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## Fluid Migration and Brittle Tectonothermal Evolution in the Central Fennoscandian Shield

- Recorded by Fracture Minerals and Wall Rock Alteration

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recorded by fracture minerals

*On the cover:* A back-scattered electron image of a cubic pyrite crystal (bright) growing together with adularia (grey) on a fracture surface in drill core KFM07A at a borehole length of 882.95 m. The minerals precipitated during the Palaeozoic, sometime between 460 and 277 Ma. The scale bar is 200  $\mu\text{m}$ .

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## ABSTRACT

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The Forsmark area in central Sweden has been investigated as a potential geological host for a final repository of spent nuclear fuel by the Swedish Nuclear Fuel and Waste Management Company (SKB). High quality drill core material from the upper 1 km of the central Fennoscandian Shield has been obtained during the site investigations and has provided a unique opportunity for detailed fracture mineralogical investigations. In this thesis, a multi-analytical approach for recognising events of fluid migration and brittle tectonothermal evolution by analysis of fracture minerals and wall rock alteration is presented. The basis for this study has been the establishment of a relative sequence of fracture mineralisations obtained by investigations of cross-cutting relations and mineral overgrowths. Based on this sequence, representative fracture mineral samples have been selected for further analysis, e.g.  $^{40}\text{Ar}/^{39}\text{Ar}$  dating, stable isotopes, trace element geochemistry and fluid inclusions. Statistical analysis of the orientation of fractures lined with different minerals has also been carried out.

Four major events of fracture mineralisation have been distinguished in the Forsmark area. The two first events are associated with hydrothermal alteration of the wall rock, causing a red-staining due to hematite dissemination. The alteration is characterised by chloritisation of biotite, saussuritization of plagioclase and partial replacement of magnetite by hematite. The oldest event occurred sometime between 1.8 and 1.1 Ga, possibly during a late stage of the Svecokarelian orogeny. Precipitation of epidote, quartz and chlorite occurred at temperatures between ca. 200° and 350°C in preferably sub-horizontal to gently dipping fractures or steep, WNW-ESE to NW-SE fractures.

These fractures are cut by fractures sealed with hematite-stained adularia and albite, prehnite, hematite-stained laumontite, calcite and chlorite which are prominent along steep, ENE-WSW to NNE-SSW and NNW-SSE fractures. These minerals precipitated under hydrothermal conditions at temperatures between 150° and 250°C.  $^{40}\text{Ar}/^{39}\text{Ar}$  dating of fracture filling adularia and K-feldspar fragments in breccias shows that a major event of hydrothermal circulation associated with both reactivation and formation of fractures occurred in the area at 1.1 to 1.0 Ga, probably due to far-field effects from the Sveconorwegian orogeny. This event was followed by a period with some dissolution of fracture minerals.

During the Palaeozoic, sometime between 460 and 277 Ma, fluids emanating from a sedimentary cover rich in organic material migrated downward into the basement, mainly during reactivation of older fractures, but formation of new fractures is also inferred during this period. It is suggested that far-field effects from the Caledonian orogeny and/or the overburden of the Caledonian foreland basin is responsible for this tectonothermal event. The youngest generation of fracture minerals is dominated by clay minerals and thin precipitates of calcite in hydraulically conductive fractures and in the upper part of the bedrock. Minor occurrences of pyrite and goethite have also been found. This event is poorly constrained in time, and precipitation may have occurred episodically from the Late Palaeozoic to the present. These minerals are mainly found in sub-horizontal to gently dipping fractures, inferred as Proterozoic structures. However, some fractures in the upper part of the bedrock may have formed relatively recently due to stress release during e.g. Quaternary deglaciations.

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*Keywords:* Fennoscandian Shield, fracture minerals, hydrothermal alteration, palaeohydrogeology, stable isotopes,  $^{40}\text{Ar}/^{39}\text{Ar}$  dating, Forsmark, nuclear waste repository, Sveconorwegian, Caledonian.

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## PREFACE

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*This doctoral thesis includes a summary and five papers, the papers are:*

- I. Palaeohydrogeology: A methodology based on fracture mineral studies.** Tullborg E-L, Drake H, Sandström B, 2008. Applied Geochemistry 23, 1881-1897. Reprinted with permission from Elsevier.  
*- Tullborg wrote the paper in collaboration with Sandström and Drake. Sandström contributed with results from the Forsmark area, figures, tables and discussion.*
- II. The occurrence and potential origin of asphaltite in bedrock fractures, Forsmark, central Sweden.** Sandström B, Tullborg E-L, de Torres T, Ortiz J E. 2006. GFF 128, 233-242. Reprinted with permission from The Geological Society of Sweden.  
*- Sandström carried out planning, sampling and writing. Tullborg contributed with planning, sampling and discussion and de Torres and Ortiz performed analyses of organic compounds and biomarkers and contributed with knowledge of these.*
- III. Fracture-related hydrothermal alteration of metagranitic rock and associated changes in mineralogy, geochemistry and degree of oxidation: a case study at Forsmark, central Sweden.** Sandström B, Annersten H, Tullborg E-L. 2008. International Journal of Earth Sciences, doi: 10.1007/s00531-008-0369-1. Reprinted with permission from Springer.  
*- Sandström carried out planning, sampling, writing and microanalysis (SEM-EDS and LA-ICPMS) and microscopy. Annersten contributed with Mössbauer analysis and discussion and Tullborg with planning, sampling and discussion.*
- IV. Episodic fluid migration in the Fennoscandian Shield recorded by stable isotopes, rare earth elements and fluid inclusions in fracture minerals at Forsmark, Sweden.** Sandström B, Tullborg E-L. Resubmitted to Chemical Geology after revision.  
*- Sandström carried out planning, sampling, writing, microanalysis and microscopy. Tullborg contributed with planning, sampling and discussion.*
- V. Brittle tectonothermal evolution in the central Fennoscandian Shield as recorded by paragenesis, orientation and  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology of fracture minerals at Forsmark, Sweden.** Sandström B, Tullborg E-L, Larson SÅ, Page L. Submitted to Tectonophysics.  
*- Sandström carried out planning, sampling, writing, microscopy and SEM-EDS. Tullborg contributed with planning, sampling and discussion, Larson with discussion and Page with  $^{40}\text{Ar}/^{39}\text{Ar}$  analysis and discussion.*

*Other related contributions by the author during the Ph.D. programme which are not included in the thesis are presented in the appendix at the end of the summary.*

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## POPULÄRVETENSKAPLIG SAMMANFATTNING

ett område strax sydost om Forsmarks kärnkraftverk, cirka 12 mil norr om Stockholm, har omfattande platsundersökningar genomförts mellan 2002 och 2007 inför valet av en plats där ett slutförvar av använt kärnbränsle kan byggas. Dessa undersökningar har genomförts av Svensk Kärnbränslehantering AB (SKB) och har omfattat provborringar ner till cirka 1 kilometers djup i berggrunden. Enligt SKBs metod skall kärnbränslet isoleras på 500 meters djup nere i urberget och eftersom det använda kärnbränslet bör vara isolerat i cirka 100 000 år så är en stabil berggrund en viktig förutsättning för lokaliseringen av ett slutförvar. Ett vanligt vetenskapligt sätt att uppskatta framtida förändringar är att studera händelser tillbaka i tiden. Genom att studera sprickor i berggrunden och de mineral som fällt ut i dessa så kan man ta reda på när berget har spruckit upp och sprickor har reaktiverats (spruckit upp igen) och vilka förhållanden som rådde i berget vid dessa händelser.

