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INFCE/DEP./WG.7/13

GRANITE-REPOSITORY - GEOCHEMICAL ENVIRONMENT

Co-Chairmen/WG7/54
1979-04-20

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Submitted in support of the INFCE
discussions by the Swedish Delegation.

Sweden - Granite Repository - Geochemical Environment

Abstract

Some geochemical data of importance for a radioactive waste repository in hard rock are reviewed. The ground water composition at depth is assessed. The ground water chemistry in the vicinity of uranium ores is discussed. The redox system in Swedish bedrock is described. Influences of extreme climatic changes and of repository mining and construction are also evaluated.

INTRODUCTION

The following paper is a reprint from the report "Handling and Final Storage of Unreprocessed Spent Nuclear Fuel" part II Technical (section 3.5 pp. 111 - 121 plus chapter 3 reference list) issued by the Swedish Project Kärnbränslesäkerhet (KBS) in September 1978.

3.5 CHEMICAL ENVIRONMENT

3.5.1 Groundwater composition

The chemical environment in and around a rock repository for high-level waste is important for the durability of the canisters and the buffer material and for the possibilities of dispersal of the waste substances. These questions can be illuminated with the aid of chemical equilibrium calculations based on the composition of the groundwater and knowledge of the natural occurrence of the materials in question. In some cases, it can be shown that the natural form of occurrence of the material has remained unchanged for many millions of years, thereby providing answers to questions concerning its durability.

An survey of the composition of Swedish groundwater has recently been published by Wenner et al. /3-69/. A large body of data from rockwells in Finland,giving average levels and ranges of variation

Table 3-3. Probable composition of groundwater in crystalline rock at great depth, according to Rennerfelt and Jacks /3-76/.

Analysis	Units	Probable interval	Min. value x)	Max. value x)
Conductivity	µS/cm	400 - 600		1100
pH		7.2 - 8.5		9.0
$KMnO_4$ cons.	mg/l	20 - 40		50
COD_{Mn}	"	5 - 10		12.5
Ca^{2+}	"	25 - 50	10	60
Mg^{2+}	"	5 - 20		30
Na^{+}	"	10 - 100		100
K^{+}	"	1 - 5		10
Fe-tot	"	1 - 20		30
Fe^{2+}	"	0.5 - 15		30
Mn^{2+}	"	0.1 - 0.5		3
HCO_3^-	"	60 - 400		500
CO_2	"	0 - 25		35
Cl^-	"	5 - 50		100 xx)
SO_4^{2-}	"	1 - 15		50
NO_3^-	"	0.1 - 0.5		2
PO_4^{3-}	"	0.01 - 0.1		0.5
F^-	"	0.5 - 2		8
SiO_2	"	5 - 30		40
HS^-	"	<0.1 - 1		5
NH_4	"	0.1 - 0.4		2
NO_2	"	<0.01 - 0.1		0.5
O_2	"	<0.01 - 0.07		0.1

x) The estimated probability that a value will fall between the min. value and max. value is 95 %. Higher values occur locally, see /3-77/ and /3-75, p. 13-17/.

for many components, has been presented by Laakso /3-70/ and other valuable data are supplied by Lahermo /3-71/. Feth et al. /3-72/ present analyses of groundwater from dioritic rocks in California and Nevada. Data from Böhmen are presented by Pačes /3-73/, who also presents groundwater analyses from similar bedrocks from other areas. In this way a good review of conditions in different climates and environments is obtained. The work which has preceded this present report includes analyses of groundwater from Swedish bedrock compiled by Gidlund /3-37/, Rennerfelt /3-74/ and Jacks /3-75/, see table 3-3. Jacks also provides a survey of pertinent chemical equilibria and relevant literature.

3.5.2 Chemistry of groundwater in contact with uranium ores

Like the uranium minerals in most uranium ores, spent fuel is composed of more than 95% uranium dioxide. Knowledge of how the uranium minerals are affected by groundwater can therefore shed light upon the long-range situation in a rock repository. The ores have normally been exposed to the action of the groundwater for many millions of years.

