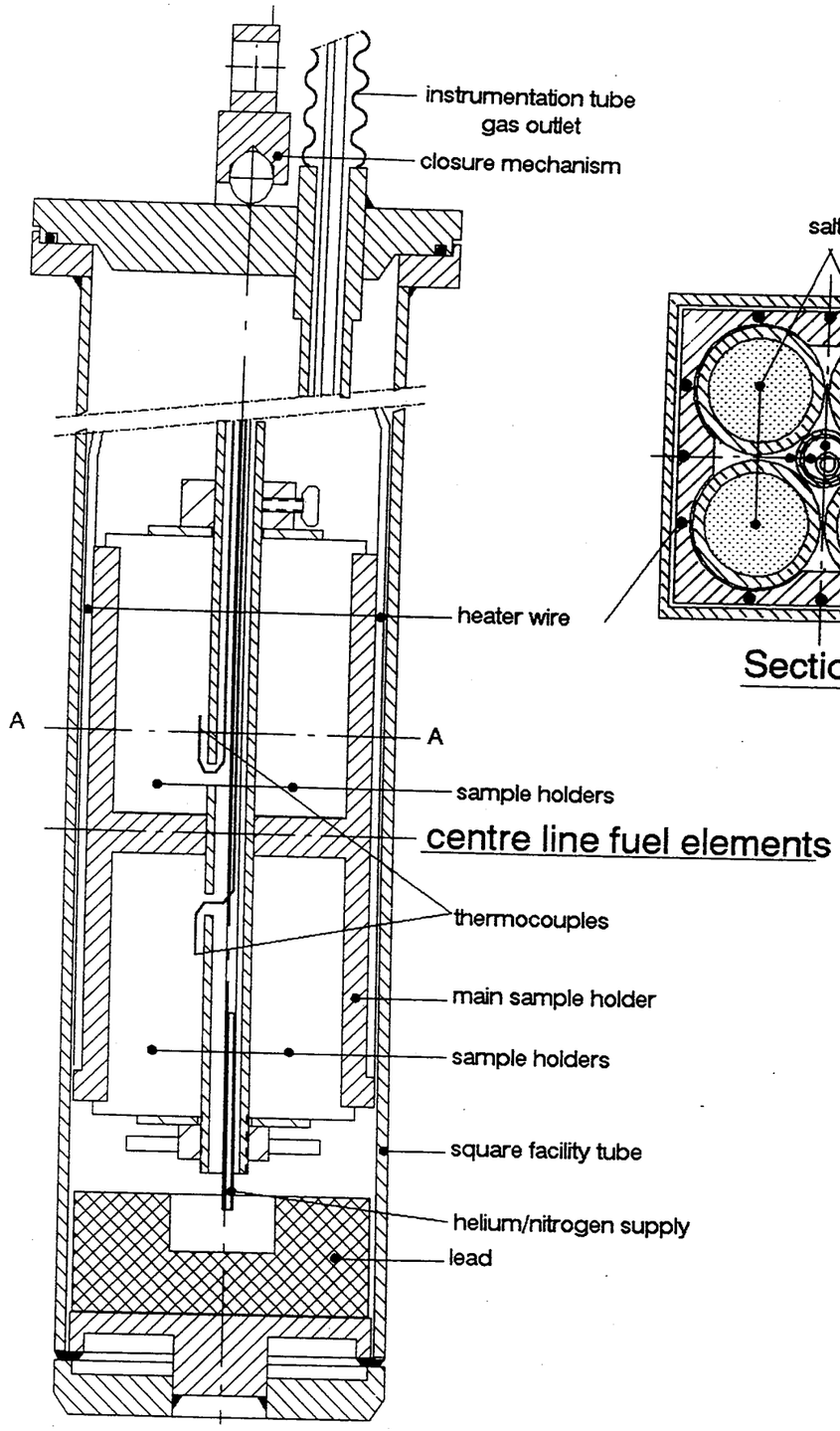


Figure 3: ***HEated Irradiation of SAlt (HEISA) instrumentation.***



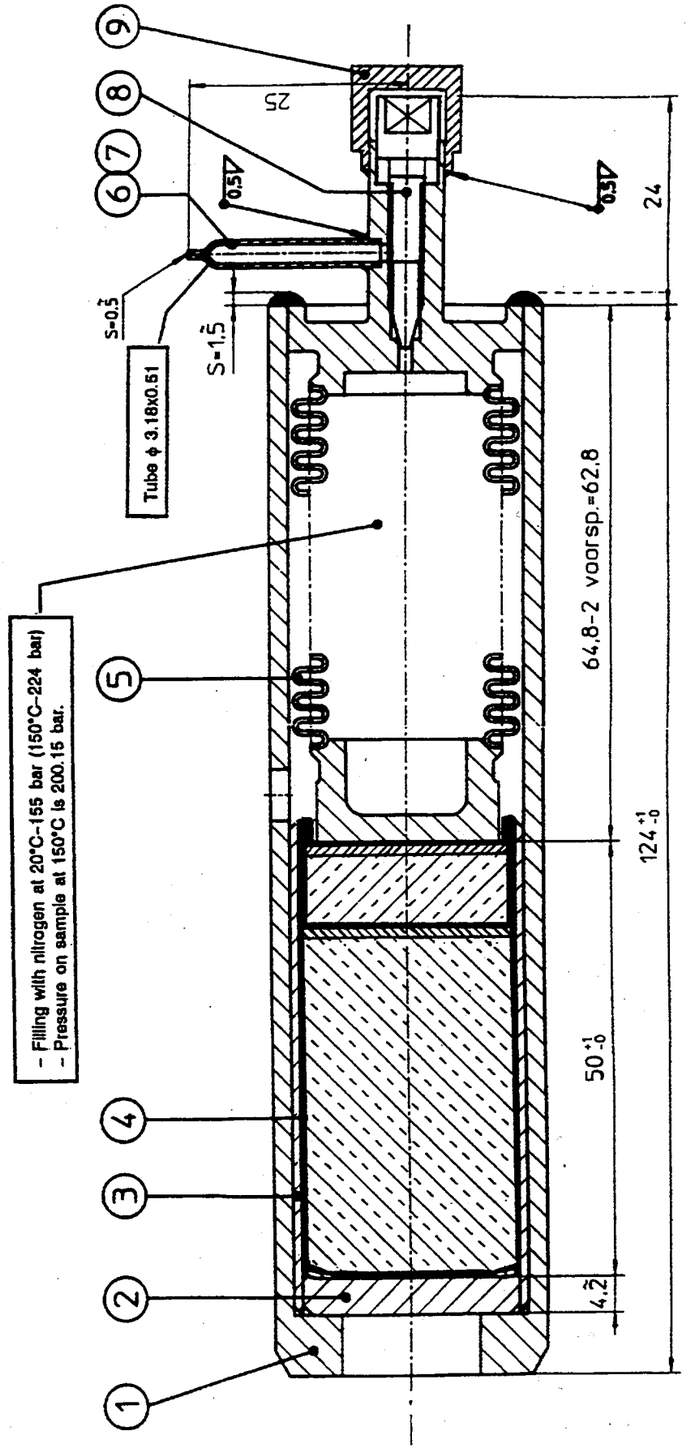


Figure 4: *Design of the pressurizing sample holders.*

## 2.2. Procedures

We will distinguish two types of procedures: handling and analyses. The procedures which we group here under the name handlings imply preparation of the samples for the experiments or for observation, such as sample machining and are described in 2.2.1. The analyses refer only to the observation or measurement, i.e. stored energy analyses, and are described in 2.2.2.

### 2.2.1. Pre- and post- irradiation handling

The samples are kept in dry drawers or in the "salt room". Both places are dried by a constant flow of compressed air. Handlings took place either in the dry drawers (precision cuts) or in the salt room (machining etc...). Some thin sections of each sample type were made in order to study their general pre-irradiation microstructure. The thin sections were conventional optical microscope preparations.

Prior to irradiation the samples had to be machined to fit the holders into two different sizes, i.e. : tablets and cylinders. The tablets have a diameter of 23.76 mm and a height of 10 mm, while the cylinders have a diameter of 24.26 mm and a height of 40 mm. Before introducing the polycrystalline samples in the gold jackets and acetate replica of the position of the grain boundaries in their top and bottom surfaces was taken. The samples were also weighted and their dimensions measured. To some polycrystalline samples known quantities of brine were added prior to their introduction in the jackets. The gold jackets were sealed by cold welding and placed inside the stainless steel holders and the whole made ready for irradiation. The procedures are described in detail in [García Celma, 1991 and 1992; and García Celma and Van Wees, 1992].

