INTER AND INTRACRYSTALLINE CREEP PROCESSES OCCURRING DURING SALT IRRADIATION

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ABSTRACT

Results of the experiments performed by our group in the last eight years show that salt creeps due to irradiation provided the dose rate is low enough. This article summarizes the observations and arguments on which our conclusion is based.

Optical microscopy observation of salt samples before and after gamma-irradiation experiments was performed. The operation of radiation-induced intracrystalline creep processes was inferred from the presence in the irradiated samples of microstructures indicative of both incipient and very advanced creep which were not present in the unirradiated samples. The observations could be made by ordinary optical microscopy because dislocations in NaCl are decorated blue by the Na-metal colloids developed by irradiation. These intracrystalline creep microstructures were moreover observed to be coupled to radiation damage anneal at the microscopic level, and to contribute to the attainment of radiation damage steady state.

Intercrystalline creep processes were also proven to take place during irradiation. Amongst them the best observed was recrystallization because its operation annihilates all the crystal defects returning the NaCl to its original colourlessness. Recrystallization consumes the energy previously stored in the crystal defects. Two types of recrystallization were observed. One of them consists of the migration of new (and dry) grain boundaries created by intracrystalline creep. The other recrystallization type consists of the migration of already existing wet grain boundaries and is known as Fluid Assisted Recrystallization (FAR).

1. INTRODUCTION

Gamma-irradiation damages the NaCl crystals by developing lattice defects such as F- and H-centres. F-centres can precipitate forming Na-metal particles of colloidal size and properties. In order to develop colloids, a sufficient concentration of F-centres is necessary. A crystal containing F-centres and small quantities of colloidal sodium ($<10^{-5}$ molar fraction) becomes yellow. Higher amounts of colloidal sodium in the NaCl lattice turn the rock salt blue. The coloration of the salt therefore is related to the amount of damage, and thus observation of damage variations is possible by optical microscopy.

Three series of irradiation experiments were performed, all three at constant temperature (100°C) and at approximately constant dose rate (either 4 or 15 kGy/h), and each series up to ten different total doses between 0.02 and 45 MGy . Each experiment contained 16 different salt samples but different experiments of the same series had equivalent samples. These were the experiments called GIF B experiments which took place in the framework of the HAW-project. The methodology applied can be read in García Celma et al., [1995 a].

The samples consisted of natural rock salts, synthetic polycrystals and synthetic monocrystals. The natural rock salt samples were taken from the Asse Mine (Germany) and from the Potasas del Llobregat Mine (Spain). The synthetic polycrystals were prepared in the laboratory using as original material salt powder of analytic purity. The synthetic high purity single crystals were purchased from Harshaw Ltd. and were also poor in OH^- contents.

Equivalent samples were subject to equivalent irradiation experiments but differed from one another in that they could contain some added brine or not. Or they could have equal amounts of brine added but one of them was subjected to enhanced pressure (of 200 bar) during irradiation while the other was not.

This article reviews the observations made in thin sections of these samples and the interpretations made with the help of many other measurements and observations performed from 1986 on. We will first consider the links between the microstructures and the colloid development, and then the way in which intracrystalline creep anneals the radiation damage and will finish considering intercrystalline creep. Intercrystalline creep is in this case mostly "static" recrystallization, since the samples were not subject to directional stress. More information on the same samples and experiments can be found in De las Cuevas and Miralles, [1995]; Donker and García Celma , [1995 a and b]; Donker et al., [1995] ; García Celma and Donker [1994 a and b]; and García Celma et al., [1993, and 1995 a and b]

2. INCIPIENT RADIATION DAMAGE DEVELOPMENT AND MICROSTRUCTURES.

For the F-centres to precipitate and develop colloids in a NaCl crystal, they need to be kept from recombining with the H-centres. According to Hobbs et al., [1973] perfect edge dislocation loops (011) develop and coalesce in irradiated NaCl crystals. H-centres are fixed as Cl_2 molecules at both sides of the extra half plane of these loops, and thus kept from recombining with the F-centres. The F-centres on their side could precipitate in the dislocation lines as suggested by Hobbs [1976].

García Celma and Donker, [1994 b] show that, since dislocation structures are mostly blue (colloid containing) and they constitute creep microstructures which were not present before irradiation, colloids do not only develop together with the dislocations and at the dislocation lines, but migrate and anneal together with them.

However, even if complementary defects are fixed at different crystallographic sites regarding a given crystallographic direction, nucleation of defect aggregates such as colloids would be greatly improved if a field of diffusion would be imposed to the homogenous crystal. This field can be created by the existence of important sinks such as dislocation walls ((sub)grain boundaries).

For all the irradiated samples microstructural observation shows that the first colloid development takes place at the grain boundary region. The portion of the crystal in contact with the exterior surface is devoid of colloids and constitutes a white band of variable thickness (2 to 7 μm) which is limited inside the crystal by a dark blue line parallel to the crystal boundary. In crystals with incipient colloid development, from this dark blue line towards the centre of the crystal the blue colour gradually fades out. Levy et al., [1980] observed the white rims at the grain boundaries of irradiated salt samples and showed them to be coupled to a higher content of impurities. They interpreted these rims as due to difficulties of colloid nucleation caused by impurities. Figures 1 and 2 show the aspect of these microstructures, see as well García Celma and Donker, [1994 a and b] and García Celma et al., [1995 b] for other appearances of the same microstructure.

This microstructure presents the same characteristics as the void distribution in irradiated metals described by Bullough and Nelson, [1974]. Bullough and Nelson, [1974] show that, in the area of the crystal boundaries, voids are absent at the exterior surface, while at a short distance within the crystal a sharp line parallel to the exterior surface marks an area of a very high void concentration behind which, towards the interior of the crystal, the void concentration gradually diminishes. This distribution would be reached thanks to the creep mechanism described by Nabarro, [1948] who observed that irradiation produces deformation of metals, and proposed a mechanism consisting of preferential diffusion of vacancies and interstitials, produced by irradiation, towards crystal boundaries, where they disappear. This diffusion would create a concentration gradient and therefore a stress field would develop which could drive crystal defects.

Jiménez de Castro and Álvarez Rivas, [1990] show that most of the stored energy produced by irradiation of NaCl is found at the surface of the crystal.

On the basis of the microstructural observations of the white-blue boundaries García Celma et al., [1995 b] developed a version of the Jain Lidiard model with a description of the dislocation density gradient to be expected at a grain boundary and a definition of the grain boundary itself in terms of dislocation density and calculated the colloid density as a function of the distance to the grain boundary. They found a distribution of colloids equivalent to that of the voids in Bullough and Nelson, [1974] and to that of the colloids found in the microstructure reproduced in Figures 1 and 2.

