To:

SSM - Swedish Radiation Safety Authority 171 16 Stockholm

SKB has recently issued a new technical report (TR-13-21 Prototype Repository. Hydro-mechanical, chemical and mineralogical characterization of the buffer and tunnel backfill material from the outer section of the Prototype Repository). It contains several statements that are incorrect and misleading as summarized here.

1. **Hydraulic conductivity of HLW buffer**

SKB has performed tests for determining the hydraulic conductivity of clay intended for surrounding the copper-lined canisters. The conductivity of samples taken from the Prototype laboratory and from equally dense, untreated clay have been compared. The problem with these tests is that the extremely high hydraulic gradient (difference in hydraulic head per flow length) changed the material and gave much too low conductivities. The gradient was 10000 meter/meter across the 1.5 cm thick samples meaning that the samples were exposed to a pressure of 150 m water head (1.5 MPa), which is known to give much too low (evaluated) conductivity values (Figs. 1 and 2). The internationally recommended maximal gradient is 30 m/m) and a recently performed by Al-Thaie at the Luleå University of Technology in Luleå and reported in his doctoral thesis (Diss. Dec. 3, 2014) University demonstrates that such high gradients underestimate the conductivity by at least 5 times (Fig.3). This discrepancy does not disqualify the KBS-3V concept but significantly reduces the safety factor of the isolating capacity of the buffer and, in particular, shows that the investigators do not realize and understand the mechanisms associated with percolation of clay materials, which imply that high hydraulic gradients cause consolidation (compression) of the clay sample and erosion of permeable channels causing clogging of them at constrictions, both leading to underestimation of the evaluated (Darcy) conductivity. The investigators' choice of such high hydraulic gradients can be taken as an attempt to deliberately demonstrate that the hydraulic conductivity is at least 5 times tighter than it actually is. If they had used a gradient of 100.000 they would have been able to show that the clay is totally impermeable.

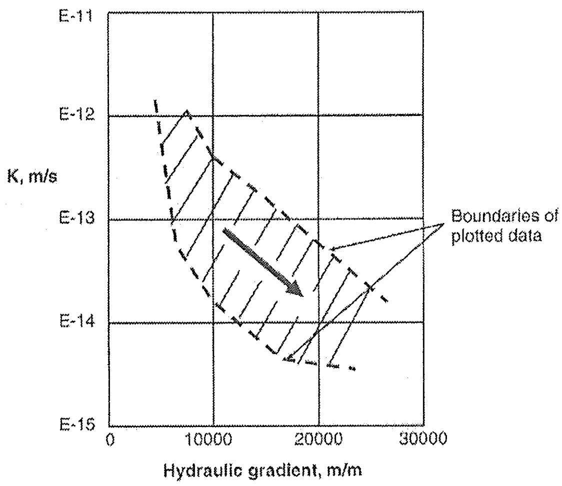

Fig.1. Variation in hydraulic conductivity obrained for for different samples of FEBEX bentonite versus the hydraulic gradient. After Villar, Romero and Lloret.