After irradiation the sample dimensions were measured, the samples weighted and new acetate replicas made. Then the samples were cut in two, one piece was sent to the Barcelona University, while the other piece was kept at ECN. With this piece of the sample first a thin section was made, and then pieces were selected for stored energy analyses. The thin section was used for observation in a conventional optical microscope and for Image Analysis measurements.



### 2.2.2. Pre- and post-irradiation analyses

Most of the planned analyses change the properties of the studied material (e.g. stored energy analysis anneal crystal defects out and dry out the samples) or require cutting the sample. Therefore pre-irradiation analyses are in fact analyses performed on non-enhanced- irradiated samples of the same batch as those being irradiated. Non-enhanced-irradiated samples are subject to the same analysis as irradiated samples in order to know which has been the effect of the irradiation on the sample properties. Not all the analysis were performed with all the samples, and the analytic work was shared between the ECN department at the Utrecht University and the Barcelona University [Mönig et al., 1990; De las Cuevas and Miralles, 1995 and Donker and García Celma, 1995 (this volume art. Nos..15 and 17)]. The pre- and post-irradiation analysis of each sample are always performed at the same laboratory. The different analytic techniques are grouped here below and the information which is obtained from each is summarized. Detailed information on detection limits etc.. can be found elsewhere in this volume [De las Cuevas and Miralles, 1995 and Donker et al., 1995 ( this volume art. Nos.15 and 19 )].

*Light Absorption* (LA) analyses were performed on irradiated salt because some of the crystal defects produced by irradiation absorb light of a characteristic wave length. LA analyses consist of registering the transmitted spectra of (originally) white light after traversing the salt samples and analysing which wave lengths have been absorbed. The type of defect, its relative frequency and even the defect size can be inferred from the spectra characteristics. These analyses were performed at the Barcelona University, and the results are reported in [García Celma et al, 1992 and De las Cuevas and Miralles, 1995 (this volume, art. nr.15)].