Therefore the existence of crystal exterior surfaces (or of the grain boundaries) could be important for the stabilization of radiation damage. The existence of material discontinuities functioning as huge sinks for crystal defects combined with the effect of the different diffusivities towards them create gradients of defect concentration which make the precipitation

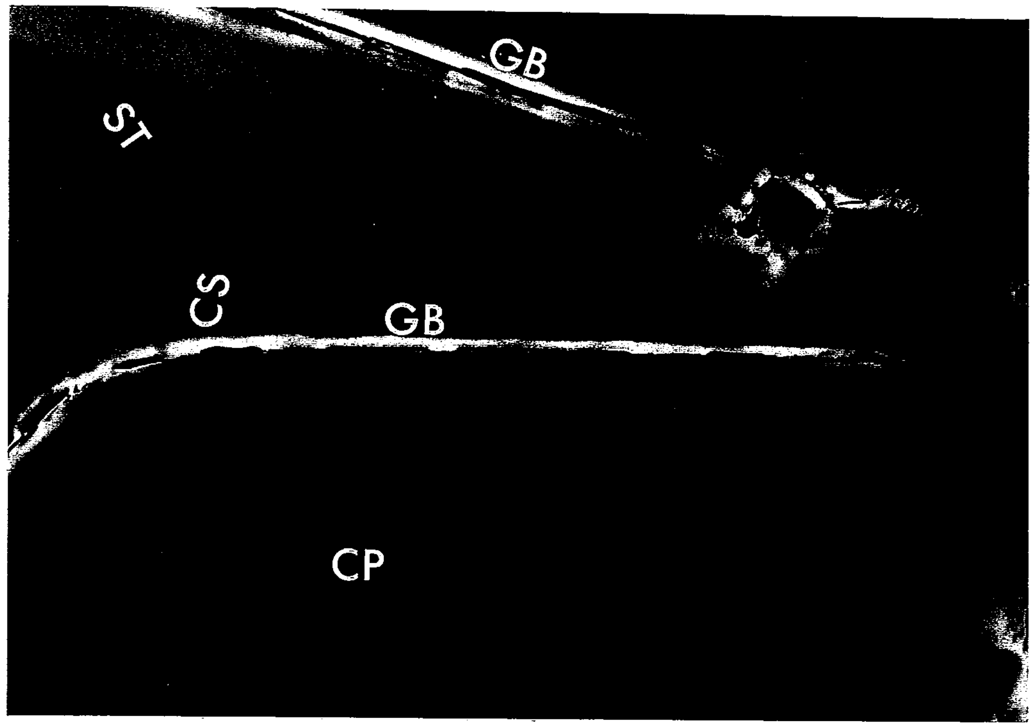


Figure 1 : *Colourless bands at grain boundaries (GB). Slip traces (ST) cross slip (CS) structures and cellular patters (CP) indicated. See as well Fig.4. Asse Speisesalz sample, 15 Sp-800. Irrad. cond.: 100°C, 4 kGy/h, 4.6 MGy. Mag. 216 X*

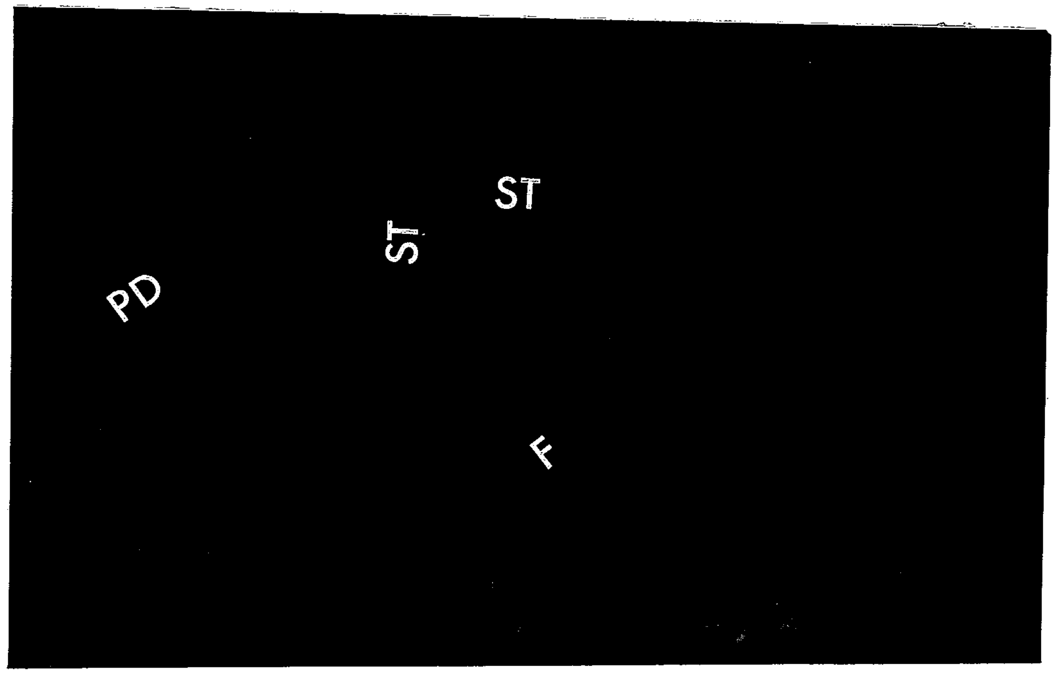


Figure 2. *Colourless rims limited by intense blue lines developed at (001) fractures (F) and also at plastic deformation (PD) structures. Slip traces (ST) <011> indicated. Asse Speisesalz sample T1 Sp-800. Irrad. cond.: 100°C, 15 kGy/h, 2.6 MGy . Mag. 338 X.*

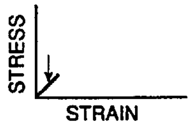
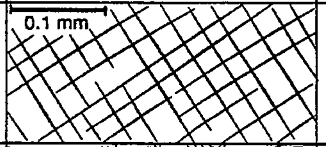
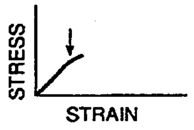
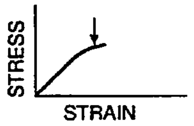
of different defects in different areas of the crystal possible. At the lattice level, of course, the defects would precipitate in their corresponding places, with the difference that the possibility to reach their sites before recombining is greatly improved by the existence of a gradient which acts differently on the different defects (due to their different diffusivities).