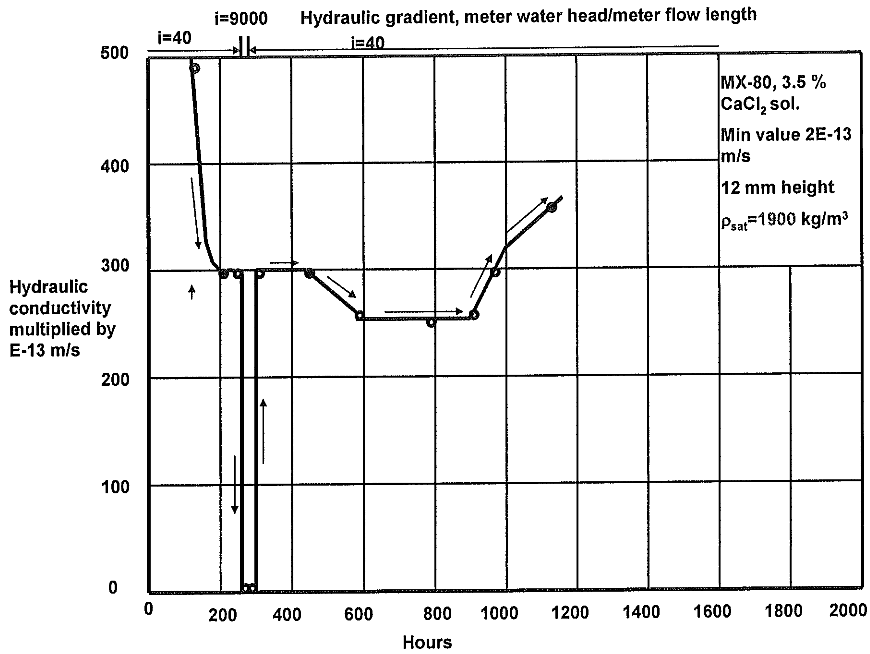

Fig.2. Change in evaluated hydraulic conductivity of MX-80 clay in oedometer percolation tests by changing the hydraulic gradient. The first 250 h represented water saturation and percolation under a hydraulic gradient of 40 meter/meter. The period 250-255 h represents percolation under the gradient 9000 meter/meter, followed by percolation under the gradient 40 meter/meter. Clay density at water saturation 1900 kg/m^3 (Pusch, Yong and Nakano; High-level radioactive waste disposal, WIT Press).

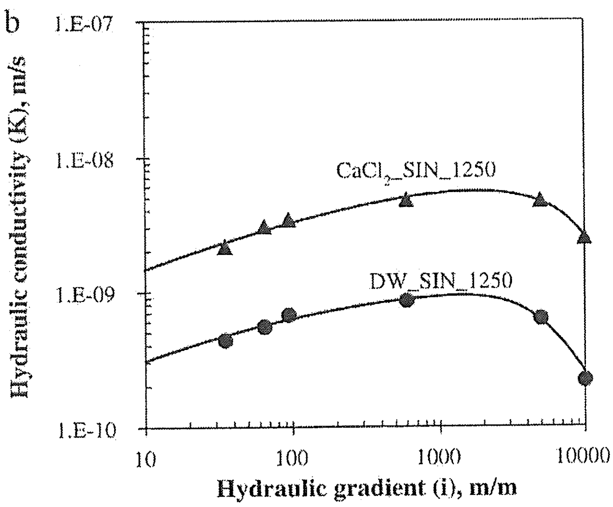

Fig.3. Impact of the hydraulic gradient (K) on the evaluated (Darcy) conductivity for a moderately smectite-rich clay percolated by distilled water (DW) and 3.5 % $CaCl_2$ solution. Dry density 1250 kg/m^3 (Al-Thaie, 2014).

These studies univocally support the generally accepted rule of using significantly lower hydraulic gradients than SKB for getting relevant data of the hydraulic conductivity.

2. Stiffening of HLW buffer

SKB knows well that stiffening of the buffer is the major threat to its function as strain-distributing medium of the canister and to its self-sealing capacity in case of fracturing and loss of contact with the surrounding rock. It is therefore expected that detailed presentation of the specific stress/strain properties would be given sufficient space to demonstrate and discuss the difference between untreated and hydrothermally treated clay, especially from 525-550 mm distance from the Äspö canister center and from equally dense clay at larger distance, all at mid-height canister, but this is not the case. The data are there, in fact, but without adequate interpretation. Thus, Table 2-3, referring to triaxial tests gives for the hydrothermally treated, trimmed sample at 545 mm distance, the peak strength 2700 kPa for 4.3 % strain while the reference clay with the same density had a strength of 2950 kPa at 10.4 % strain. The large difference in strain up to and beyond failure is strong evidence of significant stiffening of the hydrothermally treated clay while the discussion in section 2.3.5 of the report only states that the strain of "the field-exposed material is smaller compared to the specimen of reference material". The tests were too few to make a truly accurate comparison of hydrothermally treated and untreated clay, which makes one suspect that SKB is trying to hide that significant stiffening actually occurred. SKB's report does not show any attempt to derive a practically useful mathematical model for calculating the expected buffer/canister strain in a real repository as function of the buffer density.

SKB knows well that stiffening of the buffer clay with time is the most threatening change that the buffer clay undergoes in a KBS-3V repository because it limits the allowed rock shear displacement that the canisters are allowed to undergo, and because it dramatically changes the self-sealing capacity of the clay caused by such impact and by channeling generated by gas percolation. A number of loading tests made by others were available to SKB when it planned and performed its experiments and they illustrate a significant stiffening by hydrothermal treatment under temperature gradients as illustrated by the graphs in Fig.4.

