

”Radiation damage in alkali halides”

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Abstract

After the Institutions in Germany, responsible for the development of safe conditions in the different parts of the nuclear energy fuel cycle, concluded that the effects of radiation damage did not affect the safety of the application of nuclear power, the German research in the area of radiation damage in NaCl connected with the problems of nuclear waste was terminated during the past decade.

In Germany, rock salt has always been the primary option for storage of HLW. In contrast with the German situation, we note that in the Netherlands, when considering the possible host rocks for storage of high level nuclear waste, the attention is not focused exclusively at NaCl formations. Also storage in clay has been investigated (as a part of the CORA research program) during the past years. Despite this ambivalent approach in the Netherlands:

- (i) the research on radiation damage in NaCl has been maintained at a modest level with limited financial support and this has led to many new discoveries and ideas, which might help us to deal with the problems associated with radiation damage in NaCl and
- (ii) there are strong recommendations made by the CORA Committee of the Dutch Government to continue the research on radiation damage in rock salt in the Netherlands, because it is felt that more knowledge should be acquired and suitable storage concepts, taking into account the effects of radiation damage, need to be developed along with effective monitoring plans for storage sites allowing retrievable storage of HLW.

In this brief report, we will show that the decision made in Germany to stop their very limited research efforts on radiation damage in NaCl (and other possible hosts for storage of HLW) and abandon this research area completely, has been made on the basis of incomplete and incorrect information. We are convinced that with cooperative efforts made by research groups in a number of European countries we could have made significantly more progress in solving the problems associated with radiation damage in rock salt during the past decade. Unfortunately, at several occasions representatives of important German institutes responsible for the development of safe solutions for the management of HLW have been active in Germany and on the European level to block research efforts in the area of radiation damage in rock salt.

I. Introduction

This report describes the developments after the workshop held in Bad Bentheim (Germany) in 1994, where Dutch and German scientists presented their recent results on radiation damage in NaCl. The workshop was organized by the Dutch Committee OPLA (Committee for on shore storage of nuclear waste). The OPLA committee decided that it would be good if Dutch and German scientists would discuss in a small group the controversies on the subject of radiation damage in NaCl. This meeting was remarkable, because we in Groningen have sensed at many occasions that the enthusiasm at the German side to discuss with us the subject of radiation damage was minimal. The workshop was attended by scientists from the institutions BGR-Hannover-Germany, GSF-Braunschweig-Germany, Stoller Ing. Technik-Dresden-Germany, University of Paderborn-Germany, ECN-Petten-NL, COVRA-Borsele-NL, University of Utrecht-NL, University of Groningen-NL.

The chairman of the Bad Bentheim workshop, Dr. Hageman, who was in 1994 also the chairman of OPLA, concluded in his closing statement of the workshop that (i) the problems of radiation damage in NaCl should be considered seriously and (ii) the best way to do this was to set up collaborations between the groups participating in the workshop. Unfortunately, these words never materialized although there have been serious attempts from my side to start collaborations between our group and German scientists. Although some German scientists told me that they were very much in favor of a collaboration, the attempts were blocked by high ranking officials (who unfortunately did not participate in the workshop). The conclusion of these persons was that they wanted to close the files on radiation damage in NaCl and I regret that almost one decade after the workshop in Bad Bentheim German scientists have not contributed to the progress in this research area.

In our opinion the activities of the scientists in Germany on the subject radiation damage in NaCl have been very limited, defensive and they were aimed exclusively at formulating replies to critical questions from the numerous groups in Germany that are opposed to nuclear power and storage of nuclear waste in rock salt. This is in sharp contrast with the conclusions drawn at the end of the workshop held in Bad Bentheim. After this workshop, scientists from the different German institutions responsible for the development of safe solutions for high level nuclear waste have not generated new knowledge on radiation damage in rock salt, the most wanted host rock for storage of HLW in Germany. As an alibi these institutes hid themselves behind the incorrect statement that the problems relevant to storage of HLW associated with radiation damage in NaCl have been solved.

In this report we will show that this position cannot be maintained any longer and that the best solution for the future is to recognize the existing problems and set up close collaborations between the groups in the Netherlands and Germany that could contribute to solutions. In principle

there is a sufficient reservoir of scientists available, but the most important problem appears to be that high ranking persons, who decided that Germany does not want to have anything to do with radiation damage in rock salt, should make a move to allow and even stimulate those scientists in Germany, who are able to contribute to the solution of this still existing problem, to carry out the necessary research.