*Differential Thermal Analysis* (DTA), consists of heating a sample within a holder included in an isothermal block and measuring the temperature reached by the sample holder and the temperature reached by a reference holder also included in the isothermal block. The two measurements are then compared with each other. If the sample contains energy which is liberated at a given temperature, the temperature of the sample becomes higher than that of the reference. It also can happen that the sample becomes cooler than the reference. These differences are registered. This is what constitutes the "first run" of a measurement. After the first run the sample is allowed to cool down to room temperature (without opening its holder) and the "second run" can start. The second run consists of carrying out again the same heating process as the first run.

The difference between the first and the second run gives the energy balance of the irreversible processes. The irreversible process which interests us most is the anneal of the crystal defects produced by irradiation. These measurements were performed at the Thermodynamisch Centrum Utrecht, and the results can be found in [García Celma and Donker, 1994; Donker and García Celma 1995 a and 1995 b, ( this volume , art. nr.17 and 18)] .

*ThermoGravimetric analyses* (TG), measure the weight changes undergone by a sample that is being heated in much the same way as the DTA does. The sample temperature and weight are registered along with time. Since the heating rate is constant, weight changes with time correspond to weight changes with temperature. Interpretation of these analyses, in our case, gives mostly information on some endothermal (heat consuming) processes, such as vaporization of the brine included in natural samples. The measurements register the weight loss associated with the release of the gases from the samples and the temperature at which this takes place. In our case these analyses give valuable information on the brine contained at grain boundaries and in fluid inclusions, and on the dehydration processes of accessory minerals. These analyses have been performed at the Barcelona University and the results can be found in [De las Cuevas and Teixidor, 1992 and De las Cuevas et al.,1995, ( this volume, art. nr. 10 and 12)].

*Chemical Analyses* produce information on the type and amounts of impurities present in the samples. Combining them with microscopical sample observation it is possible to know whether the halite contains many lattice impurities or not. Intracrystalline impurities influence the radiation damage development in the NaCl crystal while the intercrystalline accessory minerals can only influence intercrystalline processes. Chemical analyses also measure the nature and amount of gases contained before irradiation and heating and those contained after irradiation [De las Cuevas et al., 1995, (this volume, art. nr. 12)]. Mainly the amount of hydrogen and of chlorine produced during irradiation are very important. Another relevant chemical analysis result is the determination of the amount of Na-metal produced by irradiation. This determination is done indirectly, by dissolving the salt samples in water and measuring the amount of hydrogen which is obtained ($2\,Na + 2\,H_2O = 2\,OHNa + H_2$). These analyses have been performed at the Barcelona University, and also at Braunschweig by the GSF with other samples. The results can be found in [De las Cuevas et al., 1995, and Mönig et al., 1995 (this volume, art. Nos. 12 and 16 )].

*Microscopic analysis*, consisted of conventional optical microscope observation of the thin



sections made from irradiated and non-enhanced-irradiated samples. Microscopic analyses help interpretation of other analyses, e.g. the presence of hydrated minerals can justify endothermal signals in the DTA record of a sample. A particular type of microscopic analysis is the microstructural analysis which gives information on the mechanisms by which mono- and poly-crystals adapt to stressing situations, in our case to radiation damage. The microstructural analyses of irradiated salt samples are eased by the fact many microstructures can be coloured by irradiation [García Celma and Donker, 1994]. Damaged parts of crystals, when observed by transparency under the microscope, tend to be yellow, brown, red and blue, while non-enhanced damaged parts are colourless. The amounts of colourless versus coloured material can be measured using a conventional image analysis system [García Celma et al., 1988].

## 3. PARTICULARITIES OF GIF A EXPERIMENTS

It has been postulated that, by continuous irradiation with gamma rays, the energy stored in radiation damage defects in natural rock salt will not reach a saturation level through gradual anneal of the defects but will steadily grow until a very high concentration of defects is reached at which a spontaneous (and explosive) back reaction would take place [Den Hartog et al., 1990]. In the GIF A experiments we were looking for a possible saturation of radiation damage in natural rock salts at concentration values of defects inferior to those needed for the postulated back reaction. Therefore these experiments required a single type of natural sample to be progressively irradiated up to very high doses.

A couple of pure single crystals were as well irradiated in this facility to produce samples for experiments in which we intend to determine the size and size distribution of radiation damage defects (Na-colloids in particular) by Small Angle Neutron Scattering (SANS). However, the SANS method and/or the installation used were inadequate for this purpose. Anyway the pure samples could be used to check the radiation damage theory on other points.

### 3.1. Samples used in GIF A experiments

Most samples used in GIF A experiments were of a rather pure natural rock salt, but also

pure synthetic single crystals have been used.

The *pure single crystals,* also poor in $OH^-$ contents, were purchased from *Harshaw* Ltd. They are frequently used as standard crystals in radiation damage experiments. They were all machined into tablets, and labelled "SANS + number" when used in GIF A.