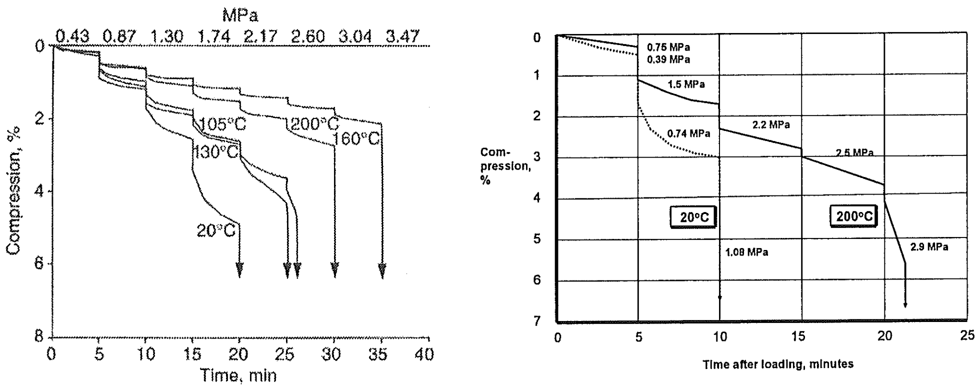

Fig.4 Results of uniaxial load testing of hydrothermally treated MX-80 buffer clay with dry density 1590 kg/m^3 (2000 kg/m^3 at saturation). Left: MX-80; (Pusch/Yong, 2006; Microstructure of Smectite clays…). Right: SWY-1; (Pusch/Yong/Nakano, 2011: High-level Rad. Waste Disposal). The graphs show that heating to about 100°C and more under hydrothermal conditions under a realistic temperature gradient caused stiffening and brittleness in half a year in the lab.

The same mistake concerns the interpretation of the unconfined compression tests and the lack of expressions of the stress/strain/time relationships and parameter data etc. The reader finds the presentation unclear and unnecessary complex but a few data can be used for drawing conclusions of the main differences between (water saturated) samples from the field experiment and from untreated ones (references) with similar density. For reference samples with a dry density of 1550 to 1675 kg/m^3 (1980 to 2055 kg/m^3 at water saturation) the axial strain at failure was 6-8 % while the hydrothermally treated sample close to the canister with a dry density of 1614 kg/m^3 reached the peak strength 3300 kPa at about 4.5 % strain. The discussion in section 2.4.5 gave the conclusion that there were no large differences between the behaviour of reference and hydrothermally exposed samples, which denies the fact - as for the triaxial tests - that there was an obvious and significant increase in stiffness of the latter. This was in fact even more obvious for the curves in Fig. 2-24 copied here.

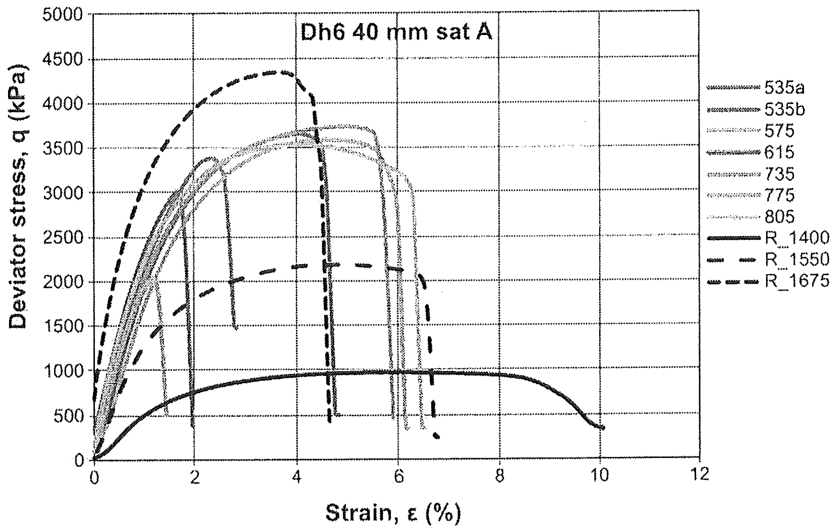

Fig. 5. Unconfined compression tests according to SKB's Fig. 2-24. Most "low-temperature" samples showed ductile behaviour and failed at more than 4 % strain, while the hydrothermally treated ones failed at 1.5 to 3 % and gave steeply rising and – after brittle failure – steeply dropping curves.