II. The differences

Of course there have been many differences of opinion between us, here in the Netherlands, c.q. Groningen and Germany over the past decades. We don't intend to present here all these differences. The objective of this section is to show only the most important differences, i.e. those playing a central role in the ultimate discussions about the consequences of radiation damage on the storage of HLW in rock salt.

II,1 The issue of the applicability of the theoretical models to calculate the damage levels under storage conditions

A very common argument used by our German counterpart is that the theoretical models can be used to calculate the damage percentages in natural rock salt under storage conditions, even though we have carried out experiments on samples, which had been irradiated at extremely high dose rates (which are a factor of $\sim 10^4$ higher than under storage conditions).

The German evaluations of damage percentages under storage conditions have been carried out with theoretical models of the first generation, which implies that these models are in fact modified Jain-Lidiard models, with increasing numbers of model parameters describing more and more complex systems of radiolytic reactions. The problem with this class of models was that a number of very important features of the damage production could not be explained. It should be emphasized that the deviations were observed between the experimental results of lab-irradiated samples and this class of theoretical models. The most important deviations were: (i) the amount of damages in many samples does not show any sign of saturation with increasing dose, (ii) for many samples (including natural rock salt) the production of radiation damage as a function of the irradiation temperature deviates appreciably from the theoretically expected behavior and (iii) these models are not capable to describe the production of voids, an important product of the radiolytic processes in NaCl.

This implies that it is totally inappropriate to use these theoretical models to calculate the damage percentage under conditions so different from those used in the laboratory. We reject the attempts to apply the above mentioned class of models to calculate the amount of damage under storage conditions. At this moment, even with the new generation of models we are not in the position to calculate the production of radiation damage under storage conditions, because the range of conditions where tests in the laboratory have been carried out is far too narrow. Of course, the ultimate goal of the

development of radiolysis models for heavily irradiated NaCl is to predict the production of radiation damage under storage conditions, but unfortunately the new generation of radiolysis models is very complex and these models contain many independent model parameters, which can be adjusted to describe the experimental results. We are convinced that much more experimental and theoretical research is required to be able to provide the necessary accuracy for dependable predictions.

II,2 The issue of the efficiency for energy storage

Reviewing the efficiency for energy storage η , which can be expressed as a percentage (%) as proposed by Mönig (MOE 02)

$$\eta = [E/D] \cdot 10^5$$

where E is the stored energy expressed as usually in J/g and D is the dose, which is expressed in Gy (1 Gy= J/kg).

Usually, the efficiency of the production of stored energy is well below 0.07%. This applies to the experiments carried out under laboratory conditions. We emphasize that it is highly improbable that these numbers also apply to irradiation experiments under storage conditions. As argued elsewhere in this report, we assume that under storage conditions the amount of radiation damage generated at the same dose is much larger than for laboratory experiments. The fact that for laboratory experiments the efficiency is usually well below 0.07% shows again that even for a maximum dose correction factor F (see below in section II,6) the efficiency is still of the order of 1%.

Our conclusion is therefore that much higher stored energies than observed under laboratory conditions and dose rates up to 300 MGy are feasible. In fact our approach to calculate the possible stored energy using the factor F is realistic and should be applied for safety reasons as long as extensive experimental results show otherwise. The proposition that the stored energy values, observed after laboratory irradiation, are the same as in storage sites is a dangerous one, because it might lead to enormous underestimations of the stored energy.

II,3 The saturation issue

From the German side it has been argued at many occasions that the value of the stored energy saturates at rather low values. To support this point of view the results obtained by Donker et al (DON 96) are mentioned. These results show a saturation behavior for the stored energy vs the dose, but it remains to be seen if these results are representative for the real behavior of (doped/natural) NaCl during exposure to very high doses of irradiation. There are several reasons to have doubts about the results published by Donker et al. During the experiments reported by Donker et al the samples were irradiated in the HFR rack at the Petten power plant by a collection of used fuel elements. The elements are exchanged regularly by new used fuel elements, which helps to keep the average dose rate at a sufficiently high level.

Immediately after the emplacement of a new set of used fuel rods the dose rate increases by about a factor of 10 and the heat production per unit volume increases correspondingly. Donker et al claim that the temperature of the samples is kept constant at 100 °C by means of the heat exchanged by He-gas in the sample compartment. It remains to be seen if this is possible, and unfortunately, besides a model calculation there is no direct proof for this claim, because during the experiments the sample temperature was not measured directly and no precise tests have been carried out to be sure that the temperature measured during the irradiation run was the same as the sample temperature.