Groundwater which moves through a stratum of permeable sandstone and, on its way, passes a uranium ore which has been formed in the pores of the sandstone has been studied by Cermanov and Panteleyev /3-78/. The groundwater flow is slow, the ore has a large specific surface area and the process has been going on for several million years. The groundwater has been studied in seven sections on its path, see figure 3-8. For each section, the range

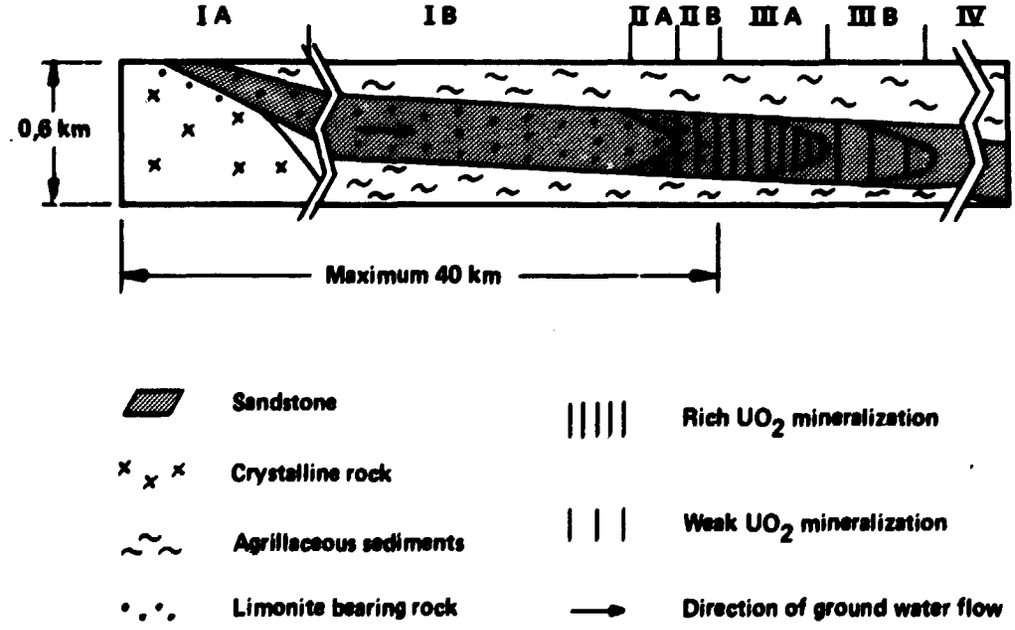

Figure 3-8. Schematic illustration of the geological situation and the different zones around a uranium ore exposed to ground water. (According to Germanov and Panteleyev /3-78/).

of variation of the obtained data is given. An extract is provided in the following table, 3-4. E_h is the groundwater's electrical potential in relation to a hydrogen electrode, i.e. the water's redox potential, given here ir millivolts.

Table 3-4. Chemical groundwater data according to Germanov and Panteleyev /3-78/.

Section zone	Number of samples	Eh mV	pH	Organic carbon mg/l	Uranium µg/l
IA	6	450 - 250	7.0-7.4	1.4 - 1.8	5 - 300
IB	12	300 - 212	7.1-7.7	3.9 - 6.0	21 - 150
IIA	23	250 - 147	6.7-7.7	7.3 -10.4	40 - 720
IIB	9	220 - 57	6.7-7.5	10.4 -16.0	125 - 5000
IIIA	8	-72 --195	7.2-7.7	9.9 - 8.0	3100 - 34
IIIB	12	-165 --212	7.5-7.8	8.0 - 6.9	32 - 3
IV	12	-7 --186	7.6-8.0	6.4 - 5.4	3.0 - 1.6

As is seen in the table, the concentration of uranium in the groundwater increases towards the ore, which shows that uranium is first dissolved from the bedrock. According to the same study, this takes place primarily through the formation of uranyl-carbonate complexes. In the actual ore zone, zone III A, the uranium content of the water decreases sharply, which means that most of the dissolved uranium has precipitated. The existence of the ore deposit in itself shows that the chemical environment leads to precipitation. In the sections downstream of the ore, the uranium levels are very low. The table also shows that the precipitation of uranium takes place in connection with a transition from positive to negative Eh values in the transitional zone between oxidizing and reducing conditions in the groundwater. In the case cited here,this transition takes place at a depth of around 400 m.