Keeping in mind that the temperature was not higher than 85°C and maintained at this level for less than two years one expects that further stiffening will happen in a real repository in the thermal period of many hundred years. The risk of loss of the self-sealing capacity that is required in case of stress-generated fracturing or shrinkage of the buffer, and of critical strain of the canisters caused by the stiff buffer, is obvious (Fig. 6). The SKB authors state that other mechanisms than chemical must have led to the admitted stiffening, which hypothesis brings us to examine the chapter on chemical and mineralogical analyses of the buffer.

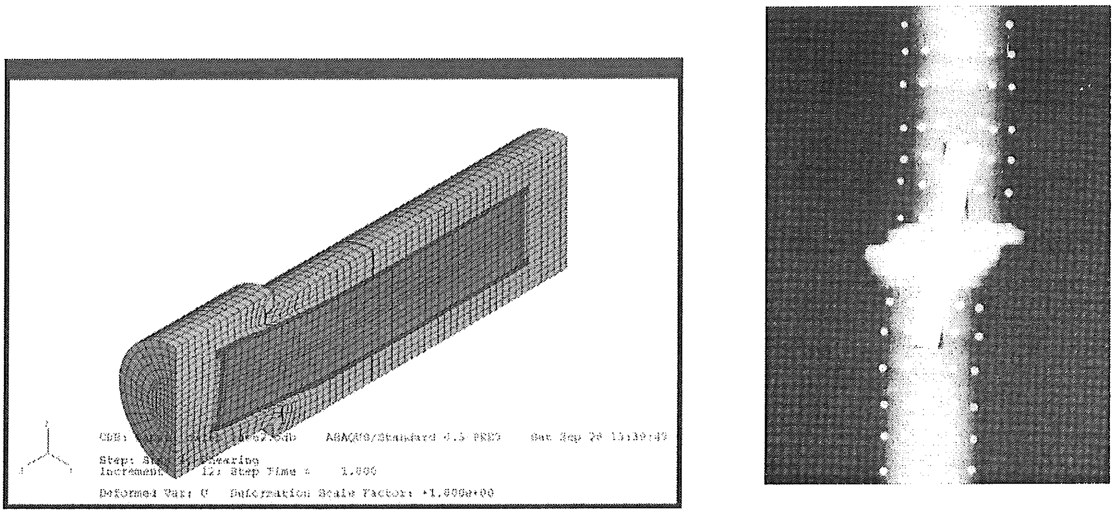

Fig. 6. Strain and displacement of SKB canister. Left: Illustration of FEM-calculated deformation of ductile clay-embedded (yellow) SKB canister (blue) by 0.1 m tectonically induced instant shearing along a fracture intersecting the deposition hole normally to it (Börgesson). Right: Formation of heterogenenities in the clay (black wedges) by shearing of a stiff canister. X-ray image of model test with leadshots in the clay to show the strain pattern. The bright discs in the center is the shear box arrangement (Pusch, Geol. Storage of Rad. Waste, 2008). Density at water saturation about 2050 kg/m^3).

These studies, performed by SKB and various other investigators, univocally support the conclusion that the hydrothermally treated buffer in the Prototype project stiffened considerably and that the allowed instantaneous shearing of 0.1 m will generate unacceptably high canister stresses.

3. Chemical and mineralogical changes of HLW buffer

The SKBs report focuses on comparing the chemical and mineralogical properties of untreated and hydrothermally treated samples of MX-80 clay buffer material from the Prototype. A first and obvious fact is that the clay excavated after 8 years was not water saturated and not uniformly wetted despite artificial wetting that is not described in the report. For the reviewers of the concept, SKB's conclusions concerning the relationship of chemical, mineralogical, and physical performances, especially respecting the transport (hydraulic conductivity) and rheological properties (strength and stress/strain behaviour) would be of interest. Unfortunately, these relationships are nearly absent in the report text, The most important ones would concern the montmorillonite crystal constitution and the formation of precipitates. The firstmentioned would explain changes in hydraulic conductivity and the other the recorded stiffening of the hydrothermally treated clay.

As to chemistry, chloride and sulphate accumulation near the canisters was noticed as expected from SKBs earlier lab tests with wetting of smectitic clay under temperature gradients. The trend of increased Ca content in this zone was expected from earlier experiments while SKB's the finding that Na had increased in the central and colder parts, is in contradiction with other literature-reported MX-80 experiments. Mg exceeded Fe in the octahedral layer according to SKB's report and accumulated in the hottest part in agreement with earlier experiments. Cu was found at 400 µm distance from the canister in the incompletely water saturated clay that was found to be fissured and fractured as observed in a number of previous experiments like the wellkown Czech Mock-up test (Pacovsky). Comparison with such experiments would have greatly improved the lengthy description of various advanced test types that are less informative than determination of the Atterberg liquid limit.