Harshaw crystals, as received, contain a stored energy of between less that 1 J/g and up to 14 J/g [García Celma and Donker, 1994]. This energy disappears by anneal at 500°C in an open canister in an argon atmosphere during one hour. Schutjens [1991] showed that the random distribution of free dislocation lines found in the Harshaw crystals is substituted by an organized structure during anneal. The annealed Harshaw crystals do not give any endo- or exothermal signal. In the first experiments (GIF A1 and GIF B1) non-enhanced-annealed Harshaw crystals were used. Annealed crystals have been used in the second set of experiments of the GIF A (GIF A2).

The *natural samples* used in GIF A were samples of the *Speisesalz* from the 800 meter depth level of the Asse Mine, Remlingen, Germany. This is a very pure (99,9 % halite) rock salt which is also very dry (0.05 % weight of brine), has a granular structure with grain diameters varying between 3 and 10 mm. The composition and characteristics of this salt and salt formation are described in [Jockwer, 1981; Peach, 1991; Spiers et al., 1986; Urai et al., 1986; García Celma et al. 1988 and Gies et al.,1994]. The Speisesalz samples used in this experiment were all tablets and their code is " TSp + a number".

### 3.2. Procedures of GIF A experiments

Tablets of Sp-800 were machined, introduced in golden jackets and sealed. Also SANS samples were prepared. Thirty nine Sp-800 samples and sometimes a SANS sample were included in eight non-pressurizing sample holders. The non-pressurizing sample holders were placed into a HEISA. And the HEISA was placed in a GIF between freshly spent fuel elements (GIF A).

The temperature was fixed at 100°C. The dose rate was the highest which could be reached in a GIF, but decayed quickly as show by the dosimetric measurements plotted in Fig.2



a. At about 28 days after irradiation start a sample was taken out of the facility and substituted by a non-enhanced-irradiated sample, and another dosimetry was performed. See Fig. 5. In each experimental cycle a dosimetry was performed before placing the samples in the radiation field and some dosimetries more were performed during the following 28 days.

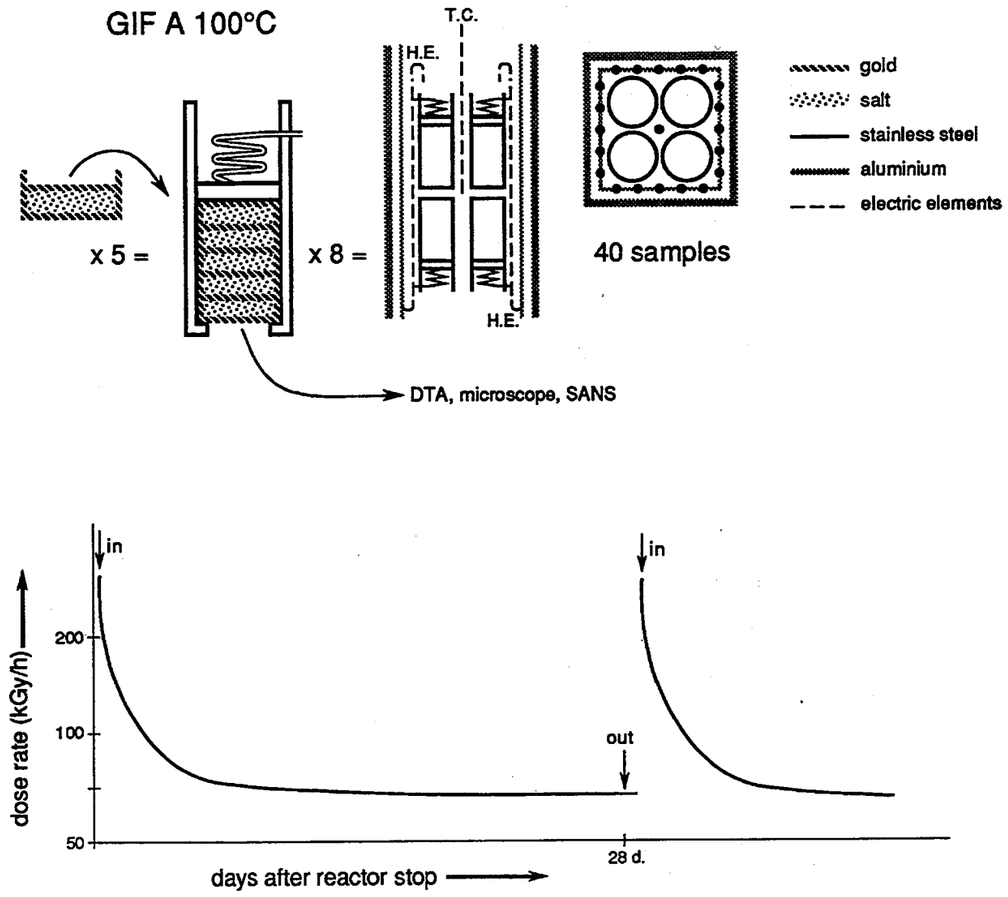


Figure 5: *Schematic representation of the procedure in GIF A. In the schematic representation of the dose rate evolution* in *and* out *indicate when samples are replaced.*

After each reactor cycle (about 28 days) the new freshly spent fuel elements were placed in the GIF A, a new dosimetry was performed, and the HEISA containing the new sample and the thirty nine samples which had already been irradiated during one month were replaced in GIF A. Repeating this procedure after each reactor cycle gives a somewhat variable dose rate traject for the samples, an example of which is given in Fig 6.