3. INTRACRYSTALLINE CREEP AND DEVELOPMENT OF RADIATION DAMAGE STEADY STATE (SATURATION).

First we will summarize a couple of creep principles. As long as most dislocations can move, the stress within a crystal lattice can be resolved in a plastic way, and the crystal creeps. Dislocation motion brings dislocations together. Negative interferences annihilate the dislocation, repairing the crystal lattice. Positive interferences produce rearrangements in lower energy structures such as dislocation walls [Hull and Bacon, 1984]. Dislocation walls, due to the preferential diffusion along them, produce a gradient of defect concentration which forces diffusion towards these walls. Thus impurities are forced towards grain and subgrain boundaries where also the highest dislocation wall density is present.[Sabharwal et al.,1975; Harris and Fiasson, 1976; Geguzin et al., 1976; Riggs and Wuttig, 1969; Filippov and Gaidukov, 1993]

Hobbs [1976] observed that the perfect edge dislocation loops which develop by irradiation in ionic crystals, grow and coalesce and give rise to dislocation forest development. Dislocation forests are considered to be the reason of hardening in crystals, because in dislocation forest the dislocations pin each other hindering each others mobility and thus the plastic behaviour of the material. They are the reason of the hardening undergone by NaCl when irradiated. Dislocation forests cannot be observed in salt by optical microscopy.

Senseny et al., [1992] gave a list of the substructural processes taking place under increasing total strain for rock salt and of the microstructures thus produced and related them to the three stages of a creep curve. This was summarized by García Celma and Donker [1994 b] as shown in Fig. 3. The hardening stage (stage I) corresponds to the development of dislocation forest development. Where the creep curve bends to enter stage II, glide is active. In stage II cross slip is added to glide, and at the end climb can take place. Rotation recrystallization is added to the rest of the processes in the accelerated creep part, stage III. All this under the assumption that temperature and confining pressure are high enough and strain rate low enough to allow plastic behaviour of the rock.

DISLOCATION MOTION MECHANISM	DIAGNOSTIC MICROSTRUCTURE		ONSET OF MECHANISM AND ITS EFFECT ON "A CREEP CURVE"
	MORPHOLOGY	NAME	
DISLOCATION DEVELOPMENT	NOT VISIBLE IN OPTICAL MICROSCOPY	DISLOCATION (FORESTS)	
GLIDE		SLIP TRACES	
CROSS SLIP		CRENULLATED SLIP TRACES	
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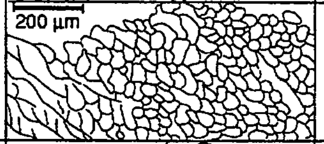
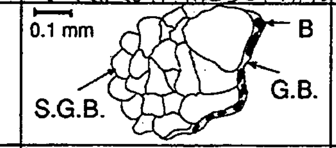
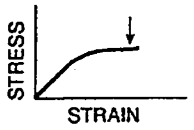
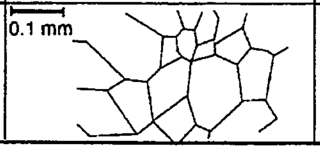
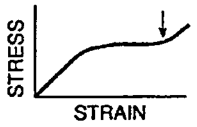
		CELLULAR PATTERNS	
CLIMB	 	SUBGRAINS (POLYGONS)	
DISLOCATION WALL MIGRATION		FOAM-TEXTURE	

Figure 3. *The diagnostic creep microstructures and the mechanisms of dislocation rearrangement which produce them. GB stands for grain boundary, B for brine, and SGB for subgrain boundary. Reproduced from Garcia Celma and Donker, [1994 b]. Based on Senseny et al., [1992].*

All the microstructures here described are diagnostic of different dislocation motion mechanisms and have been observed after chemical etching of deformed NaCl [Urai et al. 1987]; in aditively coloured samples they are decorated by colloids as well [Amelinckx, 1964].

Dry (sub)grain boundaries are generally present in the non irradiated Asse Speisesalz samples, and cellular patterns could have been in some crystals or some of the Asse samples, but the slip traces of Fig. 4, the subgrains in Fig. 5, and the new grains of Fig. 6 certainly develop during irradiation of Asse Speisesalz and of Harshaw single crystals which did not have any of these structures before irradiation.

The (blue) deformation structures which were not present in the original material are the microstructural expression of incipient creep (slip traces, Fig. 4) and final creep (foam texture) (Fig. 6) [Senseny et al., 1992]. Therefore it has to be concluded that dislocation motion occurred during irradiation. Moreover, there are many crystals which show intermediate structures (Fig. 5), suggesting change from one creep mechanism to another, and although we have to recur to the deformation literature to know which microstructure is transforming into which [Senseny et al., 1992] (Fig. 3), it cannot be doubted that the samples crept during irradiation and that this creep kept them from further hardening under the influence of the gamma-radiation.

The observed dislocation arrangements are in all stages colloid-decorated. Therefore the sodium colloids have to have migrated during irradiation tracking the deformation structures. Probably a dislocation line cannot drag along a big colloid. It seems reasonable to assume that colloids are annealed from some places and re-developed in others, e.g. are eliminated from places where dislocations went through (from the cores of the developing (sub)grains). This means that an anneal mechanism of radiation damage defects, which is different from that usually considered in the theoretical models, has to be taken into account. It also means that the rate controlling mechanisms of this anneal are the rock creep mechanisms.