Lisitsyn and Kuznetzova /3-79/ have further shown that the groundwater upstream of such an ore area is oxygen-bearing while oxygen is lacking in the water downstream, and that the sandstone upstream is rust-coloured by limonite /$Fe(OH^3)$ and FeO(OH)/, while the sandstone downstream is grey and lacks these compounds of trivalent iron. It is also interesting to note that various microorganisms are found mainly in the actual zone of transition and to some extent also in the rust-coloured limonitic part, but that they are not found, with the exception of nitrogen bacteria, in the unaffected, oxygen-free part of the sandstone located downstream of the uranium ore.

These relationships between the groundwater's uranium content, redox potential and oxygen content are not accidental, but rather determined by chemical laws. They conform to a more general model for the formation of uranium ores in sedimentary rock. In this model, oxygen-bearing water leaches out the low concentrations of uranium in the upper parts of the bedrock by converting it to a hexavalent, easily soluble form. When the water later becomes reducing as a result of reactions with organic material and compounds of bivalent iron, it is no longer able to transport the uranium, which is then precipitated in tetravalent, poorly soluble form. These precipitations can be concentrated to mineable deposits /3-80/. An analysis of transport and precipitation conditions with respect to the stability fields of the various uranium compounds in relation to the pH and Eh of the groundwater has been carried out by Hostetler and Garrels /3-81/. Other contributions have been made by Batulin et al. /3-82/ and Dahl and Hagmaier /3-83/.

In the KBS studies, this has been expanded to analysis of the hydrolysis and redox chemistry of the actinides in water /3-84 and 3-85/ in the laboratory as well as theoretically. This work shows that neptunium and plutonium are reduced to insoluble tetravalent (in the case of plutonium, possibly also trivalent) compounds at higher Eh values than uranium. This means that their occurrence in nature is similar to that of uranium and that their dispersal with the groundwater is prevented by reducing conditions in the same manner, but more easily than in the case of uranium. That such is really the case on a geological time scale is illustrated by the natural reactor at the uranium deposit at Oklo, where transuranium elements, including plutonium, have remained undispersed until complete natural decay /3-86 and 3-87/.

3.5.3 Swedish conditions

The principles and reactions discussed above apply generally. However, the composition of the Swedish groundwater at depth differs in some respects from the Russian groundwater. The sulphate content is generally lower while the pH is somewhat higher. However, these differences do not affect the retention of the uranium and other actinides in the bedrock.

A more fundamental difference lies in the fact that the Swedish bedrock, as well as the prevailing soil strata, possess lower permeability than the sandstones which were studied in the Russian examples, and that the Swedish groundwater systems are less extensive and exhibit lower flow volumes. In addition, the continental glaciation removed older weathering products, which, together with the climatic trend following the ice age, results in the fact that the water under Swedish conditions meets fresh, little-weathered and unoxidized material only a few dm below the surface. The main limonite formation therefore takes place above a depth of one metre, and even at this slight depth, oxidation is severly limited. Such an easily-oxidized material as pyrite has, for example, been found in the form of 0.2-0.6 mm large particles with thin weathering layers of limonite in moraine where the particles have been accessible for weathering for more than 6 000 years /3-88/. Similarly, apparently fresh pyrite is often found in rock fractures near the surface. This means that the transition between oxidizing and reducing conditions which occurred at

a depth of about 400 metres in the Russian example in general occurs right near the water table under Swedish conditions.

Eh values which are positive, but which fall rapidly with increasing depth, have been measured in the groundwater in an esker /3-89/, while Eriksson and Khunakasem /3-90/analyzing some 70 groundwater samples from an esker formation in the Piteå area, showed that conditions there correspond to a reducing environment.

The redox potential in deep groundwater has been studied in water samples from Stripa and the Finnsjö area /3-91/, see table 3-5.

Table 3-5. The redox potential in deep groundwater from Precambrian Swedish bedrock, according to Grenthe /3-91/.