To explain the differences between our observations and the results of Donker et al we assume that as a result of the variations in the dose rate the sample temperature varies. During the experiments carried out in Groningen, the dose rate is kept at a fixed value by controlling the value of the beam current of the electron accelerator. The temperatures of the samples are measured and controlled continuously. In addition, we have carried out many irradiation experiments using a sample holder with 15 compartments, which were kept at 15 different temperatures between 50 and 150 °C. The advantage of this method is that eventual excursions of the temperature during the irradiation run can be recognized very easily as discontinuities of our experimental observations. Our experiments have shown that (i) the amount of stored energy depends on the irradiation temperature and (ii) details of the behavior of the amount of stored energy as a function of the dose (e.g. saturation) also depend on the irradiation temperature.

We are convinced that our experimental results showing no saturation of the stored energy of a large variety of samples during irradiations at about 100 °C are representative for the real situation of heavily irradiated rock salt, while saturation (for irradiation temperatures of about 100 °C) is observed for some special samples, depending on the dopant. Saturation effects have regularly been observed at temperatures well above 100 °C. In recent experiments (den Hartog et al, unpublished) we have found for heavily irradiated pure NaCl more than 20 % metallic Na and chlorine, which corresponds with stored energies of almost 2 kJ/g, although we have assumed in the past at first sight that pure NaCl might be one of the few sample types that could possibly show saturation of the formation of radiation damage. For many other samples (like natural rock salt and K-doped samples) the efficiency of the formation of radiation damage (i.e. stored energy) is significantly higher than for pure NaCl.

Our conclusion on the issue of saturation of stored energy during exposure to high doses of ionizing radiation at about 100 °C is that the typical behavior of NaCl is that there is no saturation of the stored energy at values of about 200 J/g. Much higher stored energy values are possible.

II,4 The issue of the agreement between theoretical calculations and experiment; what should be done in the future

Starting with our first publication on the subject of heavily irradiated NaCl in the open literature in 1985 (van Opbroek and den Hartog OPB 85) we have attempted at many occasions during the past two decades to compare our experimental results with those obtained with the existing simulation models. In the beginning, when only a limited amount of experimental data was available it seemed that we were quite successful, but soon after we started to produce our own results with our irradiation facility it appeared at numerous occasions that the models were far from ideal and they were not even capable to explain very important features of the damage formation processes.

Our optimism to find suitable models for the radiolysis processes in NaCl decreased significantly until we took a new direction for our modeling work. After introducing the second generation of radiolysis models we were able to explain several important properties of the damage formation, such as the "unsaturated growth" of damage with increasing doses and also the formation of vacancy voids could be described. Unfortunately, there is a significant disadvantage, because the new models contain many new and independent model parameters. Fitting these parameters to describe the experimental results is not an easy task and in particular the most critical and ultimate task to calculate the amount of damage under storage conditions using the models with the fitted parameters should be carried out extremely carefully. At this moment and with the available experimental information this is impossible.

The most important contributions to solve these problems in the future should come from experimental observations under increasingly varying conditions. This implies that the dose range should be increased, as well as the range of dose rates, even though this will be extremely difficult and time consuming. Another indispensable contribution to reaching the ultimate goal of testing the models under the extreme conditions of storage of HLW is to carry out in situ irradiations in a real storage site in a monitoring regime. This can of course be achieved only if sufficient support is obtained from the population, which is often not in favor of the possibility to store HLW in "their" underground environment. It is hoped that this support can be gained after all necessary laboratory research has been carried out successfully and with positive results, providing us with convincing evidence that storage of HLW in rock salt is safe and that storage of HLW is the best possible solution.

II,5 The issue of the very high doses used in the experiments in Groningen

During the first two years of the still ongoing investigations on radiation damage in NaCl in Groningen we have limited our irradiation runs to the range of total doses expected in the vicinity of the canisters with HLW (in the storage concept in the Netherlands ~ 25 Grad, which is equivalent to 250

MGy). For safety reasons we have expanded the range of doses to 150 Grad (=1500 MGy). The first important reason to use the more extended range of doses was that it was not certain, that the natural NaCl crystals in the vicinity of the canisters with HLW can be compared with the materials used in our experiments. Extra production of stored energy was expected in samples with certain (combinations of) impurities and mechanical deformation of the crystals during irradiation. The second reason was that there are enormous differences between the laboratory irradiations and the conditions in the vicinity of the canisters with HLW. The difference in dose rate is in the range 1000 – 10000!