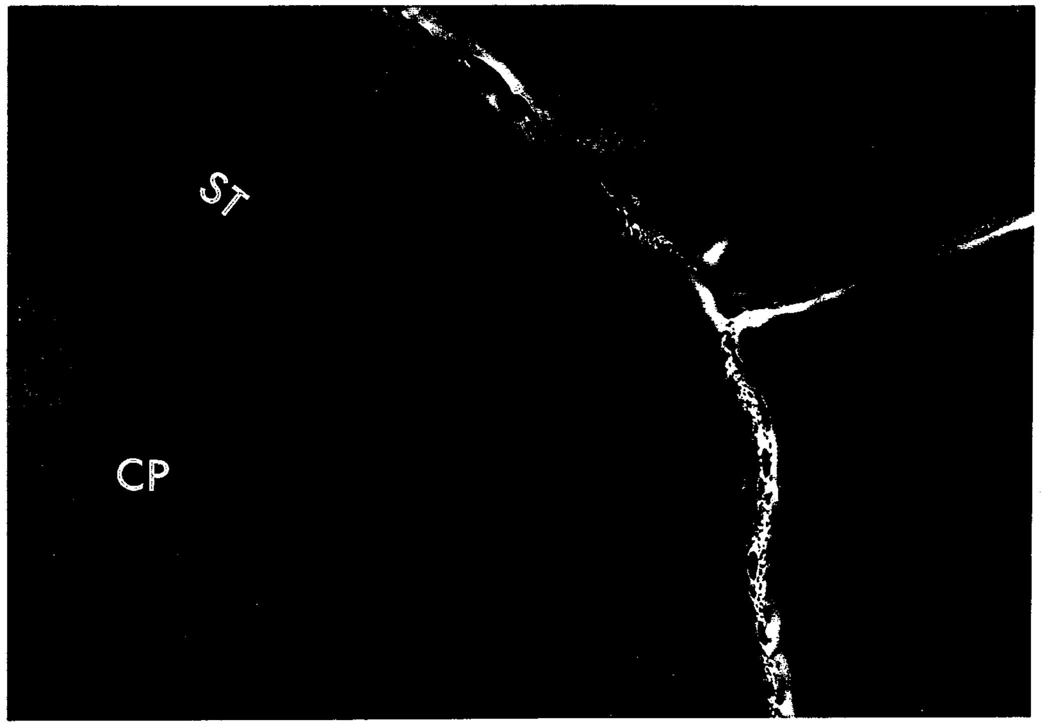


Figure 4. *Slip traces (ST), to cross slip and cellular patters (see as well Fig.3). Colourless grain boundary with black structures. Asse Speisesalz (15 Sp800). Irrad. cond.: 100°C, 4 kGy/h and 4.6 MGy. Mag. 216 X.*

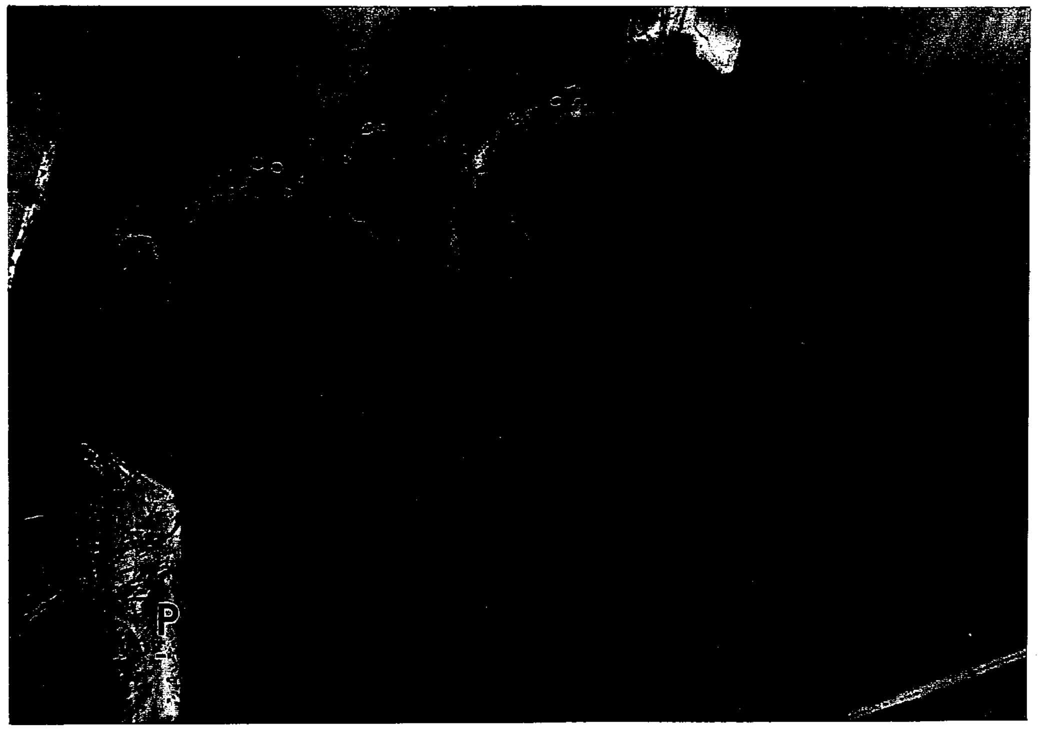


Figure 5. *Subgrain boundaries developing from rearrangement of cellular patterns. The grain boundary between the polyhalite (P) and the halite contained many fluid inclusions, the black structures. Asse Speisesalz (T3 Sp800). Irrad. cond.: 100°C, 15 kGy/h, 4 MGy. Mag. 216 X.*

García Celma and Donker [1994 a] observed that anneal associated to creep structures is more active in natural rocks than in pure undeformed single crystals. Since the samples on which this statement was based were irradiated simultaneously, the reasons for this must lie in the differences between the samples; either intracrystalline enhancement of creep by H_2O (dislocation mobility enhancement by H_2O) [Carter and Hansen, 1983] is very important, or polycrystallinity is. Most probably both.

Regarding the enhancement of dislocation mobility by H_2O it can be seen that around polyhalites, at which boundary with halite many fluid inclusions are present before irradiation, a blue aureole develops for low total doses while the most intense recovery processes (see Fig.6) are observed for high doses. On the other hand, the competence contrast between polyhalite and halite could also cause deformation structures to develop at their boundaries by partitioning of the otherwise homogeneous stress field to which the samples are subject during irradiation. These homogeneous stresses must then be the result of confined thermal expansion of the samples since the experiments take place at 100°C and all microstructures appear in the samples regardless of whether they were subject to enhanced pressure during irradiation or not.

Two reasons can be suggested to explain the enhancement of anneal associated to creep during irradiation in polycrystals as compared to our single crystals: grain size differences between the considered samples and grain shape interferences when volume changes are required.

The influence of grain size is due to the fact that the volume of preferred diffusion paths is important in damage development and creep. This volume is much higher in the natural rocks than in the single crystals. The single crystals used in the experiments consist of tablets with a diameter of 24.28 mm and a length of 10 mm, and without subgrains. In Sp-800 samples the individual crystals can be approximated by spheres with a mean diameter of 5 mm which moreover include a web of subgrains of a diameter of about 200 micron. Preferred diffusion and sinking of lattice defects towards dislocation arrangements in grain and subgrain boundaries has to be more important in Sp-800 samples than in the pure single crystals. This could produce differential stresses within the crystals [Nabarro, 1948] which would enhance dislocation mobility.

Let us consider the shape interferences. When a grain becomes larger the neighbouring grains will have to accommodate their shapes to this expansion. In our case, volume increase due to dislocation loop development and accommodation of the Cl_2 molecules and to thermal dilation (because of the 100°C at which the experiments were performed) has to be expected. Thanks to different intensities of slip and/or climb of dislocations in the different crystallographic planes, the crystal shape can change when placed under differential stress by an expanding neighbour. The differential stress is produced by partitioning of deformation depending on the relative crystal exterior shapes, e.g. if the boundary between two crystals displays a sharp angle and one of the crystal expands more in this direction because (e.g.) it contains more impurities (which ease dislocation loop nucleation), then the homogeneous stress field is partitioned into differential stresses and dislocation migration (or crack development) takes place. It is therefore easier for colloids to interfere with passing dislocations than would be the case when the slip planes are only determined by irradiation-produced dislocation loops mobility, as in the single crystals.