Mer än 200 prover har valts ut från nästan 18 kilometer borrhävar under arbetet med denna avhandling. Genom att studera hur sprickor med olika sprickmineral korsar varandra har en sekvens av olika gamla generationer av sprickmineral byggts upp. Utifrån denna sekvens har prover valts ut för vidare analys med olika metoder. Bland annat har kalifältspat valts ut för radiometrisk datering med hjälp av  $^{40}\text{Ar}/^{39}\text{Ar}$  metoden. Denna metod bygger på att en naturligt förekommande radioaktiv isotop av kalium ( $^{40}\text{K}$ ) sönderfaller till argon med en känd hastighet (s.k. halveringstid). Genom att mäta kvoten mellan moderisotopen kalium-40 och dotterisotopen argon-39 i provet, så kan en ålder räknas ut. Vidare har sprickmineral (kalcit och pyrit) analyserats med avseende på förhållandet mellan stabila isotoper av syre, kol och svavel. Denna typ av analyser kan ge information om vilka förhållanden som rådde då mineralen föll ut i sprickan, såsom temperatur, varifrån kolet kom (t.ex. från organiskt material eller från det omgivande berget) och om man haft mikroorganismer närvarande.

Fyra större händelser av sprickmineraliseringar har urskiljts: 1) De äldsta sprickmineraliseringarna bildades någon gång för mellan 1,8 och 1,1 miljarder år sedan vid en temperatur av 200 till 350°C. Detta kan ha hänt under ett sent skede av en omfattande bergskedjebildning i området (den Svekokarelska), för knappt 1,8-1,7 miljarder år sedan. 2) För 1,1 till 1,0 miljarder år sedan cirkulerade varma lösningar (runt 200°C) i berggrunden i Forsmark samtidigt som äldre sprickor reaktiverades och nya bildades och stora mängder sprickmineral föll ut. Detta skedde sannolikt som ett resultat av påverkan från en stor bergskedjebildning (den Svekonorvegiska) som framförallt omfattade västra Skandinavien. Under denna tid omarbetades och veckades stora delar av berggrunden i t.ex. sydvästra Sverige. Efter denna period skedde en viss upplösning av äldre sprickmineral. 3) Någon gång för mellan 460 och 277 miljoner år sedan trängde lösningar ner i berggrunden från överliggande lager av sedimentära bergarter. Dessa lösningar var tydligt influerade av organiskt material och rester av detta material har hittats som asfaltit (oljeliknande substans) i sprickor ner till cirka 150 meters djup i dagens berggrund. Huvudparten av dessa lösningar hade en temperatur på 60°-100°C, även om de lokalt kunde vara varmare. Lösningarna cirkulerade framförallt i äldre sprickor men en viss nybildning av sprickor skedde också. Troliga anledningar till denna aktivitet i berggrunden är effekter från bildandet av den skandinaviska fjällkedjan i väster (Kaledoniderna) och då en omfattande bassäng med bl.a. vittringsprodukter från Kaledoniderna bildades öster om bergskedjan. 4) De yngsta sprickmineralen återfinns i vattenförande och i ytnära sprickor. Det är oklart när dessa mineral har fällt ut och det är möjligt att det skett vid olika episoder från för 277 miljoner år sedan fram till idag. Det har framför allt skett i gamla, sedan länge existerande, sprickor och zoner i berget.

Resultaten som presenteras i denna avhandling visar att berggrunden i Forsmark, även ur ett geologiskt tidsperspektiv, är mycket stabil. De flesta sprickorna bildades för mer än 1000 miljoner år sedan. Reaktivering och viss nybildning av sprickor har också skett under senare perioder (Paleozoikum). Utfällningar av sprickmineral i vattenförande sprickorna sker fortfarande om än i mycket blygsam skala. En mindre del subhorisontella sprickor, framför allt i de övre delarna av berggrunden, kan ha bildats senare vid t.ex. tryckavlastning efter de sista istiderna i området.

## 1. INTRODUCTION

Site investigations for a potential geological host for a final repository of spent nuclear fuel have been carried out by the Swedish Nuclear Fuel and Waste Management Company (SKB) at two locations in Sweden; the Forsmark area in central Sweden and the Laxemar/Simpevarp area in south-eastern Sweden (Ström et al. 2008). During these investigations, drilling into the Fennoscandian basement has provided high quality drill core material from the upper 1 km of the continental crust. Due to the long life span of a final repository (100,000 years), stable bedrock is one of the most important prerequisites for site selection. A common scientific approach for predicting future changes is to look back into the past. Fracture minerals can provide a record of past tectonothermal events in the brittle regime.

The fracture mineralogy investigations were initiated in order to obtain knowledge of the low- to moderate-temperature geological evolution in the Forsmark area (Fig. 1). In this thesis, the results of detailed investigations of fracture mineralogy and wall rock alteration in the Forsmark area, ca. 120 km north of Stockholm are presented. This includes the formation and reactivation of fractures, events of fluid migration (palaeohydrogeology) and fluid-rock interaction throughout the geological evolution. In addition, data from the fracture mineralogical studies are important for the modelling of migration and retardation of radionuclides after a possible leakage from the repository and for modelling of oxygen consumption in the fracture system during recharge of oxygenated waters in response to e.g. the construction of a repository or during a future glaciation (e.g. Puigdomenech et al. 2001).

Fractures form when the bedrock responds in a brittle manner to stress. Brittle deformation occurs in the upper crust where the temperature is below ca. 300-350°C, in typical continental crust with a geothermal

gradient of 20-25°C/km, this approximates to the upper 15 km (Davis and Reynold 1996). Precipitation of fracture minerals occurs when a fluid present in fractures becomes oversaturated. This often occurs due to changes in fluid characteristics such as chemistry, temperature, pressure, pH or Eh. Such changes can be promoted by processes such as fluid mixing, water-rock interaction, fracturing and subsequent drop in hydrostatic pressure or circulation and cooling or heating of fluids in a hydrothermal system. Prevailing conditions control precipitation of minerals, thus these can be used as indicators of past hydrogeochemical conditions.

Calcite is a common fracture-filling mineral and precipitates under a wide range of conditions, from high-temperature hydrothermal to low-temperature groundwater circulation. Calcite is also known to respond quickly to changes in temperature and chemistry of the fluid, making it one of the most suitable minerals for palaeohydrogeological investigations.

Fracture mineralogical investigations aiming at characterising palaeohydrogeological events have been carried out at several locations in both crystalline and sedimentary rocks (e.g. Larson and Tullborg 1984, Lindblom 1987, Wallin and Peterman 1999, Blyth et al. 2000, Budai et al. 2002, Juhász et al. 2002, Neymark et al. 2002, Drake and Tullborg 2009).