This procedure was repeated each month, from December 1989 until May 1994. Each month the samples which had not been retrieved received an additional dose of about 40 to 30 MGy. The experiment was restarted once, but anyway, at the end of the second experiment some samples which had not yet been retrieved and had received a total dose of 1223 MGy, and there existed at least one sample for each 40 to 30 MGy increment from 40 up to 1223 MGy.

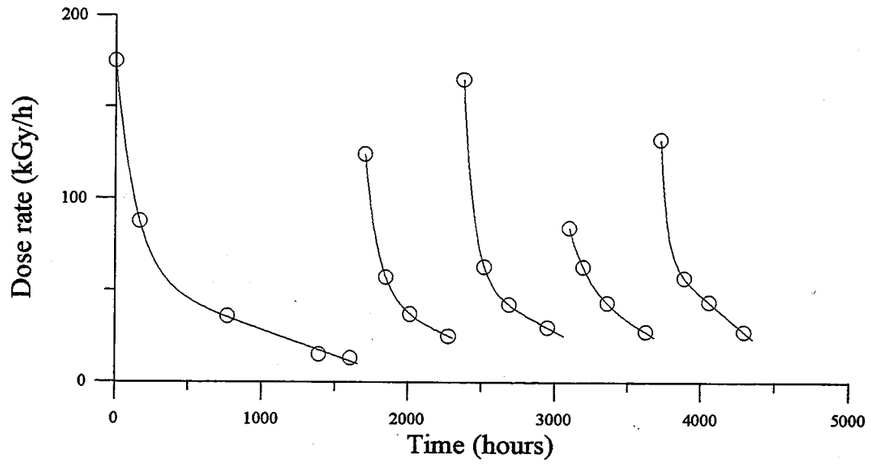


Figure 6: *An example of the dose rate trajectory in GIF A.*

After retrieval, the samples were cut into two pieces and a thin section was made. One piece of the sample remained at Utrecht and from it we performed DTA analysis, and the other piece was sent to Barcelona where they performed LA, and TG analyses and measured the production of hydrogen by solution in water in order to determine the amount of damage in terms of amount of Na-metal. This is reported in [De las Cuevas and Miralles, 1995, ( art. nr.15, this volume].



## 4. PARTICULARITIES OF GIF B EXPERIMENTS

GIF B experiments were planned to produce information on the effect of pressure and of impurities on radiation damage development at conditions as near as possible to that of a radioactive waste repository. Therefore various types of samples had to be irradiated up to the same total doses for a whole set of total doses and the effect of (low) dose rates and of (enhanced) pressure had to be studied as well for different total dose (time) increments.

Dose rate had to be as low and as constant as possible in each experiment to approach radioactive waste conditions. The effect of low dose rate in natural samples had to be analyzed (figure 6). This was done by performing different series of experiments at different dose rates. These series were GIF B1, GIF B2 and GIF B3; each series consisted of irradiating up to different total doses 16 samples at a time.

Here we will give some information on the samples, first on the planned differences between them, and the reason for using these samples, and then they will be summarily described in 4.1. The procedures and a résumé of the reasons to adopt them are given in 4.2.

### 4.1. Samples used in GIF B experiments

In the GIF B experiments, to compare the effect of identical irradiation in samples differing only in "wetness', 'purity', mono- or poly-crystallinity or degree of deformation samples differing from each other in only one of these factors have been simultaneously irradiated. We irradiated together pure dry and undeformed single crystals of NaCl, pure polycrystals and natural rock salts (neither pure, dry, monocrystalline nor undeformed).

#### 4.1.1. Pure single crystals

Pure single crystals, purchased from Harshaw Ltd., have been described in 3.1. They were all irradiated as tablets. In the first series of GIF B experiments (GIF B1) they were not annealed prior to irradiation. In the following set of GIF B (GIF B2) experiments they were annealed prior

to irradiation, and in the third set (GIF B3) a combination of annealed and non-enhanced-annealed Harshaw crystals was irradiated. The Harshaw crystals are labelled "H + a number".

### 4.1.2. Pure polycrystals

Since natural rock salt is always impure, pure polycrystals had to be produced. The starting material from which all the pure polycrystals were produced was the same, namely NaCl powder of "pro-analysis" quality as produced by Merck to which 0.2 % weight bi-distilled water was added. Two types of pure polycrystals were produced differing in their microstructures, the pressed powder and the synthetic salt polycrystals.

The *pressed powder* samples were produced by cold pressing up to 1.7 kbar, maintaining the pressure during 3 min, and slowly taking the pressure away. The thus obtained cylinders were further machined into either cylinders (" PP + number" ) or tablets (" TPP + number" ). Sometimes, brine was added to the finished samples or tablets. The brine was produced using NaCl powder pro-analysis and bi-distilled water.

The individual crystals of the pressed powder samples display brittle deformation structures and have a mean diameter of 60 μm. The poria between crystals in the Pressed powder samples are irregular and therefore grain boundaries are not shared by two grains but consist only of the limits of individual crystals. Fig 7. These samples yield an exothermal stored energy of 4.2 J/g before irradiation [García Celma, 1992; García Celma and Van Wees, 1992; García Celma , 1991; García Celma et al., 1993].