Regarding the intracrystalline processes taking place during irradiation of polycrystals it is important to take into account that at the start of a deformation episode different grains can be in different states of strain, that strain is higher at grain mantles since by developing dislocations in the mantles the cores of the grains are protected from deformation, and that depending on the orientation of each grain with respect to the deformation the rate of each process will be different in each grain. Thus one grain can display advanced recovery structures while another may start to develop damage microstructures.

We assume that the fact that the stored energy accumulated in lattice defects saturates for natural samples at values lower than that corresponding to colloid saturation as calculated using the Jain-Lidiard model for the corresponding experiment is due to the operation of creep processes [Donker and García Celma, 1995 a]. The experiment in which saturation of damage was reached was such that fluid assisted recrystallization had to be excluded of the possible reason for the low stored energy values. Note then that the low value of stored energy at saturation could be due not only to the development of a possible creep steady state and therefore stabilization of stored energy attached to dislocations, but also to the enhanced anneal of the rest of radiation damage defects which crept together with the dislocations [Donker and García Celma, 1995 a].

Figure 6 a. *Anneal of colloids produced by dislocation wall migration ("dry" recrystallization). Note however, that these bleached areas can contain polyhalite (P). Asse Speisesalz (T41 Sp-800). Irrad. cond.: 100°C, 15 kGy/h , 15.9 MGy. Mag. 34 X*

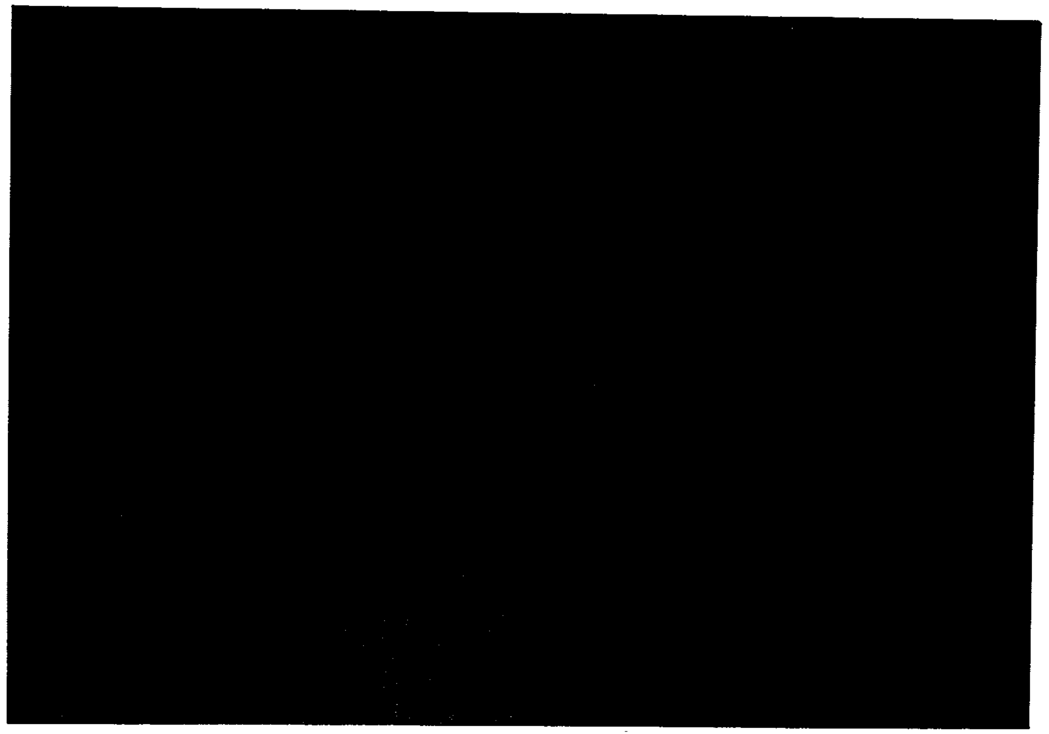


Figure 6 b. *Foam texture developed by dislocation wall migration ("dry" recrystallization) inside the bleached areas observed in Fig. 6 a. Asse Speisesalz (T50 Sp-800). Irrad. cond. : 100 °C, 15 kGy/h , 15.9 MGy. Mag. 134 X.*

Taking into account the properties of creep [Guillope and Poirier, 1979; Poirier, 1976], this additionally means that, at least if the irradiated samples are not allowed to expand during irradiation, lower irradiation dose rates (deformation rates) enhance recovery and the associated anneal versus damage development. Saturation of radiation damage (steady state) would be reached at lower levels of damage for lower dose rates in polycrystalline samples.

4. INTERCRYSTALLINE CREEP PROCESSES (RECRYSTALLIZATION) AND RADIATION DAMAGE ANNEAL.

The only intercrystalline processes which have been observed in the samples being considered are recrystallization processes. First of all, the well known "dry" recovery recrystallization, which takes place as a consequence of the creep processes described above, and then an additional type of recrystallization, which depends on the existence of brine at the grain boundaries and which is called Fluid Assisted Recrystallization (FAR).

The "dry" recrystallization is a consequence of the development of dislocation walls which result of the combined action of glide and climb inside a crystal. With increasing deformation (radiation-induced in our case) the misorientation between parts of a crystal across the dislocation walls becomes so important that the dislocation walls acquire the properties of grain boundaries and try to reduce their surface energy. This is reached by migration until the grain boundaries become straight and meet at triple points delineating angles of 120 degrees, what constitutes the characteristic high equilibrium "foam" texture (see figure 3). Wherever a foam texture is found it has to be assumed that grain boundary migration (recrystallization) took place. Note that recrystallization always implies that, the material which constituted the old crystal (which is being invaded by the migrating grain boundary) is incorporated step by step in the lattice of the new grain, but the lattice defects of the old crystal are not (except for perhaps some impurities). In this way the previous defects are annealed and the structure is recovered [Guillope and Poirier, 1979; Senseny et al., 1992]. This has been the case in the structure of figure 6.

Fluid Assisted Recrystallization (FAR) on the contrary consists of "wet" grain boundary migration. A "wet" grain boundary can be observed in Fig. 7. FAR proceeds by solution of damaged NaCl in the brine at the grain boundary voids, transport in solution, and reprecipitation of newly produced NaCl. In this way the wet grain boundary migrates [Urai et al., 1986; Urai, 1983]. Note that this process also eliminates the defects which were present in the old crystal, since it is dissolved in the brine.