In this thesis, the brittle tectonothermal evolution and different events of fluid migration in Forsmark have been deciphered using a multi-analytical approach. The work has contributed to the understanding of the stability of the crystalline bedrock at the Forsmark site. The basis of the work has been the establishment of a relative sequence of fracture mineralisations based on cross-cutting relations and overgrowths. Based on this sequence, samples have been selected for geochemical analyses including stable isotopes, major and trace elements, biomarkers, fluid inclusion and radiometric dating.

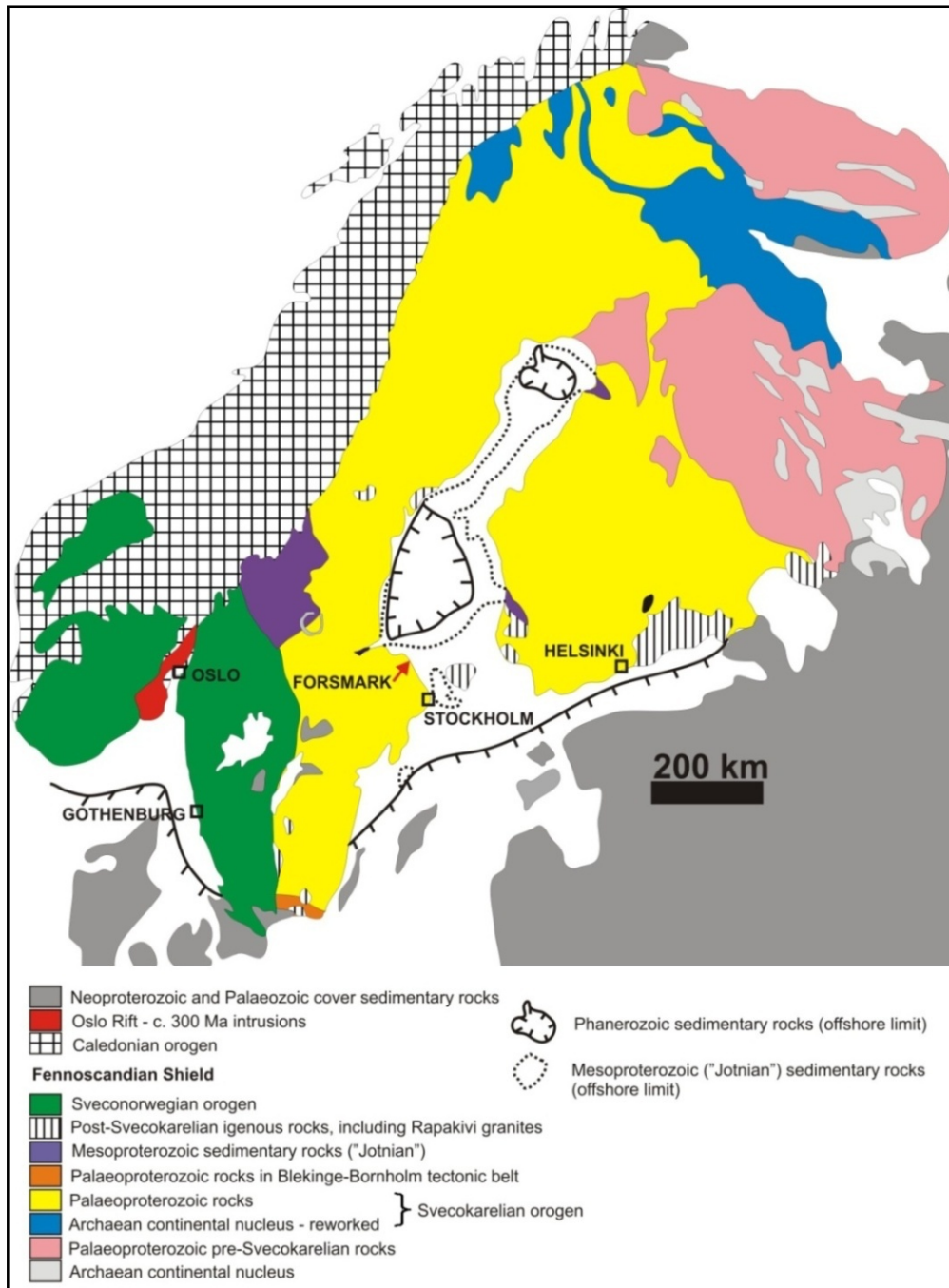


Fig. 1. Major tectonic units in the Fennoscandian Shield. The location of the Forsmark area is indicated by the arrow. The map has been modified after Winterhalter (1981) and Koistinen et al. (2004).

## 2. FINAL REPOSITORY OF NUCLEAR FUEL IN SWEDEN

The site investigations carried out by SKB in Sweden are aimed at finding a suitable location for a final repository of spent nuclear fuel that requires no monitoring by future generations. The spent nuclear fuel consists mainly of uranium dioxide. The objective is to isolate the spent

nuclear fuel for 100,000 years, which is the time it takes for the fuel to reach a radioactivity level that is not harmful to the environment.

The repository is based on the KBS-3 method (SKB 1983) where the spent nuclear fuel will be isolated at a depth of 500 metres in crystalline bedrock. The repository is based on a multiple barrier principle (Fig. 2). The first barrier is the copper canister in which the fuel is encapsulated. The canister is



embedded in a bentonite clay buffer; the second barrier. The third barrier is the bedrock which should protect the canister from mechanical damage and provide an environment free of dissolved oxygen and low water flow rates. The work presented in this thesis contributes to the understanding of the stability of the third barrier; the bedrock.

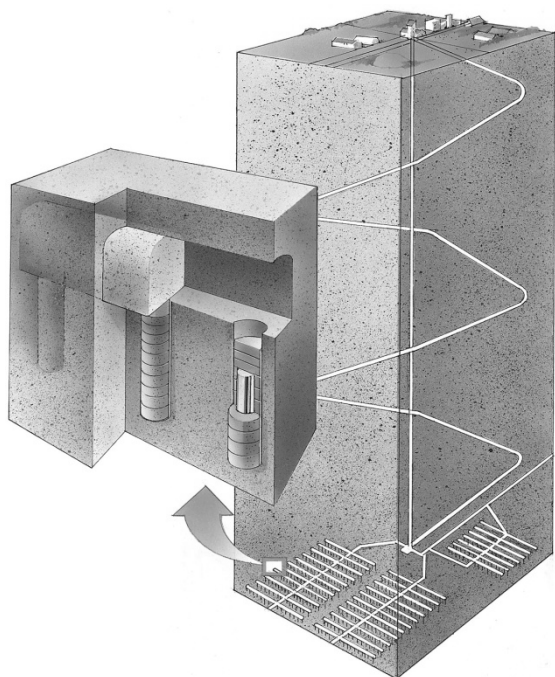


Fig. 2. Illustration of a final repository of spent nuclear fuel according to the KBS-3 method. Copper canisters with encapsulated spent nuclear fuel are embedded in a bentonite clay buffer at a depth of 500 meters in crystalline bedrock. Copyright SKB 2009, illustrator Mats Jerndahl.