Already in our paper published in 1985 (van Opbroek and den Hartog OPB 85) we have established from our theoretical calculations that the dose rate effects might be appreciable. In many cases the stored energy (SE) value behaves as $SE = \alpha \cdot (\partial D / \partial t)^{1/2}$

It should be recognized during the experimental program that these dose rate effects might cause appreciable differences between the laboratory experiments and the real situation under storage conditions. In fact the amount of stored energy under storage conditions will be appreciably larger than for laboratory experiments. Because it is practically impossible to decrease the dose rate appreciably, we have tried to produce samples in the laboratory, which are comparable with those in the vicinity of the canisters with HLW after a few hundred years of storage. The most practical way to do this is to increase the dose by a factor between $(1000)^{1/2}$ and $(10000)^{1/2}$ or between about 32 and 100. Unfortunately, for a long time due to the limitations of our irradiation facility we were not able within reasonable time periods to reach doses higher than 300 Grad (=3000 MGy).

Recently, we were able to improve our irradiation facility and we have carried out irradiation runs up to doses of 2000 Grad (=20.000 MGy). These doses are required when we want to include dose rate effects in the evaluation of the consequences of the production of radiation damage under storage conditions (see below).

Our conclusion with regard to the issue of the very high doses used in the experiments in Groningen is that it was necessary to increase the laboratory dose drastically in comparison with the dose expected in the storage site with HLW. It appeared to be the most effective way to deal with the problem of the dose rate extrapolation. First, because of the uncertainties of the sensitivity of natural rock salt in the vicinity of the waste canisters (even though we have studied systematically the damage formation in a wide range of NaCl samples) and secondly, because of the dose rate effects in combination with the enormous gap between the laboratory dose rates and those expected in storage sites.

II,6 The issue of the very large differences between the dose rates in laboratory experiments and those under storage conditions

During storage the dose rate in rock salt close to the canisters with HLW is in the range of 10^{-2} Mrad/hr (or 0.1 kGy/hr), whereas in laboratory experiments the dose rate is, depending on the purpose of the experiment, typically 10 - 100 Mrad/hr (or 0.1 - 1 MGy/hr), while recently we were able to increase the maximum dose rate to values well above 1000 Mrad/hr (=10 MGy/hr). It is clear that these enormous differences in dose rate will have consequences for the formation of radiation damage. This is particularly the case when we are interested in the advanced stages of damage formation, because a multitude of temperature dependent defect reactions and defect mobilities play a role. Any model simulating the damage formation using these defect reactions and mobilities will result in dose rate dependent damage formations and the dose rate effects might be quite significant, as we have observed for some of the models that have been developed by our group.

As mentioned in the preceding section we have found in some cases (e.g. in connection with the work of van Opbroek and den Hartog, OPB 85) a rather pronounced dose rate dependence, which can be described analytically by $SE = \alpha \cdot (\partial D / \partial t)^{1/2}$

In more complex models with increasing numbers of model parameters which should be optimized or fitted to ensure the best matching between theory and the experiments we have noticed that the dose rate effects depend on the choice of the parameter sets. This is rather disturbing, because with many model parameters often there is more than one acceptable solution, implying that the existing experimental information is insufficient to make a good choice for the model parameters. One of the fundamental reasons for this is that the range of the irradiation conditions used for the tests is insufficient. It is clear that in order to extrapolate the dose rate over a factor of 10^4 , we need extensive and critical information about the dose rate dependence of the damage production.

It is clear from this discussion, that it is not easy to resolve the situation. We should *not*, as it has been proposed by some people, simply ignore the dose rate effects or just recognized that these effects exist and do nothing to solve the problem. Instead, it is necessary to find practical methods to deal with the problems. A first and rather approach was chosen by our group several years ago, when we measured the sensitivity of a limited number of NaCl samples to low dose rate ionizing radiation (OPLA research project 1986-1990). These experiments turned out to be very delicate and extremely time consuming. The results of these experiments on a small number of samples clearly revealed a significant dose rate dependence, which was in reasonable agreement with our expectations on the basis of the then existing theoretical models. A second approach, which is a bit speculative but much less time consuming, is to increase the dose very strongly. As a first approximation one can use the required dose correction factor associated with the situation of a rather strong dose rate effect

$$F = \left\{ (\partial D_{\text{lab irr}} / \partial t) / (\partial D_{\text{storage}} / \partial t) \right\}^{1/2}$$

Because the difference in dose rate is very large, the dose correction factor F is also large. If we apply the formula in a strict way, we calculate for F a value between 32 and 100!