García Celma et al., [1988] showed that FAR extensively takes place (after irradiation) in heavily irradiated samples and eliminates radiation damage. In their experiments FAR operation was proven by the existence of fluid inclusions containing H_2 and decorating the growth surfaces of the recrystallized grains. Hydrogen is produced by the reaction:

$$2\ Na + 2\ H_2O \rightarrow 2\ NaOH + H_2$$

which takes place during solution of the Na-colloids at the grain boundary void.

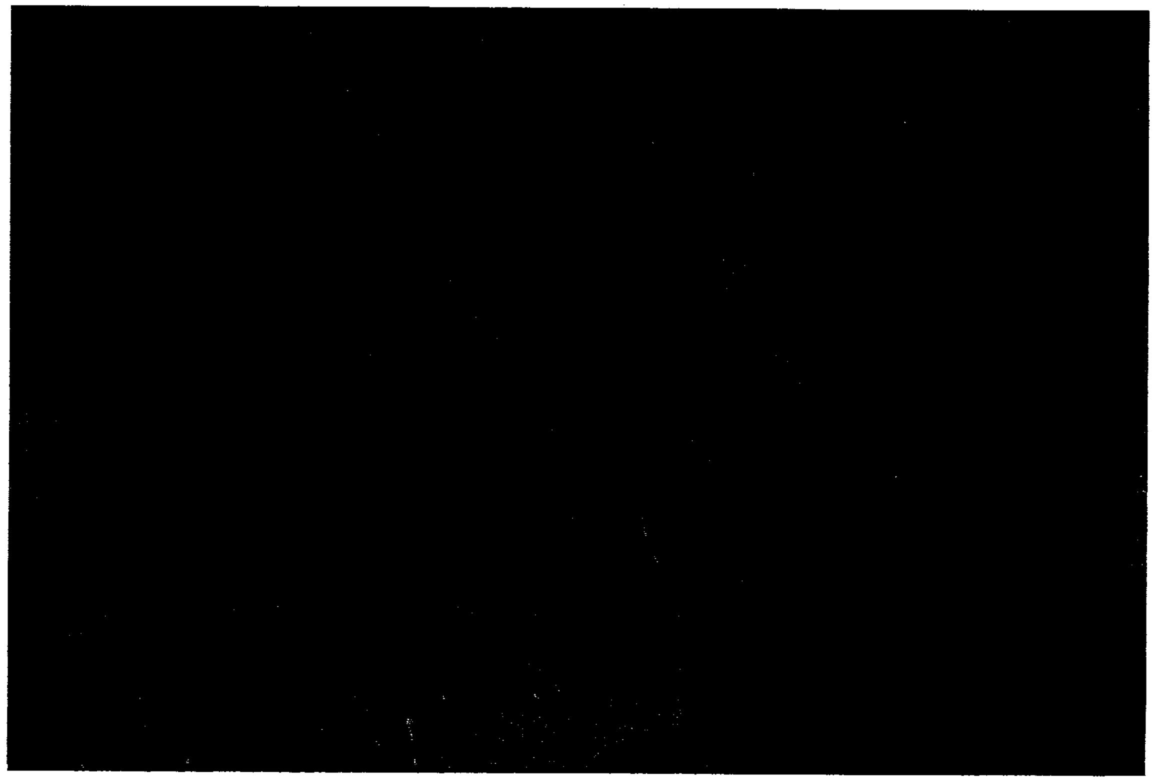


Figure 7: *Wet grain boundary. The voids in the grain boundary surface at various depths contain brine or have lost it during section preparation in which case they are observed as black. Micrograph of a "thick" thin section of an Asse Speisesalz sample (8Sp-800) . Mag. 216 X*

Characteristic of FAR-produced areas is that their contacts with the blue material are, on one side a wet grain boundary, and on the other side the previous position of the grain boundary. FAR-produced areas present the morphology of overgrowths. Moreover, FAR-produced areas have different substructures (e.g. subgrain boundaries) than the rest of the crystal, and are perfectly white (see Fig. 8), except when re-irradiated (see Fig.9).

Whether Fluid Assisted Recrystallization could proceed without decomposing brine and/or with brine in the vapour phase was a matter of discussion. If recrystallization can proceed without decomposing brine and can also proceed when the brine is in the vapour phase, can proceed during irradiation and cyclically, then recrystallization could operate continuously in a repository. These questions were solved by García Celma et al., [1993]. They showed that the solution of irradiated NaCl in H_2O, which is the process responsible for brine decomposition during FAR, only decomposes the H_2O when the F-centres have precipitated as Na-colloids. Therefore, as long as colloids have not developed, FAR can proceed while the amount of brine at the grain boundary is maintained. García Celma et al., [1993] also proved that solution and reprecipitation of NaCl, which is per definition FAR, takes place at 150°C and atmospheric pressure, in which conditions brine has to be in a vapour state. Their proof was that grain growth takes place in samples subject 150°C and atmospheric pressure. Therefore, FAR can take place even if the brine is in the vapour phase. Since samples not subjected to these temperatures did not recrystallize, the enhancement of FAR by temperature was proven.

The occurrence of FAR during irradiation was also proven by García Celma et al., [1993] since, in order to produce variations in grain sizes, the grain boundaries have to migrate and grain boundary migration is per definition recrystallization and they showed grain growth to take place during irradiation (compare Fig. 10 with Fig. 11). To explain their quantitative microstructural determinations they also had to assume that recrystallization took place more than twice on the same material during the experiments. These experiments had been performed with samples consisting of cold pressed NaCl powder of a starting microstructure as shown in Fig. 10 and an end microstructure as that of Fig 11. These recrystallization was enhanced by brine; and thus inferred to be FAR.