### 3. GEOLOGICAL SETTING

Forsmark is located on the shoreline of the Baltic Sea, ca. 120 km north of Stockholm in central Sweden (Fig. 1). The area belongs to the Svecofennian domain, a major part of the Fennoscandian Shield (Koistinen et al. 2001). The Forsmark area is dominated by two suites of calc-alkaline meta-igneous and granitic rocks (Fig. 3) which intruded the area at 1.89 to 1.87 Ga and 1.87 to 1.85 Ga, respectively (Hermansson et al. 2007, 2008). Metamorphism occurred at amphibolite facies with a main phase of penetrative ductile deformation between 1.87 and 1.86 Ma

during the Svecokarelian orogeny (Hermansson et al. 2008). After 1.85 Ga, ductile deformation was restricted to discrete zones along the already present high-strain belts in the region. The present erosion level cooled below 300°C between 1.73 and 1.67 Ga, and it is inferred that the bedrock started to respond in a brittle manner between 1.8 and 1.7 Ga (Söderlund et al. 2008). Mesoproterozoic sediments (“Jotnian”) were deposited on the Fennoscandian Shield sometime between 1.5 and 1.27 based on dating of sub-Jotnian Rapakivi granites (Andersson et al. 2002) and post-Jotnian mafic intrusions (Söderlund et al. 2005a). These sediments probably covered the Forsmark area. During the Sveconorwegian orogeny between 1140 and 920 Ma, the bedrock in western Scandinavia was heavily reworked (Bingen et al. 2008, Fig. 1). Associated with this orogeny, a foreland basin was formed, covering a large part of the Fennoscandian Shield (Larson et al. 1999). At the end of the Neoproterozoic, a sub-cambrian peneplain had developed in the Forsmark area which largely corresponds to the present bedrock surface (Lidmar-Bergström 1996). A sequence of Cambrian to Silurian sediments was then deposited on this peneplain, the thickness of this sequence was probably less than 500 metres in central Sweden (Gee and Sturt 1985). Docking of Baltica with Laurentia between ca. 510 and 400 Ma resulted in the formation of the Caledonian orogeny in western Scandinavia (Gee and Sturt 1985; Roberts 2003) (Fig. 1). A Caledonian foreland basin developed during the Devonian (Larson et al. 1999, 2006). The thickness of these sediments in the Forsmark area has been estimated to ca. 2-3 km (Cederbom et al. 2000). Most of these sediments were eroded during the later part of the Palaeozoic (Cederbom et al. 2000), but a thin sedimentary cover persisted until the Cenozoic (Cederbom et al. 2006, Söderlund et al. 2005b). During the Quaternary, the Forsmark area has been characterised by alternating cold glaciations and warm interstadials (Fredén 2002).

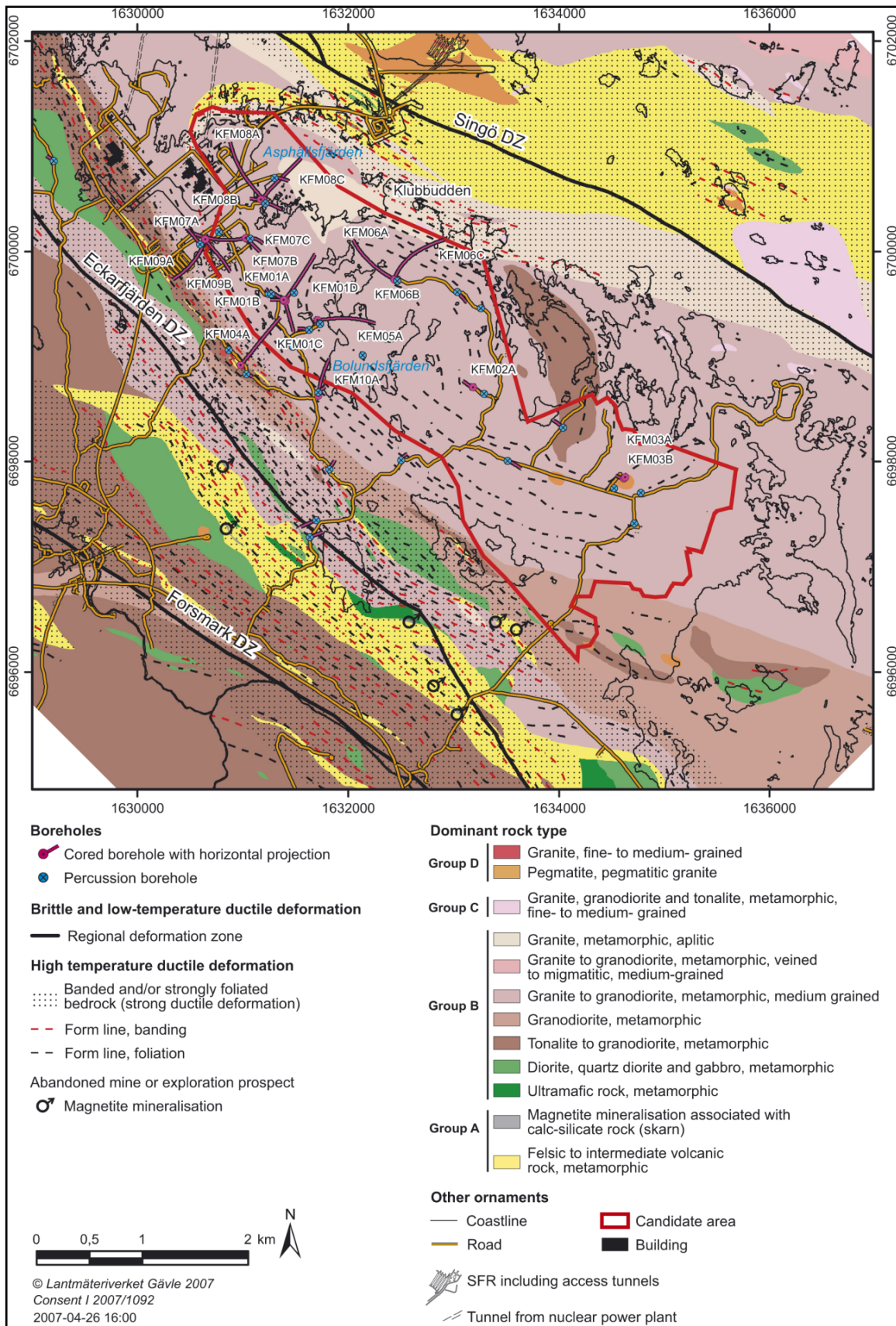


Fig. 3. Bedrock geological map of the Forsmark site with projections of the sampled boreholes, adapted from Stephens et al. (2007).