Sample No.	Site	Eh, mV
7	Stripa	-152
8	Stripa	- 31
18	Stripa	-173
19	Finnsjö area	-191
21	Stripa	-210
14	Stripa	- 26
20	Finnsjö area	-157
24	Stripa	-140

In the samples from Stripa, the pH was around 8.2, and in those from Finnsjö, 7.7. The slight deviations exhibited by the values for samples 8 and 14 were probably caused by contamination by small quantities of air. Otherwise, the obtained values show that the redox potential of the Swedish water samples very nearly corresponds to the above-cited Russian values for the nearly uranium-free groundwater which passed through the uranium ore zone. Chemical analysis has also shown that samples of Swedish groundwater from great depth in the KBS test areas have a very low oxygen content /3-74/.

3.5.4 Redox systems in the bedrock

Up to now, only a few determinations of redox potential or oxygen content have been carried out on deeper groundwater in Sweden. However, these results can be accorded wider applicability by considering the redox systems in the bedrock with which the water can react. These are composed primarily of different iron compounds, mainly chloride and similar iron-bearing silicate mine-

rals with layer lattices, generally containing both bivalent and trivalent iron. The redox reaction can be written

$$2FeO_{sil} + H_2O_{sil} = Fe_2O_{3\ sil} + 2H^+ + 2\ e^- ;$$

(sil indicates that the component is part of the silicate structure.) Following /3-92/, Gibbs' free energy for this reaction can be calculated to be 9.7 kcal, which gives

$$Eh(V) = 0.21 + 0.03 \log (Fe_2O_3)/(FeO)^2 - 0.06\ pH;$$

where (Fe_2O_3) and (FeO) stand for the chemical activity of the respective component in the mineral. Mineral analyses show that both FeO and Fe_2O_3 in these minerals can vary between 0.1 and approx. 40% by weight, where the higher value approximately indicates a saturated, solid solution. It is then possible to write

$$Eh(V) = 0.21 + 0.03\ (\log 40 + \log \overline{Fe_2O_3}/\overline{FeO}^2) - 0.06\ pH =$$

$$= 0.26 + 0.03 \log \overline{Fe_2O_3}/\overline{FeO}^2 - 0.06\ pH;$$

where $\overline{Fe_2O_3}$ and $\overline{FeO}$ indicate the concentration of these components in the mineral, expressed as percent by weight. With these concentration limits, the following approximation is obtained:

$$Eh(V) = 0.26 \pm 0.1 - 0.06\ pH;$$

which for pH = 7.7 gives Eh $= -0.2 \pm 0.1$ V, i.e. -200 ± 100 mV.

Iron-bearing chlorite characterizes the rock fractures in all of the KBS test areas. The mineral is, alone or together with other layer-silicates such as biotite, illite and smectite, a normal constituent of the bedrock, and especially its fissures. The deep groundwater, within a pH interval of 7-9, should therefore exhibit an Eh interval between -60 and -380 mV. In limonite-/$Fe(OH)_3$ and FeO(OH)/-bearing fracture zones, however, positive Eh values can be expected. As an example of the prevailing conditions, 18 analyzed samples of fissure-filling layer-silicates from relatively shallow tunnels in Gothenburg /3-93/ can be cited. Their Eh values, calculated for a pH of 7.5 to permit comparisons with previously cited data, fall between -120 and -230 mV. Two samples from borehole 2 at Finnsjö both indicate Eh values near -200 mV.

Other minerals, such as magnetite, pyrite (FeS_2) and pyrrhotite (FeS), which contain bivalent iron, are less common than chlorite, but nevertheless occur, generally in small quantities, in the Swedish bedrock. The sulphides particularly weather easily in oxidizing environments. As a result, they constitute Eh indicators, whose general occurrence also shows that negative Eh values prevail in the bedrock.

Mineralogical and mineral-chemical observations thus show, along with the cited Eh and oxygen determinations, that the groundwater at depth in the Swedish bedrock, aside from locally occurring limonite-bearing fracture zones, is characterized by negative Eh values. In this respect, it is very similar to previously discussed Russian groundwater.