The *synthetic rock salt* was produced by first pressing the starting material cold at 1 kbar during 8 hours, and then annealing it during 30 days at 500 bar and 150°C. During the anneal the salt crystals develop real grain boundaries shared by two grains while the brittle structures of the crystals disappear. The new grain boundaries are straight and meet each other at triple points making angles of 120 degrees. This shows that they migrated until their exterior surfaces reached a very low free energy configuration. These are typical recrystallized granular structures called "foam-textures" by evident morphogenetic reasons. See fig 8. These samples are also machined after production to fit the holders and are labelled " SS + number" or "TSS + number". To some



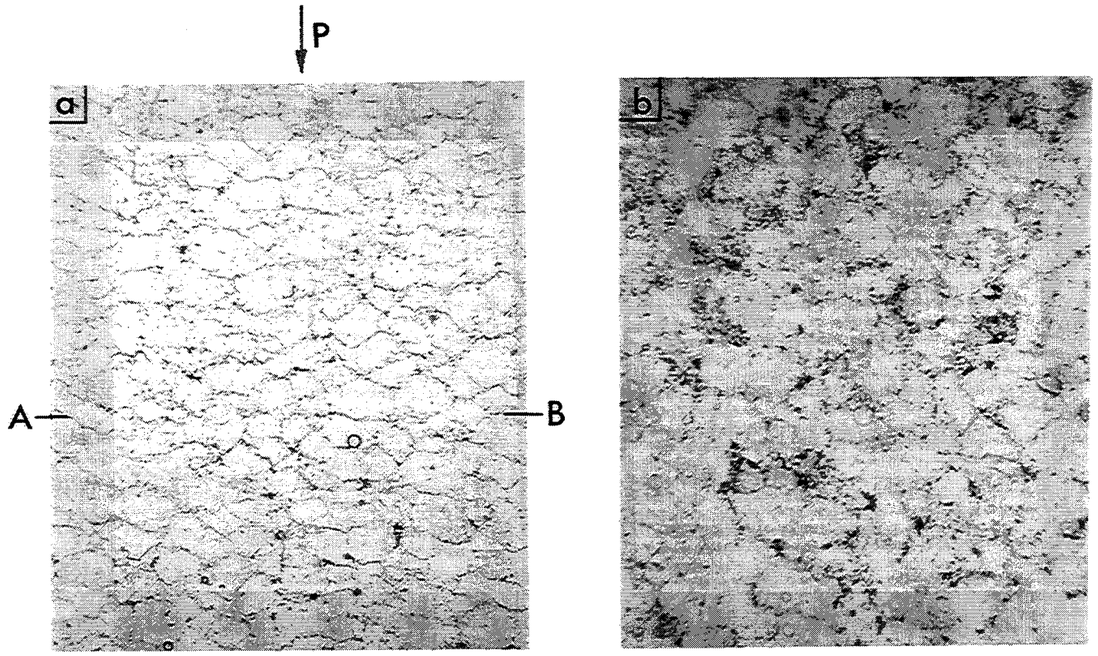


Figure 7: *Microstructural aspect of Pressed Powder samples in two perpendicular sections a): section parallel to the direction of the pressure (P) used to produce it. b): section perpendicular to the direction of the pressure (A-B in 7a). Mag. 5x.*

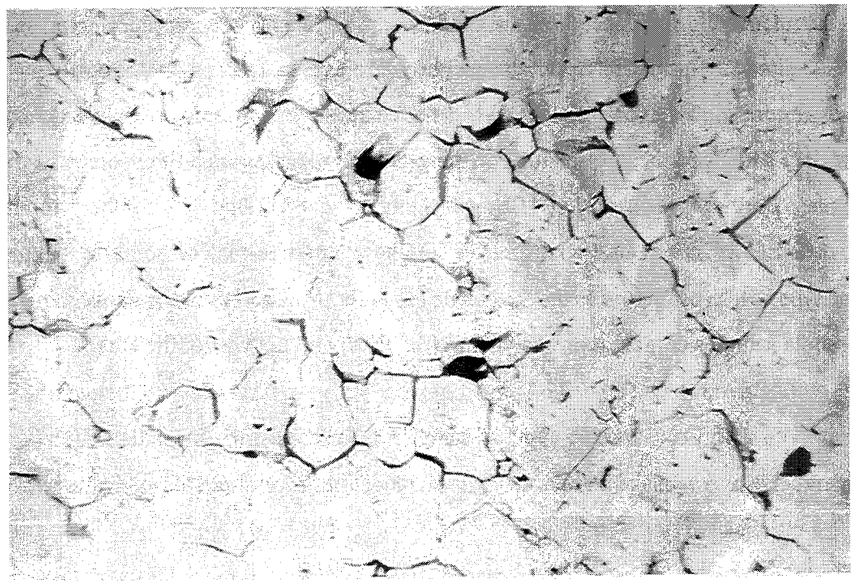

Figure 8 : *Microscopical aspect of synthetic rock salt. Observe the polygonal shape of the grains. Mag. 5x.*

of them pure brine was added before the irradiation. SS samples yield a stored energy of 4.7 J/g before irradiation [García Celma, 1991 and 1992; García Celma and Van Wees, 1992].