At present, four major groundwater types are found at the Forsmark site: The upper ca. 150 metres of the bedrock are highly transmissive and characterised by an abrupt change from fresh, dominantly meteoric water in the upper 30 to 100 metres, to brackish marine water. The brackish marine water is dominated by components from the Littorina Sea and glacial melt water from the last deglaciation. At greater depths, brackish to saline non-marine waters dominate (Laaksoharju et al. 2008, Smellie et al. 2008).

## 4. METHODOLOGY

More than 200 fracture samples have been selected from 22 drill cores. The total length of these drill cores is nearly 18 km. The locations of the boreholes are shown in Fig. 3. The basis for fracture mineralogical studies is the establishment of a relative sequence of fracture mineralisations based on cross-cutting relations and overgrowths. The relative formation of fracture minerals has been established in every sample. From this data, a more general sequence of fracture minerals has emerged. However, a complete record of events can only be found in the ideal case since periods of non-saturated fluid and dissolution can leave out or even erase records of past fluid migration and brittle tectonothermal events. Studies of the relative order of fracture mineralisations have mainly been carried out using standard polarising microscope, binocular microscope and scanning electron microscope.

A common problem encountered when working with fracture minerals, especially from drill cores, is the small sample volumes. The fracture surface intersected by the borehole represents a very small part of the actual fracture surface. Therefore, the established sequence serves as an important basis for the selection of representative samples for e.g. stable isotopic analysis and radiometric dating. Short summaries of the main analytical methods used in this thesis are presented below.

### 4.1 SEM-EDS

Mineral identification was aided by analysis with a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). SEM-EDS was also used for studies of the textural relationships between different generations of fracture minerals.

### 4.2 Stable isotopes

The stable isotopic composition of fracture minerals can be used to recognise past hydrogeochemical conditions. Stable isotopic ratios of oxygen, carbon and sulphur are often presented as deviation ( $\delta$ ) in per mil (‰) units in relation to a given standard, which are defined as follows using oxygen as an example:

$$\delta^{18}\text{O} = \left[ \frac{{}^{18}\text{O}/{}^{16}\text{O}_{\text{sample}}}{{}^{18}\text{O}/{}^{16}\text{O}_{\text{standard}}} - 1 \right] \times 10^3$$

The  $\delta^{18}\text{O}$  value in a fracture mineral depends on the composition of the formation fluid and the temperature. Thus, if the formation temperature is largely known, the  $\delta^{18}\text{O}$  value of the formation fluid can be estimated using fractionation factors. Fluid-rock interaction can also have a large influence on the  $\delta^{18}\text{O}$  value. The  $\delta^{13}\text{C}$  value is less temperature dependent than  $\delta^{18}\text{O}$  and the influence from fluid-rock interaction in crystalline bedrock is often negligible due to its low C content. Instead, the  $\delta^{13}\text{C}$  value mainly reflects the source of the dissolved  $\text{CO}_2$  which enters the fracture mineral during precipitation. The  $\delta^{34}\text{S}$  value can be used to separate magmatic and hydrothermal pyrite from pyrite precipitated due to the bacterial or thermochemical reduction of sulphate. The strontium isotopic composition is expressed as the  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  ratio and since Sr does not fractionate significantly during calcite precipitation and calcite does not accommodate  ${}^{87}\text{Rb}$ , the radioactive parent of  ${}^{87}\text{Sr}$ , in its crystal lattice, the measured  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  ratio in calcite is a good indicator of the  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  ratio in the formation fluid (McNutt 2000). Since the fraction of  ${}^{87}\text{Sr}$  steadily increases in rocks and fracture minerals containing Rb, higher  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  ratios in calcite may reflect younger ages of

the fluid from which it precipitated (Faure and Mensing 2005).

### 4.3 Trace elements in calcite

Trace elements in calcite can be used as indicators of formation temperature, redox conditions and dominating complexing agents (Möller and Morteani 1983, Tullborg et al. 1999).

### 4.4 $^{40}\text{Ar}/^{39}\text{Ar}$ dating

The age of a fracture mineral is not necessarily the age of the fracture, although precipitation can occur syngenetically with fracturing. More often, precipitation occurs during reactivation of an already present fracture or during fluid flow in a fracture kept open due to the prevailing stress situation. Thus, radiometric ages obtained from fracture minerals only provide a minimum age of fracture formation. However, cross-cutting relations of several generations of fracture minerals can also provide a maximum age for a fracture. A combination of cross-cutting relations and radiometric dating of fracture mineral can thus constrain the age of fracturing of the bedrock.

$^{40}\text{Ar}/^{39}\text{Ar}$  dating can be applied on K-bearing minerals. The method is based on the radioactive parent  $^{40}\text{K}$  which decays to 89.52%  $^{40}\text{Ca}$  and 10.48%  $^{40}\text{Ar}$  with a half-life of  $1.250 \times 10^9$  years. In the  $^{40}\text{Ar}/^{39}\text{Ar}$  method, stable  $^{39}\text{K}$  is transformed to  $^{39}\text{Ar}$  by neutron activation in a nuclear reactor by neutron capture and proton emission. Since  $^{39}\text{Ar}$  has a short half-life (269 years), the amount of  $^{39}\text{Ar}$  in the mineral before irradiation can be neglected. The portion of  $^{39}\text{K}$  which has been converted to  $^{39}\text{Ar}$  provides a measure of the original K content in the mineral. The  $^{40}\text{Ar}/^{39}\text{Ar}$  ratio is measured by a mass spectrometer during step-heating of the mineral by a laser. From the released  $^{40}\text{Ar}$  and  $^{39}\text{Ar}$  during each step, an age can be calculated. The  $^{40}\text{Ar}/^{39}\text{Ar}$  age is obtained from a plateau of similarly aged steps (Faure and Mensing 2005). Different minerals have different closure temperature for Ar diffusion, the mineral adularia which has been dated in this thesis has a closure

temperature of 150-250°C (McDougall and Harrison 1999).

### 4.5 Fluid inclusions

Fluid inclusions are fluids trapped within minerals during crystallisation. Observations of the ice melting and fluid homogenisation temperatures during repeated freezing and heating of the inclusions under a microscope provide information on the salinity and temperature of the fluid during crystallisation (Roedder 1984).

### 4.6 Biomarkers

Biomarker analysis is based on chromatography of organic material and is used to characterise certain organic sources and provide information of secondary processes like burial and later alteration (Meyers 2003).

### 4.7 Orientation of fractures

Fracture orientation data obtained from boreholes need weighting to ensure that the data are representative for the fracture network and independent of the borehole orientation (Terzaghi 1965). Fracture orientation data are recorded during the BIPS-based (Borehole Image Processing System) drill core mapping carried out by the on-site geologists as part of the site investigation programme and have been extracted from the SKB database Sicada. The Sicada database contains more than 57,000 fractures from the drill core mapping at the Forsmark site.

### 4.8 Geochemical analyses of altered wall rock

For characterisation of geochemical changes in altered wall rock, it is important to compensate for changes in mass and/or volume during alteration. This has been done by normalisation based on the assumption that certain elements are immobile and therefore conserved during the alteration (Gresens 1967, Grant 1986). Whole rock geochemistry was analysed by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectroscopy) and ICP-MS

(Inductively Coupled Plasma Mass Spectrometer). In situ analyses were carried out by SEM-EDS and Laser Ablation ICP-MS. The degree of oxidation was measured by Mössbauer analysis.