According to the Russian analyses, the uranium content of the water in equilibrium with uranium dioxide lies between 1.6 and 3

micrograms per litre. These values agree well with maximum uranium contents in groundwater calculated from equilibrium constants /3-85/. The average concentration of uranium in Swedish springs and wells is also around 3 micrograms per litre. On the other hand, levels of up to and over 2 milligrammes per litre have been found in shallow rock fractures and under oxidizing conditions, for example in mines with uranium mineralization /3-94/. The groundwater in the Swedish bedrock is therefore not able to dissolve and leach out uranium at great depths. That this is really the case is demonstrated by the fact that the continental glaciation has exposed uranium mineralizations in the Precambrian bedrock which have not been carried away by the groundwater, despite the fact that they were formed more than 1 000 million years ago /3-95 and 3-96/.

3.5.5 Extreme climatic changes

As was mentioned earlier, the climate in Sweden around 30 million years ago was much warmer and more humid than it is today. Plant remains from that time show that nearly tropical conditions prevailed. Changes in the level of the land also led to an intensive and deep groundwater circulation. This has given rise locally to deep weathering, which shows what can happen in extreme cases in our type of crystalline bedrock. The best-studied cases have been found in certain mines /3-28, 3-29/ and represent limonitic alterations which may reach depths of several hundred metres. This represents a transition from magnetite to hematite together with a locally strong development of limonite in connection with severe crush zones in the rock, all of which is a sign of deep and intense oxidation. Without exception, however, it is always found that the oxidation is limited to near the surface, or to the immediate vicinity of such crush zones. At a distance of some metres from the limonite formation, completely intact sulphides are often found, for example PbS and ZnS, which show that the oxidation never reached far out into the surrounding rock. This also indicates that extreme changes in climate, topography and groundwater circulation, which appear extremely unlikely today, would not lead to oxidizing conditions for a waste repository. In such limonite-bearing crush zones, as well as in less extreme cases of limonite-stained fractures, the transitional zone between positive and negative Eh values is situated at the border of the unaltered rock. In general, however, the fracture zones in the bedrock, even at shallow depth, have not become oxidized in this manner, but instead exhibit varying contents of bivalent iron in the layer silicate minerals.

3.5.6 Impact of construction and drainage

So far, we have been discussing the natural, undisturbed chemical environment around a rock repository. However, the actual process of constructing and draining a rock repository causes the groundwater to flow towards the repository and lowers the water table in the form of a funnel-shaped depression, Thunvik /3-45/. As a result, the natural chemical conditions around the repository are altered so that oxidation above the water table reaches greater depth and horizontal extent. At the same time, the rock walls of the repository are aerated. A valuable analysis of such a situation is provided by Pačes /3-97/, who has studied the chemical

conditions at the Svornost uranium mine at Jachymov in Czechoslovakia. Mining operations have been pursued here for several hundred years. The groundwater table is now 300 m below the surface. At the time of the study, the mining operations had reached a depth of about 500 m. From the surface to this depth, the following variations were measured in the groundwater (average values of 9 samples at surface and 5 samples from 480 m):

	At surface	At depth of 480 m
Temperature, ^{o}C	7.1	21
pH	6.5	7.6
Na content mg/l	3.6	95
Eh, mV	+473	-53

Despite the presence of air in nearby mining chambers, the Eh in the inflowing groundwater at the deepest level in the mine is clearly negative. The sulphate concentration (average) at this level is 280 mg/l, which is related to the fact that the surrounding crystalline bedrock is composed of metamorphic rocks, in some cases with considerable purite concentrations. 5 groundwater samples from granite in the same mine at depths of 520-720 m give instead 14 mg/l as an average value for the sulphate content. Despite the fact that the sulphate content at higher levels of the mine is very high, the groundwater remains nearly neutral or weakly alkaline owing to reactions with surrounding silicate minerals.

Of special interest is the fact that the uranium content of the groundwater is given for four samples. Some chemical data on these samples are presented in table 3-6.

Table 3-6. Groundwater data from Svornost uranium mine /3-97/.