As concerns mineralogy, no adequate XRD data were obtained for the hottest part of the buffer according to the SKB report because of technical difficulties. No alteration of the montmorillonite buffer component is mentioned and no indication of precipitation of silicious or iron-containing complexes. This is in contrast with the findings of other investigators according to which the following observations from hydrothermal tests have been made:

1) TEM-EDX analyses of samples from the clay next to the canister in the wettest hole at Äspö (R8:225) have shown that most of the montmorillonite particles underwent mineralogical alteration from montmorillonite to two different series of mixed-layer series (diVS-ml). Illite with an interlayer charge and K-deficite (<<0.89 per half unit cell) is conventionally termed dioctahedral vermiculite.

2) The chemical composition of the diVS-ml phases is characterized by 1) enrichment of Al in the octahedral layer, and 2) illitization (Fig. 7.). The octahedral substitution of Mg by Al reduced the deficit of charge in the octahedral layer. By dissolution/precipitation processes in the interlayer space tetrahedral Si migrated into the bulk pore solution being replaced by Al, initiating illitization. The two processes take place in chemically dynamic systems as shown by Pusch & Kasbohm (2002) and Herbert et al (2004).

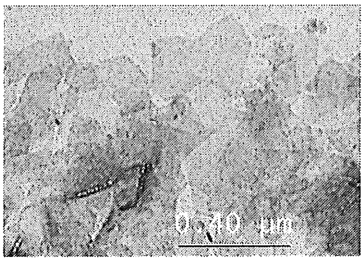

Fig. 7. Partially hypidiomorphic, pseudohexagonal illite-type particles in sample R8:225 (Äspö Prototype project).

3) The diversity of the diVS-ml series indicates obvious trends especially for the cold regions of the experiment. Al followed a decreasing trend with increasing vermiculite/smectite ratio. Fe and Mg behaved reversely with minimum Fe and maximum Mg with a diVerm/smectite ratio of 70/30. The ultimate end members of this process, i.e. dioctahedral vermiculite (=charge- and K-deficient illite) and evolution of the octahedral reconstitution, developed much slower in the hot vicinity of the canister, partly because of a limited degree of water saturation,

4) In contrast to SKB's findings the XRD diagrams for oriented mounts of material from the hottest region there is a small 10 Å peak that is not related to mica-like phases (e.g. muscovite) but to charge- and K-deficient illite,

5) Montmorillonite in the colder parts was mainly unchanged by the hydrothermal impact and the sum of the Na, Ca and Mg contents was nearly the same in the cold ad hot parts. The concentrations of Fe and Al are slightly higher in the cold parts and there was also a weak tendency for the Si-concentration to have risen in the central and colder parts, indicating initiation of cementation by silicification,

6) XRD analyses showed that Ca uptake in the hottest part had altered the typical lamellar spacing of Na montmorillonite from 12.8 to 15 Å. Quartz and cristobalite, being normal constituents in the untreated clay, remained apparently unaltered, while the chloride mineral sinjarite was neoformed in the hottest part,

7) Copper had dissolved and entered the clay especially in the hottest part. The concentration gradient suggests that the diffusion coefficient for migration of copper was on the order of E-12 m^2/s.

SKB's conclusions from its chemical and mineralogical investigations were that the buffer did only undergo insignificant chemical and mineralogical changes by the exposure to hydrothermal condition. These investigations are not representative of the evolution of the buffer in a long term perspective because of 1) the short testing time, 2) the significantly lower temperature than in a real repository (applying Arrhenius-type modelling the 85°C temperature impact is much smaller than that of 100°C), and 3) the incomplete water saturation. Since thermal impact will last for several hundred years the whole concept may have has to be changed so that the maximum temperature of the buffer clay is reduced to 60°C, which is believed to make it survive for a hundred thousand years without significant changes (Fig. 8).