## 5. SUMMARY OF PAPERS

### 5.1 Paper I

*Palaeohydrogeology: A methodology based on fracture mineral studies.* Tullborg E-L, Drake H, Sandström B, 2008, *Applied Geochemistry* 23, 1881-1897.

In this paper, a methodology is presented for palaeohydrogeological studies based on fracture minerals in crystalline bedrock. The methodology presented in this paper has generally been followed during the fracture mineral studies at the Forsmark site, although some adjustments have been made due to site specific prerequisites. The method focuses mainly on solving variations in the depth of the fresh/saline water interface and the detection of the near-surface redox front. The scheme suggested for solving the variations in fresh/saline water interface depth focuses on fracture filling calcite and includes methods such as analysis of  $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ ,  $^{87}\text{Sr}/^{86}\text{Sr}$ , trace element geochemistry and cathodoluminescence (CL). Redox front investigations should include studies of the distribution of redox sensitive minerals (e.g. pyrite, Fe-oxyhydroxides), U-series analyses, trace element analysis of fracture fillings (e.g. Ce, Fe, Mn),  $\text{Fe}^{3+}/\text{Fe}^{2+}$  analyses and Fe isotopes. It is also concluded that the methodology is site-specific and depends on the geological and hydrogeochemical evolution in the area.

### 5.2 Paper II

*The occurrence and potential origin of asphaltite in bedrock fractures, Forsmark, central Sweden.* Sandström B, Tullborg E-L, de Torres T, Ortiz JE. 2006. *GFF* 128, 233-242.

Asphaltite, a bitumen, has been found in fractures in the upper part of the bedrock at Forsmark. In this paper, analyses of stable carbon isotopes and biomarkers are applied to elucidate its origin and time of precipitation. A Late Cambrian to Early Ordovician alum shale which probably

covered the Forsmark area during this period is suggested as the most likely source rock based on similarities in carbon isotopes and biomarker composition. Downward migration of fluids emanating from this organic-rich sediment during the Palaeozoic is suggested, due to overpressure in a sedimentary overburden during the development of the Caledonian foreland basin.

### 5.3 Paper III

*Fracture-related hydrothermal alteration of metagranitic rock and associated changes in mineralogy, geochemistry and degree of oxidation: a case study at Forsmark, central Sweden.* Sandström B, Annersten H, Tullborg E-L, 2008. *International Journal of Earth Sciences*. doi: 10.1007/s00531-008-0369-1.

Fluid circulation during the Proterozoic resulted in red-staining of the wall rock adjacent to fractures due to hydrothermal alteration. In this paper, it is shown that the alteration is characterized by chloritisation of biotite, saussuritisation of plagioclase and partial hematization of magnetite. The red-staining is due to hematite dissemination within and along the grain-boundaries of the altered plagioclase. Geochemical analysis of whole rock samples and in situ techniques (SEM-EDS, Laser Ablation ICP-MS) show that the changes on the whole rock scale were limited during the alteration and are mainly manifested by enrichment of  $\text{Na}_2\text{O}$  and volatiles and depletion of  $\text{CaO}$ ,  $\text{FeO}$  and  $\text{SiO}_2$  in the altered rock. However, the element mobility was more extensive with intra- and inter-granular migration on the microscale, of e.g. Ca, K, Na, Al, Si, Ba, Rb, Sr, Ti and REEs. An increase in the degree of oxidation can be seen as partial hematization of magnetite and hematite dissemination in plagioclase. However, the change in degree of oxidation between fresh and altered rock is not significant at the  $1\sigma$  confidence level.

#### 5.4 Paper IV

*Episodic fluid migration in the Fennoscandian Shield recorded by stable isotopes, rare earth elements and fluid inclusions in fracture minerals at Forsmark, Sweden. Sandström B, Tullborg E-L. Resubmitted to Chemical Geology after revision.*

In this paper, the stable isotopic composition, REE geochemistry and fluid inclusions in fracture filling calcite and pyrite have been investigated in order to reveal past events of fluid migration in the central Fennoscandian Shield. Four major events of fluid migration are distinguished, representing precipitation of fracture minerals under conditions ranging from high-temperature hydrothermal to present day groundwater circulation. The first event caused precipitation of hydrothermal epidote, quartz and chlorite at temperatures above 200°C during the Proterozoic. This event was followed by another event of hydrothermal circulation of fluids and precipitation of mainly hematite-stained adularia and albite, prehnite, laumontite, calcite and chlorite at temperatures between 150° and 280°C, probably due to far-field effects from the Sveconorwegian orogeny around 1.1 to 1.0 Ga. Precipitation was probably promoted by fluid-rock interaction. During the Palaeozoic, an organically influenced fluid migrated downward into the basement and caused precipitation of mainly quartz, calcite, pyrite and asphaltite. Precipitation occurred at temperatures between 60° and 190°C (mainly <100°C), probably promoted by mixing of a fluid emanating from an overlying organic-rich sedimentary cover and a deep basinal brine. This was probably due to the development of the Caledonian foreland basin. Far-field effects from the Caledonian orogeny in western Scandinavia probably also influenced the area. Microbial activity has also been inferred in the fractures during this period. The youngest event of fluid migration is associated with precipitation of clay minerals and calcite with minor occurrences of pyrite and goethite in fractures in the upper part of the bedrock and in hydraulically conductive fractures. Precipitation of these fracture minerals have probably occurred episodically over a long

period, possibly from the Late Palaeozoic to the present.

#### 5.5 Paper V

*Brittle tectonothermal evolution in the central Fennoscandian Shield as recorded by paragenesis, orientation and  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology of fracture minerals at Forsmark, Sweden. Sandström B, Tullborg E-L, Larson SÅ, Page L. Submitted to Tectonophysics.*

The focus of this paper is  $^{40}\text{Ar}/^{39}\text{Ar}$  geochronology of fracture-filling adularia and analysis of the orientation of fractures lined with different fracture minerals. The basis of the work is the establishment of a relative sequence of events of fracture mineralisation. The different parageneses were characterised and adularia samples were subsequently selected for  $^{40}\text{Ar}/^{39}\text{Ar}$  dating for geochronological constraints on the sequence. The events of fracture mineralisation have been correlated to major tectonic events in the Fennoscandian Shield. Formation and reactivation of different fracture sets are also addressed. The first generation of fracture minerals (epidote, quartz and chlorite), is preferably found in sub-horizontal to gently dipping fractures and in steep, WNE-ESE to NW-SE fractures. Precipitation occurred between 1.8 and 1.1 Ga, possibly during a late stage of the Svecokarelian orogeny close to 1.8-1.7 Ga. The second generation of fracture minerals consists of hematite-stained adularia and albite, prehnite, laumontite, calcite and chlorite and is preferably found along steep, ENE-WSW to NNE-SSW and NNW-SSE fractures. Precipitation occurred around 1107 to 1034 Ma during reactivation and formation of fractures, probably due to far-field effects from the Sveconorwegian orogeny. A period with some dissolution of fracture minerals followed. During the Palaeozoic, precipitation of mainly quartz, calcite, pyrite and asphaltite occurred, preferably along older Proterozoic fracture sets, but formation of new fractures is also inferred during this period. Far-field effects from the Caledonian orogeny or the development of the Caledonian foreland basin are the likely reasons for this event. Clay minerals and calcite of the youngest

generation of fracture minerals are prominent along sub-horizontal to gently dipping fractures but are also found in steeply dipping fractures. Most of these fractures are Proterozoic but some of the sub-horizontal fractures in the upper part of the bedrock may have formed recently, due to stress release during loading and unloading cycles of sedimentary covers and glaciations.