### 4.1.3. Natural rock salts

The used natural samples proceeded either from the Asse mine, in Remlingen, Germany, or from the Potasas del Llobregat mine, in Cardona, Spain. The formations and the characteristics of the samples are described in Gies et al. [1994] and De las Cuevas et al., [1995, (this volume, art. nr.10)] respectively. Here we will only name some of their characteristics.

From the Asse mine we used the *Speisesalz* from the 800 m depth level due to its purity, and also to the fact that so many studies [Jockwer, 1981; Spiers et al., 1986; Urai et al., 1986; García Celma et al., 1988; Peach, 1991; Gies et al., 1994] have been performed with these rather homogeneous salt that it has almost become a standard for pure natural rock salts (it was also used for GIF A experiments, see chapter 3.1.). These samples are labelled " Sp-800 + number " when long (40 mm) and " TSp800 + number" when in the form of tablets (10 mm high).

The samples called Borehole samples were as well taken from the Asse mine, from the boreholes drilled to perform the HAW-test field experiment, where in principle they would have been irradiated producing a picture of the gradual progression of the damage in the borehole samples. *Borehole polyhalitic* ( "Bhp + a number" or " TBhp + a number" for tablets) and *Borehole anhydritic* (" Bha + a number" or " TBha + a number"). The borehole samples have the same type of microstructure as the Speisesalz but differ from it in their accessory mineral contents. Borehole samples contain more polyhalite (the Bhp) and anhydrite (the Bha).

We also used another type of samples from the Asse mine, the *Polyhalitic* samples, although they were only used as tablets, they contain huge amounts of polyhalite and are labelled PS + a number.

The Asse samples have been recrystallized a couple of times during their geological history, therefore they present a granular microstructure from which the original sedimentary structures have been whipped out.



The samples from the *Potasas del Llobregat* mine, contrary to the Asse samples have not suffered complete recrystallization but only developed overgrowths during diagenesis. They therefore display many relics of the originally precipitated crystals and although they can be considered just as pure as those of Asse in their bulk, at the level of the sample size used in the experiments they are rather heterogeneous. They contain mostly anhydrite and some clay as accessory mineral. They are extensively described in [De las Cuevas et al., 1995 (this volume, art. nr.10)],

## 4.2. Procedures in GIF B experiments

In GIF B experiments samples are irradiated at a low and as constant as possible dose rate, at a constant temperature of 100ºC, and up to different integrated doses. The (quasi)constant and low dose rate can be reached by placing the samples amongst old spent fuel elements in the GIF B (see Figs. 1 b and 9).

Through repeating experiments at the same dose rate but during a different length of time different integrated doses are obtained, and since a HEISA can contain eight sample holders the possibility exists of simultaneously irradiating different types of samples. Figure 7 schematizes the way GIF B experiments are operated. By combining pressurizing with non-pressurizing sample holders identical samples were irradiated simultaneously which only differed in the pressure at which they were subjected during irradiation.

### 4.2.1. Pre-irradiation procedures

Long samples and tablets had to be prepared to fit the holders. There were also two types of holders, pressurizing and non-pressurizing. Jackets were of gold but in the first experimental set (GIF B1) the samples were wrapped in silver foil and introduced in aluminium holders.

The artificial samples had to be produced and the process developed to produce them has been summarily described in 4.1.2. The samples were measured and weighted before irradiation. From the long samples an acetate peeling was made before packing them for irradiation. In a single irradiation experiment up to three identical samples could be irradiated which differed in

the amount of brine added to them prior to irradiation [García Celma and Van Wees, 1992].

### 4.2.2. Irradiation conditions

First two sets of experiments have been carried out, GIF B1 at 15 kGy/h and GIF B2 at 4 kGy/h. The wished dose rates were obtained by using different combinations of old spent fuel elements. Depending on the time elapsed since they were used in the reactor they emit a higher or lower dose rate (see Fig.9). Different irradiation duration produced samples irradiated up to different total doses. Irradiation took place at a constant temperature of 100°C.

In each experiment two identical samples and two identical tablets were irradiated which differed in the pressure applied to them during irradiation. There were six pressurizing sample holders and two non-pressurizing samples holders in each experiment of GIF B1 and GIF B2. Dosemetries were regularly performed.

In the GIF B3 experiment a smaller quantity of samples and sample types was irradiated at 15 kGy/h up to about the same total doses as in the previous two experiments, and also at 100°C and in pressurized and non-enhanced-pressurized sample holders.

The data of the irradiations are given in a separated data report (in prep) for each sample and irradiation configuration.

### 4.2.3. Post-irradiation procedures

Post-irradiation procedures consisted of first of all obtaining an acetate replica of the position of the grain boundaries in the circular surfaces of the long samples immediately after irradiation. Once the replicas had been obtained the samples were cut in two halfs one of which was sent to Barcelona and the other was kept by us. With the half of the samples which we kept, first a thin section was made, then a piece was cut from which the darkest pieces were hand picked for stored energy analysis by DTA. With the piece sent to Barcelona where $H_2$, and $Cl_2$ determinations and LA and TG analyses were performed.



All samples have always been kept dry and dark and at room temperature.