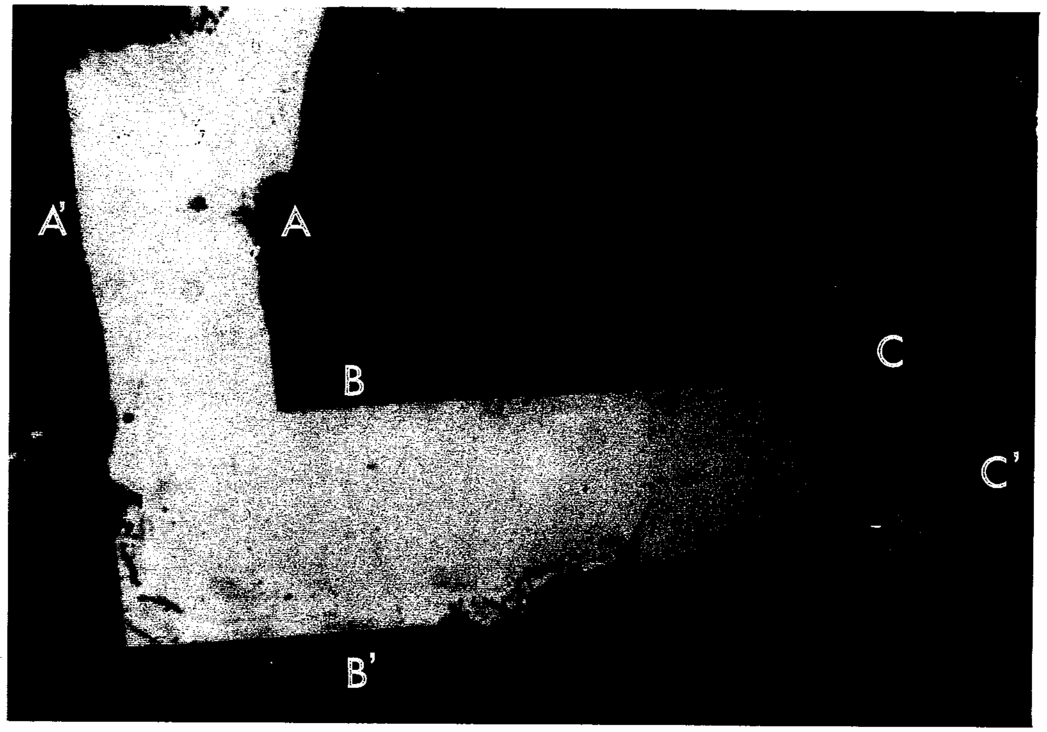


Figure 8 a : *Fluid Assisted Recrystallization. The grain boundary at ABC has migrated to A'B'C' and the colloids have been completely annealed. Asse Speisesalz sample T8 Sp-800. Irrad. cond: 100°C, 15 kGy/h, 7 MGy. Mag. 86 X.*

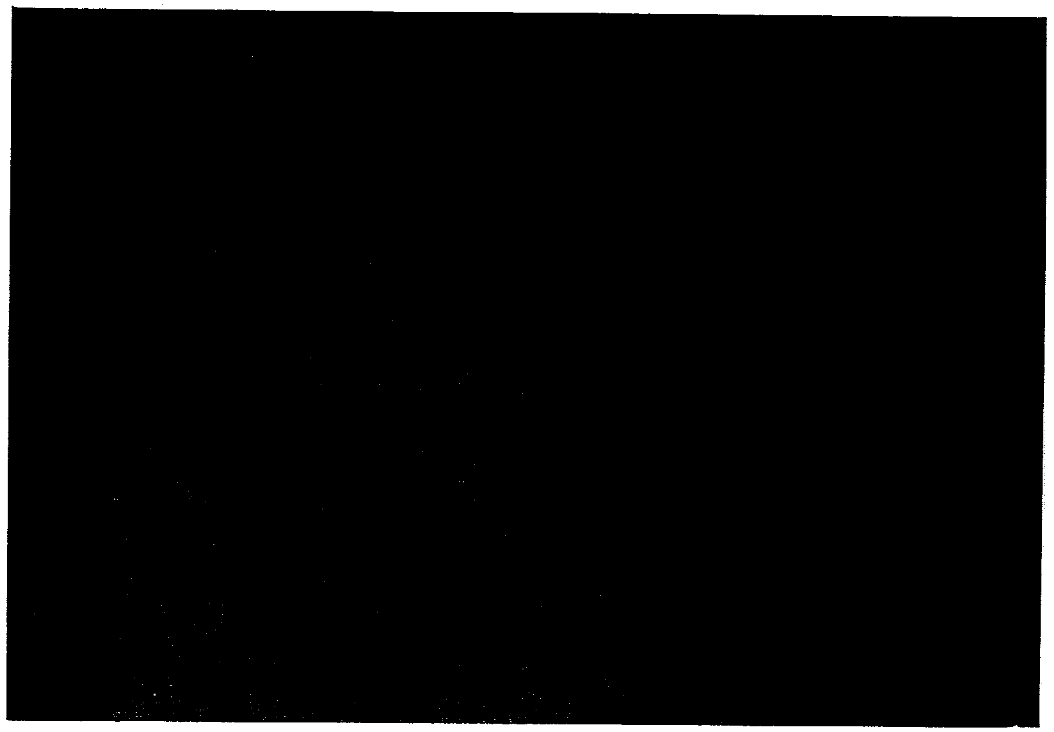


Figure 8 b: *Blow up of Fig. 8 a. Little bubbles arranged in lines consist of fluid inclusions characteristically containing* H_2 *originating from the reaction of the Na-colloids with brine which is incorporated in the growing surfaces. Mag. 216 X*

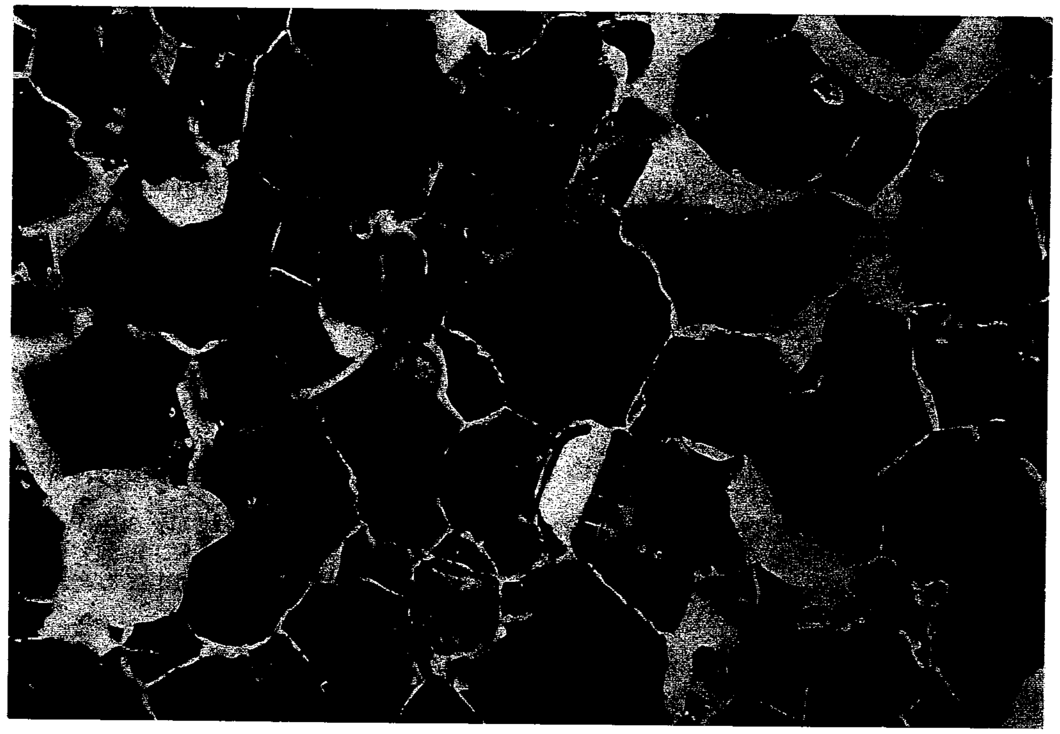


Figure 9: *Characteristic aspect of FAR grown and redecorated grains. Compare the size of the grains with that from Fig.10. NaCl Pressed Powder sample (20PP). Irrad. cond. : 100°C, 15 kGy/h, 44.6 MGy. Mag. 97 X.*