In our publications, which have appeared until now we have used doses up to 300 Grad (=3000 MGy), i.e. the factor F used until now is about 10 and it has been very modest. It should be realized that most probably the stored energy values reached during our laboratory irradiations are well below those expected under storage conditions. To avoid this problem the doses in laboratory experiments should be increased drastically. To reach the required doses by means of laboratory experiments with a dose rate of e.g. 200 Mrad/hr (=2 MGy/hr) we would have to carry out a non-stop irradiation experiment with our accelerator of about 14 months! This very long irradiation period is not practical and therefore we had to increase the dose rate to 1000 Mrad/hr (=10 MGy/hr). The disadvantage is of course that the difference in dose rate between the laboratory experiments and the storage conditions is increased even further.

The first test irradiation runs have been carried out and the results were quite spectacular. These results will be reported in the open literature in the near future.

II,7 The issue of explosive phenomena in heavily irradiated NaCl crystals and the role of the microstructure

Just like many other scientists working on problems associated with the safe storage of HLW it is our intention to find solutions for the problems with radiation damage in NaCl, which have been discovered by our group. Our approach has been quite different from the ones chosen by the German institutions and the Dutch Institute NRG (which was called in the past ECN). It is aimed at two goals: (i) to learn as much as possible about the explosive phenomena and (ii) to find possibilities, with the acquired knowledge about these phenomena, to exclude with absolute certainty that explosive decomposition of heavily irradiated rock salt in the vicinity of the canisters with HLW occurs in a storage site.

At variance with the doubts that have been expressed by German scientists of the well known institutions and workers of the Dutch organization NRG (former ECN, Petten) about the nature of the observed instability of heavily irradiated NaCl, we are convinced that the instabilities are associated with instantaneous and very violent back reactions. In fact, we have provided unambiguous proof for this in VAI 00a. In addition, we have given a rather detailed description of the processes leading to these reactions in terms of reaction models TUR 01

We have obtained systematic information about the stability of irradiated NaCl, depending on the dopant types and the irradiation dose as early as 1990 (Final report of the OPLA research on "Radiation Damage in NaCl" HAR 90). The results show that depending on the type of impurity the instability

of the samples increases with increasing dose. The results also showed that the observed instabilities were definitely not limited to samples with only very high doses and particular impurities. Some samples (e.g. NaCl:K) appeared to be more unstable than the others. In fact we have observed that only one exceptional type of samples (NaCl:Br) was stable up to the highest doses. All other samples including natural rock salt (both the ones from Asse and the Netherlands) were unstable. Another important feature of the unstable samples was that with increasing damage percentage the temperature, where the instability was observed, decreased. These observations have been reproduced at many occasions during the years after the appearance of the OPLA report (HAR 90); see also HAR 97, VAI 00a, VAI 00b.

Our conclusion was and is that the instability of heavily irradiated NaCl is a common property of this material. In exceptional cases NaCl might be rather stable, but extensive research is required to establish this preliminary observation.

By investigating the microstructure of heavily irradiated NaCl containing increasing amounts of damage we have found important clues for the nature of the instability. In the range of low and intermediate doses sub-microscopic vacancy voids are produced, probably by clustering of neutral di-vacancy pairs. During the course of the irradiation experiment the voids grow. Simultaneously more and more small nanometer sized Na-colloids and chlorine precipitates are formed, but their sizes are always much smaller than the voids. This applies in particular to the chloride precipitates (bubbles), which are appreciably smaller than the Na-colloids. Based on this information and taking into account the results of the important SEM experiments on potentially unstable samples we have concluded that the vacancy voids play a central role in the scenario leading to the explosive decomposition of heavily irradiated NaCl crystals (see e.g. DUB 01, TUR 01, HAR 02).