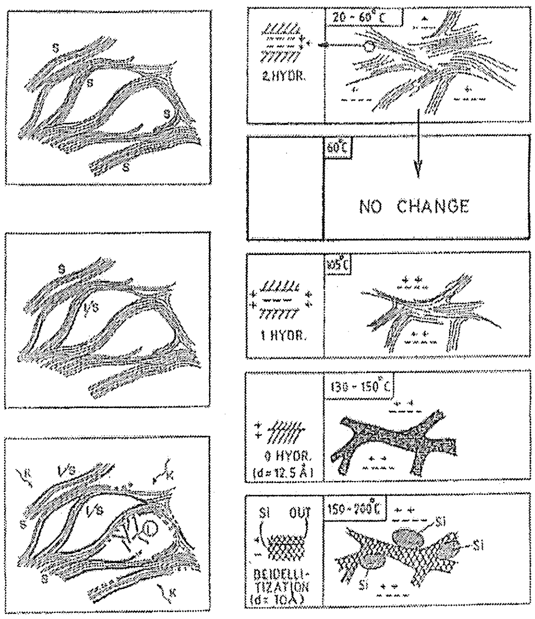

Fig.8. Proposed stages in illitization and silicification of montmorillonite buffer. Left column shows congruent dissolution, conversion to mixed-layer S/I and neoformation of illite. Right column shows heat-induced contraction of smectite stacks, permanent collapse of stacks at a critical temperature, and precipitation of silica (Pusch and Karnland, SKB TR-88-15).

4 Comments

Reconsidering the conclusions from the various rheological and hydraulic conductivity tests should lead to rephrasing and changing of the most essential buffer design criteria:

- If the presently proposed minimum acceptable dry density 1508 kg/m^3 be maintained by SKB, the design value of the hydraulic conductivity would have to be lowered for corresponding to results from testing with a maximum hydraulic gradient of 30-100 m/m using Ca-rich groundwater with the salt concentration met with at 500 m depth. If, on the other hand, SKB maintains the design conductivity E-12 m/s, the density has to be increased, which would naturally raise the swelling pressure,

- As to the rheological properties SKB would have to present their results from triaxial and unconfined compression tests in a clearer form and specify what parameters that can be taken as the basis for calculating the distribution of shear strain of 100,000 years old buffer surrounding canisters of SKB type,

- The fact that the risk of canister failure in a KBS-3V repository is more obvious than estimated by SKB because of the buffer stiffening would make the Very Deep Hole concept (VDH) attractive because of the lower probability of locating canisters where rock displacements can take place Practically all of the upper, 2 km sealed part of a 4 km deep VDH, will be heated to less than 60°C, which makes the risk of mineral conversion to nonexpendables negligible (cf. Fig.8); (Pusch et al, Nat. Science Vol 4, 2012; Pusch, "Bentonite Clay: Environmental Properties and Applications", Taylor & Francis, 2015).

5 Additional

Quick corrosion of the SKB canisters in a KBS-3V repository at Forsmark, generated by electrical potentials in the host rock by the Baltic cable to Finland, has not been thoroughly investigated by SKB. Experience shows that instrumentation installed in certain investigation boreholes at Forsmark were corroded and severely damaged in short periods of time. The crucial matter is that deposition tunnels can short-circuit parallel steep fracture zones that have different electrical potentials. For accurate prediction of such risks, the matter of electrical conductivity of the buffer and backfill needs to be tested in Mock-up tests on bench-scale followed by large-scale experiments. Comprehensive corrosion was for instance observed on the chemical analysing equipment at three occasions in borehole KFM04A according to Nissen et al, 2005 (SKB Rep. ISSN 1651-4416; SKB P-05-265). This study demonstrated the existence of a strong electrical voltage anomaly within and around it and a few more long boreholes intersecting major, water-bearing fractures zones with different electrical potentials. Similar severe corrosion would be caused by clay-filled deposition tunnels. Theoretical models, not yet tested, indicate that the most severe conditions can prevail in the course of the water saturation of the buffer, resulting in electric currents passing through the canisters that are surrounded by wetted clay at their upper and lower ends while the central parts are still dry by heating.

SKB has announced that it will issue a new report on the subject (TR-14-15) and there are reasons to examine it with respect to the described matter. If the model is tested physically and some systematic measurements of the electrical potential in representative boreholes at Forsmark are made one would be able to predict with great confidence what the risk for such canister corrosion really is.

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Roland Pusch

Prof. emeritus; PhD in soil mechanics; PhD in geology

Luleå University of Technology

Geotechnical Division

Private address: St Södergatan 57 A, 22223 Lund

Tel. 046-2111406

Mob. 070-2338132