No.	Depth m	T C^{o}	Cl^{-} mg/l	HCO_3^{-} mg/l	SO_4^{2-} mg/l	H_2SiO_3 mg/l	pH	Eh mV	U µg/l
20	445	17,6	16,7	143,4	394,6	20,3	6,55	-5	471
21	445	20,0	10,6	183,1	862,9	23,9	7,15	+57	9 000
23	486	23,9	5,3	367,9	54,3	70,2	7,80	-89	76
31	636	29,8	12,4	441,8	15,6	56,2	6,75	-62	17

The two deepest samples, 23 and 31, show how rapidly the Eh and the uranium and sulphate concentrations decrease in the deeper part of the mine. They also provide some idea of the chemical environment in the immediate vicinity of a rock repository which has been dewatered for a very long period of time and of the solubility of uranium dioxide which can be expected in corresponding situations in groundwater of a similarly bicarbonate-rich type as in the Swedish bedrock. The high sulphate contents are,

however, not to be expected in areas with sulphide-poor bedrock of the type which occurs within the areas studied by KBS.

This study shows that the effects of the construction and drainage of a rock repository on the groundwater's redox potential and its capacity to disperse uranium in solution are limited to the vicinity of the rock caverns. Immediately outside the disturbed area, the natural equilibria are restored, and the groundwater cannot cause any substantial dispersal of the uranium. When a rock repository is backfilled and the water table resumes its natural level, the local perturbations also gradually disappear.

3.5.7 Application to a waste repository

In the previous KBS study on the storage of vitrified high-level waste, it was concluded that most of the fission products are retarded so long by sorption in the bedrock that they decay before they reach the biosphere. However, spent fuel contains considerably higher levels of uranium and other actinides, many of which have very long half-lives. It has been shown above that the actinides, even if they are partially present in higher valence states at deposition, would, owing to the natural redox conditions in the bedrock, be precipitated and retained in tetravalent form for millions of years.

In a rock repository, the temperature will be elevated for a limited period of time, whereas the specified chemical equilibria apply at 25°C and atmospheric pressure. It can be shown thermodynamically that the equilibria between the concerned solid phases would not be appreciably shifted if the temperature were increased to 200°C. However, precipitation takes place more rapidly at higher temperatures, as has been demonstrated experimentally /3-98, 3-99 and 3-100/.

The cited examples show that precipitation occurs at the depth and at the pressures which are suggested for a Swedish rock repository. The disturbance of the natural redox conditions caused by the aeration and drainage of the repository have been shown to be of a local nature and negative Eh values have been found immediately outside of similarly aerated mine chambers. Extreme geological disturbances of the natural redox conditions, represented by the limonitic alteration discussed above, are also of a local nature and have remained local for a very long period of time, probably around 25 million years.

In order for it to be possible to apply conditions, observed in nature and explained theoretically and on the basis of experimental data, to a waste repository, the water must be in direct contact with the spent fuel in the same manner as it is with the uranium dioxide in the studied uranium ores. Cited data show that uranium, plutonium and other actinides will not be dispersed to the biosphere with the groundwater despite such direct contact.

However, KBS' storage proposal provides two additional barriers between the fuel and the groundwater, namely a canister and a buffer substance between the canister and the surrounding rock. The copper canister will, among other things, reduce external radiation so that the effects of radiolysis will be virtually negligible. The function and durability of these barriers are dealt

with in chapters II:4 to 6. From a geological point of view, it can be added that metallic copper is a material which is stable in natural groundwater and which has been found in small quantities at various locations in Sweden. A natural deposit of metallic copper together with uranium dioxide has been noted by Welin /3-96/. An interesting observation is that metallic copper proved to be chemically stable over geological periods of time in water with an extremely high salt content, containing, among other solutes, 176 g/l Cl^- and 110 mg/l SO_4^{2-} /3-40/.

With regard to the proposed buffer material (Na-bentonite), an abovecited study of fracture-filling materials from the Gothenburg district and the analytical data given there show that bentonite of the relevant Na-dominant type is a natural constituent of the Swedish bedrock. Groun water chemistry studies /3-75/ cited above show that this material is also generally in chemical equilibrium with the groundwater in a crystalline bedrock of roughly granitic composition.

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