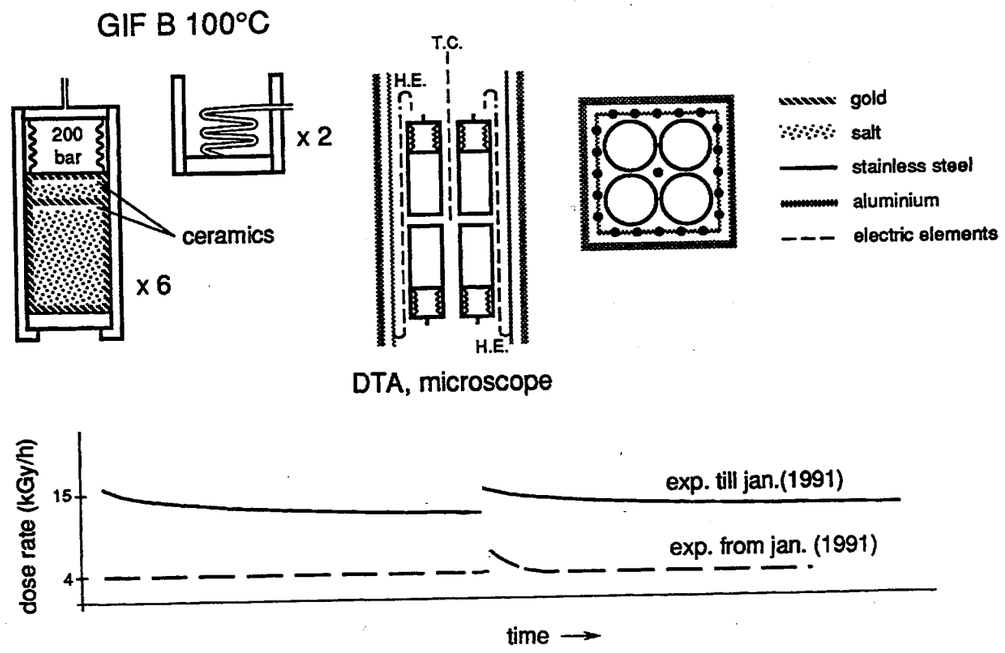


Figure 9: *Schematic experimental set-up of GIF B experiments. The different dose rates are reached by using different combinations of old spent fuel elements. Time scale is arbitrary. See as well Fig. 1 b.*

## 5. RESULTS

All obtained samples were irradiated at a constant temperature of 100°C.

In GIF A1 experiment a set of samples wrapped in silver was irradiated up to a total dose of 567 MGy. In GIF A2 experiment the samples were enclosed in welded golden jackets which

was an improvement in the design regarding eventual leaking of gases or brine. Another difference between GIF A1 and GIF A2 was that the Harshaw crystals were annealed before irradiation in GIF A2 while they were not annealed prior to GIF A1 experiments. Moreover, the quality of the fuel elements had been changed, and as a consequence the highest dose rate was lower in GIF A2 than in GIF A1 experiments. Under these conditions, in GIF A2, we were able to irradiate some samples up to 1223 MGy up to a total of 39 Asse Speisesalz samples which received total doses of between 1223 and 40 MGy in increments of about 40 MGy to 30 MGy, depending on the reactor cycle. All sample composition conditions of irradiation and results of the analysis are given in a separated data report (in prep).

In GIF B, we could perform three sets of experiments and irradiated a total of 400 samples. GIF B1 (160 samples) experiment was performed at a dose rate of approximately 15 kGy/h (decreasing from 20 to 10 kGy/h), 10 different total doses were reached varying from 0.02 to 45 MGy. In this set the Harshaw crystals had not been annealed prior to irradiation.

A second set, GIF B2 (160 samples) was performed with a dose rate of 4 kGy/h, up to 10 different total doses varying from 0.022 to 44 MGy. The sample compositions were the same as in the previous experiment with a few exceptions. The Harshaw samples were annealed prior to irradiation, and all samples were included in gold jackets.

The third set of experiments, GIF B3 (80 samples) was performed at a dose rate of 15 kGy/h, up to 10 different dose rates , also between 0.022 and 41 MGy. In this experiment only PLL, Sp800 and Harshaw samples were irradiated. The Harshaw crystals were annealed and non-enhanced-annealed prior to irradiation and both sorts were irradiated up to the same total doses pressurized and non-enhanced-pressurized.

We wished to perform experiments at yet lower dose rates and for a variety of relevant temperatures and aimed at it in the HAW-field planning, but we had to settle for laboratory experiments which are necessarily shorter ( human interference difficults to maintain the factors constant) and we also had to choose to perform the experiments at a constant temperature, that of maximum damage production reported to be 100°C [Den Hartog, 1988]. In spite of these disappointments the agreed extension of the laboratory experiments has produced a unique and very important data base which has substantially modified the theory in most of the points which



constituted questions for the HAW-field experiment on radiation damage.

## ACKNOWLEDGMENTS

We remember with pleasure our collaboration with Prof. Dr. C. Spiers, Dr. C. Peach, and Dr. R. C. M. W. Franssen, at the Utrecht University. Ir. L. Vons has been an endless source of handy ideas and ways out. P. Snip and A.H. de Vries took care of many of our experiments. To all of them our thanks.

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