## **6. SYNTHESIS OF RESULTS**

This thesis shows how detailed investigations of fracture minerals and a multi-analytical approach can successfully be applied to distinguish and characterise events of fluid migration and brittle tectonothermal evolution in crystalline bedrock. The establishment of a relative sequence of fracture mineralisations is the basis for the selection of representative samples for e.g. stable isotopic analyses, fluid inclusion thermometry (Paper I) and radiometric dating.

Four different generations of fracture mineralisations have been distinguished in the Forsmark area. The generations represent precipitation during a wide range of conditions, from hydrothermal conditions during the Proterozoic to possibly recent low-temperature groundwater circulation. Fluid-rock interaction during the two oldest events of fracture mineralisations caused red-staining of the wall rock due to hematite dissemination. The main features of this wall rock alteration are chloritisation of biotite, saussuritisation of plagioclase and partial hematisation of magnetite. The geochemical changes on the whole rock scale were limited and the increase in degree of oxidation (seen as partial hematisation of magnetite and hematite dissemination) was not significant at the 1 $\sigma$  confidence level (Paper III).

The events of fluid migration and associated fracture mineralisations distinguished are summarised below, mainly based on Paper II, IV and V.

### **6.1 Late Svecokarelian event?**

The first event of fracture mineralisation occurred between 1.8 and 1.1 Ga, under conditions close to the ductile-

brittle transition. The fracture mineral paragenesis consists of epidote, quartz and chlorite (Fig. 4-1). The semi-ductile character of cataclasites sealed with these minerals suggests that precipitation occurred close to 1.8-1.7 Ga, when the bedrock started to respond in a brittle manner (Söderlund et al. 2008). Epidote is normally found at temperatures above 200°C, (Bird and Spieler 2004) and the ductile-brittle transition is around 300-350°C (Davis and Reynold 1996), constraining the formation temperature to 200-350°C for this fracture mineral paragenesis, possibly during a late stage of the Svecokarelian orogeny. The first generation of fracture minerals is preferably found along sub-horizontal to gently dipping fractures but also in steep WNW-ENE to NW-SE fractures. This event of fluid circulation was associated with a high degree of fluid-rock interaction as evidenced by the alteration of the wall rock seen adjacent to fractures (Paper III).

### **6.2 Sveconorwegian event**

Fractures sealed with epidote, quartz and chlorite are cut by fractures sealed with hematite-stained adularia, albite, prehnite, laumontite, calcite and chlorite (Fig. 4-2). These minerals formed under hydrothermal conditions at temperatures between 150° and 250°C, probably around 200°C. Based on <sup>40</sup>Ar/<sup>39</sup>Ar ages of fracture-filling adularia and reset K-feldspar fragments in breccias, it is inferred that a major event of fluid circulation occurred around 1107 to 1034 Ma, probably due to far-field effects from the early phases of the Sveconorwegian orogeny. The event was associated with a high degree of fluid-rock interaction which caused alteration of the wall rock (Paper III). These minerals are preferably found along steep ENE-WSW to NNE-SSW and NNW-SSE fractures. It can not be determined with certainty whether all fracture minerals of this generation precipitated during a single event or if fluid circulation during a late phase of the precipitation caused resetting of the Ar-system. However, it is evident that a major event of fluid circulation and reactivation and formation of fractures occurred during this

period. A period of minor dissolution of fracture minerals occurred after precipitation of these minerals.

### 6.3 Palaeozoic event

During the Palaeozoic, sometime between 460 and 277 Ma as evidenced by  $^{40}\text{Ar}/^{39}\text{Ar}$  dating of fracture filling adularia, an organically influenced fluid migrated down into the crystalline basement. Migration occurred along already present Proterozoic structures, but formation of new fractures is also inferred. Minerals precipitated during this event are mainly found in ENE-WSW to NNE-SSW and sub-horizontal to gently dipping fractures, but they are also found along other steep fracture sets. The fluid is suggested to have emanated from an overlying sequence of sedimentary rocks, including an organic-rich Late Cambrian-Early Ordovician alum shale (Paper II). The dominant minerals precipitated from this fluid are quartz, calcite, pyrite and asphaltite (Fig. 4-3). Minor occurrences of analcime, galena, fluorite, adularia, barite and sphalerite are also found. Precipitation occurred at a temperature between 60-190°C, although most minerals precipitated at temperatures below 100°C. Microbial reduction of sulphate in the presence of organic material has also been inferred based on stable carbon isotopic composition in calcite precipitated during this period. Caledonian fracture fillings have also been recognised in south-eastern Sweden (Drake et al. 2009), suggesting that this event of fluid migration occurred on a regional scale.

### 6.4 Late Palaeozoic to possibly recent event

The youngest generation of fracture minerals is dominated by clay minerals and thin precipitates of calcite in near-surface and hydraulically conductive fractures (Fig. 4-4). Minor occurrences of pyrite and goethite are also found in these fractures. These minerals are found along mostly Proterozoic fractures and zones. However, some sub-horizontal fractures in the upper part of the bedrock filled with these minerals may have formed recently due to stress release during loading