Because in advanced stages of damage formation the size of the vacancy voids is significantly larger than the average distance between two nearest chlorine bubbles a small percentage of the chlorine molecules is captured by the voids. We have calculated that chlorine pressures of typically about 50 bar are expected in the voids. Because apart from some exceptions the Na-colloids are significantly larger than the chlorine bubbles voids collide with Na-colloids in the late stages of the damage formation. At the moment of a collision the chlorine gas of ~50 bar within the void there will be a chemical reaction with the atoms in the Na-colloid. During this highly localized back reaction the local temperature increases very rapidly and this causes the local pressure to increase very rapidly as well, which leads under certain conditions to explosion-induced crack formation. The cracks are oriented along the (100) crystallographic planes, which are cleavage planes (for a more detailed description the reader is referred to DUB 00 and HAR 02).

In general one can say that the presence of elongated voids (= oriented explosion-induced cracks) is an indication for the onset of instability of the heavily irradiated sample. Extensive crack formation ultimately leads after continued exposure to ionizing radiation to explosive decomposition of the sample within an extremely short time period. As mentioned above, voids play a central role in these phenomena. However, also the chlorine precipitates and the Na-colloids are important ingredients in the explosive back reaction. Necessary conditions for unstable NaCl are: (i) the existence of sufficiently large voids, (ii) the presence of sufficient amounts of chlorine and Na precipitates and (iii) the Na particles should be significantly larger than the chlorine bubbles, because only in this situation sufficient amounts of stored energy are transformed locally into heat, giving rise to a shock wave which triggers back reactions at a macroscopic scale.

We assume that in principle these conditions can be met under storage conditions for a number of NaCl materials, depending of the dopant(s). Therefore our main task is to find technologies to achieve appreciable reductions for the production of radiation damage in NaCl expected under storage conditions, i.e. reduction of colloid, chlorine and void production. In addition, because of the uncertainties connected with the extrapolation it will be necessary to monitor the stability of the rock salt in the vicinity of the canisters with HLW. Even if our simulation models (which can be built only on the bases of our limited experimental data, obtained in a narrow range of dose rates) would suggest that under particular conditions the expected amount of damage are moderate one should be careful. The storage strategy should allow us to take appropriate measures as soon as the irradiated rock salt reveals signs of instability.

Our conclusion is that a combined experimental and theoretical research program will allow us to build the necessary confidence that the problems, associated with radiation damage can be overcome by the development of innovating technological measures and a strict monitoring program during storage.

II,8 The issue of the localization of the explosive effects in a storage facility

Radiation damage is formed in those regions surrounding the canisters with HLW where the dose rates are sufficient. As a result of the absorption of radiation energy and the reduction of the irradiation flux with increasing distance to the irradiation source the dose rate decreases rapidly as a function of the distance from the canister. This implies that the formation of high levels of radiation damage is confined to cylindrical region in the vicinity of the canister (e.g. 50 cm).

At first sight one could think that the localization of the damage formation in the close vicinity of the canisters leads to localized, explosive effects. In fact this line of reasoning is heard from the representatives of the well known German institutions, involved in the problems of radioactive waste management. This line of reasoning, however, is not correct, because an

explosion occurring in irradiated rock salt close to one particular HLW canister will be capable to trigger coherent explosions in the corresponding layers of heavily damaged rock salt of *all* remaining canisters, wherever they are in the salt formation, by means of shock waves. This implies that the explosive effects are by no means localized. In principle all unstable, heavily damaged rock salt present in the storage site contributes to the explosion events in the storage site.

To explain this line of reasoning, let us consider the situation, that in the vicinity of a canister with HLW very locally an explosive back reaction is triggered. Due to the very rapid increase of the local temperature and the associated increase of the local pressure a shock wave is created, which travels in the salt. In the region close to the canister there is a certain mass of heavily damaged NaCl and the shock wave from the initiating trigger induces this material to release its stored energy exactly at the moment of passage of the extremely narrow shock wave. In this scenario the intensity of the wave increases as long as it traverses NaCl with sufficient stored energy. This process enables the heavily irradiated rock salt surrounding the canister to release almost all its stored energy. Of course, the narrow shock wave does not stop at the boundary of the damaged region. It is transmitted almost in an ideal way by the rock salt formation and it is not attenuated strongly. Roughly, its intensity decreases with increasing distance, r , to the source

$$I_{\text{shock wave}}(r) = I_B \cdot [R/r]^3$$

Here, R is the distance between the "central position" of the heavily irradiated zone and the boundary of the heavily irradiated rock salt; it is estimated to be about 1 m. The distance r is the distance between the "central position" and the position in the rock salt formation (outside the damage zone) where we want to calculate the intensity of the shock wave. It is of course important to know the intensity of the shock wave when it arrives at a neighboring location in the storage site, where a canister with HLW is placed. This distance depends on the storage concept.