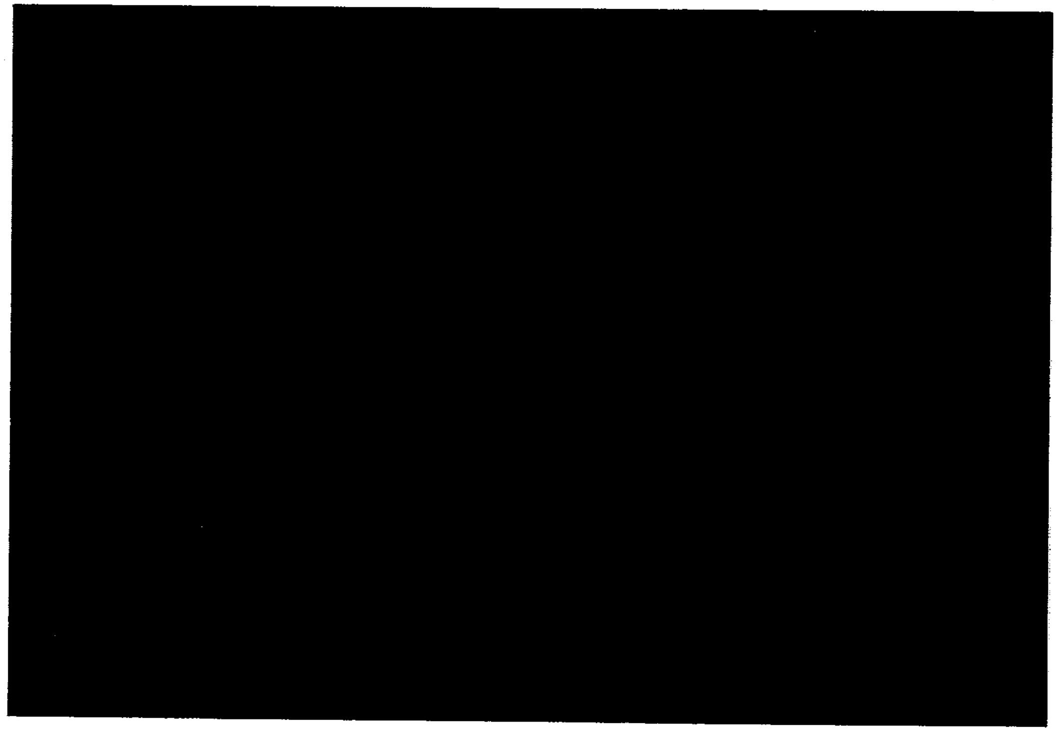


Figure 10 : *Micrograph of the original structure of the NaCl Pressed Powder samples before irradiation. Compare with Fig 9 to see the difference in grain size. Same magnification as Fig.9. Mag. 97 X.*

5. INTERPRETATIONS AND PARTIAL CONCLUSIONS

At a low dose rate (15 kGy/h) and a high temperature (100°C), and in samples not allowed to dilate, mechanical distortion produced by irradiation leads to the development of plastic deformation (creep) structures, even in the absence of an externally applied differential stress. This effect is more important in polycrystals than in monocrystals.

At 15 kGy/h and 100°C the sodium colloids produced by irradiation migrate together with the deformation structures, and are therefore either dragged by the migrating dislocation, or annealed from some places and redeveloped at others, while creep proceeds. The actual process at atomic level is not yet understood.

Anneal coupled to intracrystalline creep processes can justify that the most damaged parts of natural crystals do not contain more stored energy than pure single crystals, at least when irradiation proceeds at a dose rate of 15 kGy/h, at a temperature of 100°C and when the samples are not allowed to dilate.

For high total doses (from 12 MGy on), obtained at a dose rate of 15 kGy/h, a temperature of 100°C, and in samples not allowed to dilate, climb of dislocations leading to recrystallization competes with fluid-assisted recrystallization in eliminating radiation damage. This recrystallization could have been influenced by the presence of OH^- as lattice impurity, but is due to irradiation. An intragranular mechanism implying water-enhanced dislocation mobility could have been active but whether H_2O diffusion plays a major role in anneal of colour centres cannot be quantitatively determined. Note that diffusion of radiation damage defects towards (sub)boundaries can produce the same microstructures and we have not found any quantitative relationship between intragranular bleached areas an water content.

These creep processes show that the behaviour of the rock salt during irradiation is plastic, and suggested that a sort of steady state of radiation damage could be reached if the distorting agent (the radiation) was applied slowly enough (low dose rate) and during long enough times so as to already have produced damage enough (high total doses) for recovery to proceed. Although in themselves these microstructures do not prove that this steady state would exist, their observation drove us to perform very long experiments which did prove that saturation of radiation damage exists for natural rock salts. This means that at lower dose rates, lower values of damage at saturation ought to be found. The radiation damage simulation models also reproduce saturation behaviour [see Donker and García Celma, 1995 a] and the lower values obtained in the experiment as compared to the predictions can be justified by the bleaching associated to creep.

6. GENERAL CONCLUSIONS

The intracrystalline microstructure analyses thus have shown that crystals creep during irradiation and in this way anneal some of the produced radiation damage. On the other hand, the analyses also have shown that nucleation of defect aggregates is easied by the existence of heterogeneities mostly bound to grain boundaries. Impurities, although spatially linked to colloid depleted areas, do not hinder damage development but, are segregated to the places [(sub)grain boundaries] where, due to enhanced diffusion, anneal of colour centres and dislocations takes place. Therefore the white-blue rims at the grain boundaries are not the consequence of hindered colloid nucleation but of differential diffusion. These rims, when unequally developed ought to be the reason for the nucleation of the first bulge for fluid assisted recrystallization.

Fluid-assisted recrystallization already takes place at intergranular brine contents of 0.02 weight %, can take place during irradiation and does not decompose the intergranular brine as long as colloids have not yet developed, can also take place if the brine is in the vapour phase and continues to take place even when the whole sample has already been recrystallized.

It will depend on the rate of volumetric recrystallization and that of colloid development whether brine will or will not be decomposed by recrystallization in a repository. The relative rates of colloid development and volumetric recrystallization deserve more attention.

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