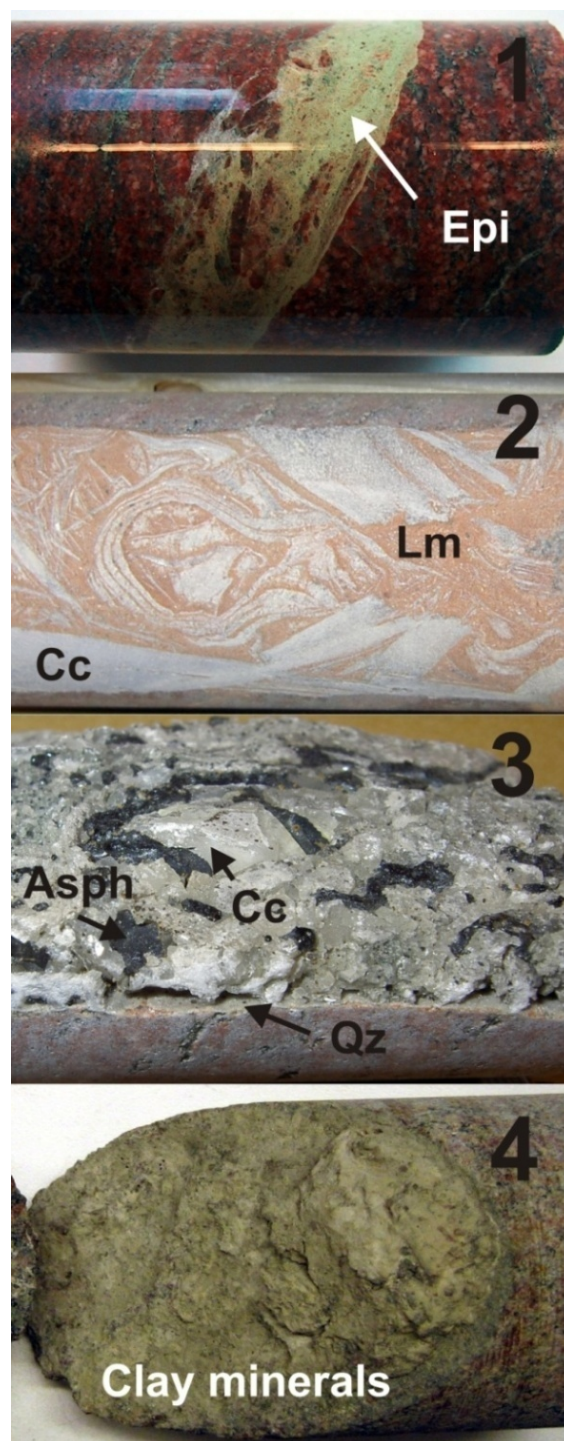


Fig. 4. Drill core samples representing the different generations of fracture mineralisation. 1) Epidote-sealed cataclasite (Epi). 2) Lanmontite (Lm) and calcite (Cc) from the Sveconorwegian event. 3) Quartz (Qz), calcite (Cc) and asphaltite (Asph) precipitated during the Palaeozoic. 4) Clay minerals from the last event of fracture mineralisation in a hydraulically conductive fracture.



and unloading cycles of sediments during the Mesozoic or during the Quaternary glaciations.

## 7. CONCLUDING REMARKS

The results presented in this thesis show that the majority of the fractures in the crystalline bedrock at Forsmark are older than 1.0 Ga. Substantial reactivation of these structures and also formation of new fractures occurred during the Palaeozoic prior to 277 Ma. Since then, formation of

new fractures has been limited although formation of some fractures, especially in the upper part of the bedrock may have formed during later episodes of loading and unloading cycles related to sedimentary episodes during the Palaeozoic to Mesozoic and Quaternary glaciations. A schematic time scale with the different events of fracture mineralisation and major tectonothermal events influencing the central Fennoscandian Shield is shown in Fig. 5.

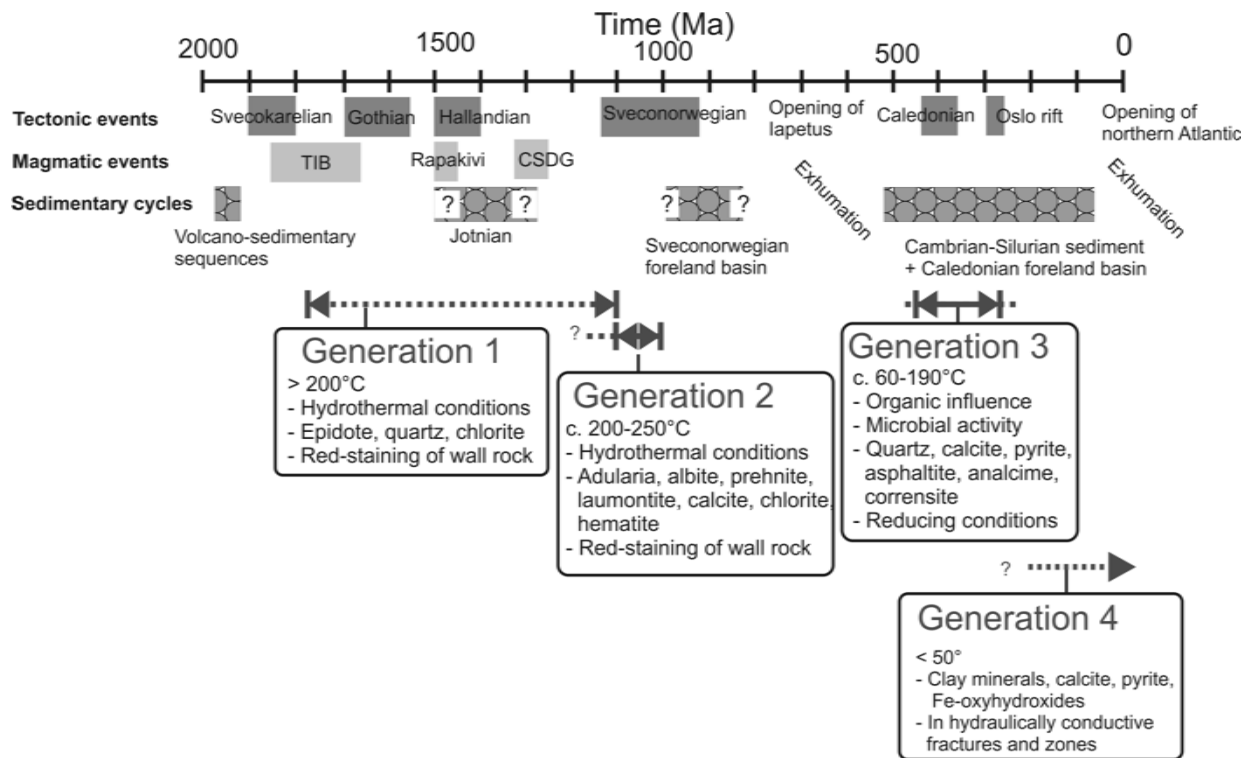


Fig. 5. Schematic time scale summarising the main events of fracture mineralisation and their formation conditions at Forsmark together with major tectonothermal events the central Fennoscandian Shield. Ages of the tectonothermal events have been compiled from Hubbard (1975), Connelly and Åbäll (1996), Åbäll and Gower (1997), Christoffel et al. (1999), Larson et al. (1999), Cederbom et al. (2000), Andersson et al. (2002), Roberts (2003), Gorbatschev (2004), Haapala et al. (2005), Söderlund et al. (2005a, 2005b, 2006), Möller et al. (2007) and Bingen et al. (2008).

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## 10. APPENDIX

*Other related contributions by the author not included in the thesis:*

*Peer-reviewed short paper:*

Sandström B, Tullborg E-L, 2007. Paleohydrogeological events in Forsmark, central Sweden, recorded by stable isotopes in calcite and pyrite. In: T.D. Bullen & Y. Wang (eds), *Water-rock Interaction: Proc. 12<sup>th</sup> Intern. Symp. on Water-Rock Interaction*, Kunming, China, 31 July - 5 August 2007, Taylor & Francis Ltd, 773-776.

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