In the old concepts it was planned to use deep bore holes for the emplacement of large numbers of HLW canisters, one on top of the others. From the discussion given above it is clear that within one stack of canisters the shock wave will be able to release all stored energy in the vicinity of all vessels in the same bore hole. Another important question will be if the stored energy associated with the canisters in the other boreholes will also be released in the same event. In our opinion this is definitely possible if no special measures are taken.

Our conclusion is that the strong coupling of the explosive events and the coherence of the shock waves associated with the localized, radiation induced explosive events should be taken into account in the safety assessment. The instantaneous release of all stored energy in a salt dome is possible; it might be initiated by coherent shock waves, if no special precautions are taken. Taking into account the total amount of HLW in a storage site in the rock salt formation and the associated stored energy the

effects of the explosions for the salt formation as a whole can be quite drastic. This is the reason why much more research should be carried out to reduce the effects of radiation damage in NaCl.

III. What can/should Germany do?

Obviously, the decision in Germany to abandon the research on radiation damage in heavily irradiated NaCl was wrong and it will take considerable efforts and time to repair this mistake. In principle, there are sufficient German researchers capable to carry out valuable research in this area, but one decade of silence in the research area of radiation damage in NaCl on the German side did have serious consequences.

There is another problem, which is far more severe. It is related with the reason why the research on radiation damage of NaCl has been abandoned. For many people it is certain that the decision to stop the research on radiation damage in NaCl was made for political reasons. High ranking persons have said that radiation damage was not a problem. This kind of approach of the scientific problem will turn out to be a major difficulty, because several scientists will have been influenced by this and moved away from a subject, which is very important, in particular for Germany. The most relevant question is now how does Germany get qualified and critical scientists back into the field of radiation damage in heavily irradiated NaCl. This question is related with many other questions regarding the nuclear energy cycle. The most important one being: Is it possible to earn back the trust of the reasonable average German citizen.

Also for the average citizens there are problems. Because for many of them it is clear that discussions about the issues, connected with radiation damage in NaCl, have been avoided systematically. This is felt very strongly by many people, who were interested to take part in the discussions about nuclear power. They ask themselves, is there anything related with radiation damage in NaCl that the authorities are hiding for us?

It is clear that in the first place the German authorities should start an open discussion about the feasibility of storage of HLW in rock salt formations (or eventually other natural hosts). The discussion should not be limited to a discussion about the best choices for a location. The role of scientists in the feasibility discussion is an important one, because it is possible in principle to benefit from the independent and critical ideas put forward by these scientists. Until now on an overwhelming scale scientists from German institutions played a role in the general debate and also the advise for the Government and quite often the information for the members of the German Parliament is provided by workers from these institutions. Of course it is important for a Government to be certain that sufficiently qualified people are available to solve all problems associated with the storage of HLW, but equally important is, especially when the issues are controversial, that there is a significant contribution from independent scientists. At the present

moment the discussion in Germany is held at two different and totally separated levels. There is a discussion among experts and there is a discussion in the German society.

This situation is unhealthy and it will not lead to solutions, because a large part of the public will say that the experts are biased, because they depend on their institutions, which on their turn depend on the Government. It will not be easy to change the situation, but changes are required otherwise the controversy will persist and the public will not trust its own Government. In conclusion: An open debate on all subjects related with the storage of HLW should be held and efforts should be made to stimulate that independent scientists make their contributions to the solutions of the "hard-to-crack" problems, among which "Radiation damage in NaCl".

It might be necessary to change the strategy of the management of HLW. *Retrievable storage* combined with an appropriate *monitoring* program has many advantages. Our group has been promoting this storage strategy since the end of the 70's of the last century, because it will help us to solve a number of "unsolvable problems" and it might help to gain public acceptance for underground storage of HLW. In the Netherlands the Government and Parliament have decided as early as 1993 that retrievability is a requirement for eventual storage of HLW. Also the research activities in the Netherlands are carried out taking into account this boundary condition. In discussions I have never detected any support from the German side for retrievable storage and it would be a significant step in the right direction if Germany would, just as the Netherlands did, decide that storage of HLW should be retrievable.

IV. References

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