Applied Geochemistry 76 (2017) 196–209

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Quaternary redox transitions in deep crystalline rock fractures at the western margin of the Greenland ice sheet☆

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ARTICLE INFO

Article history:
Received 3 June 2016
Received in revised form
29 November 2016
Accepted 1 December 2016
Available online 2 December 2016

Keywords:
Redox transitions
Fracture coatings
U-series isotopes
Greenland ice sheet
Nuclear waste disposal

ABSTRACT

When planning for long term deep geological repositories for spent nuclear fuel knowledge of processes that will influence and change the sub-surface environment is crucial. For repositories in northern Europe and similar areas, influence from advancing and retreating continental ice sheets must be planned for. Rapid transport of meltwater into the bedrock may introduce oxic conditions at great depth, which may affect the copper canisters planned to encapsulate the spent fuel. The lack of direct observations has led to simplified modelling assumptions not reflecting the complexity of natural systems. As part of a unique field and modelling study, The Greenland Analogue Project, of a continental ice sheet and related sub-surface conditions, we here present mineralogical and U-series data unravelling the Quaternary redox history in the deep bedrock fracture system close to the margin of the Greenland ice sheet. The aim was to increase the understanding of circulation of potentially oxygenated glacial meltwater from the surface down to 650 m depth.

Secondary mineral coatings were sampled from open fractures in cored boreholes down to 650 m, within and below the current permafrost. Despite continental ice sheet coverage and/or prevailing permafrost during large parts of the last 1 Ma, measured disequilibrium in the ^{238}U-^{234}U-^{230}Th system shows that water has circulated in the bedrock fracture system at various occasions during this time span. In fractures of the upper 60 m, infiltration of oxygenated surface water has resulted in a prominent near-surface ”oxidised zone” with abundant FeOOH precipitation. However, this zone must be relict because it is currently within permafrost and the U-series disequilibrium signatures of most fracture coatings show evidence of deposition of U prior to the Holocene and even prior to the last glaciation maximum which occurred less than 100 ka ago. This U deposition is found both within and below the near surface “oxidised zone” indicating temporal redox variation within this zone during the last 1 Ma. Potential Holocene leaching of U is indicated by $^{230}\text{Th}/^{238}\text{U} \gg 1$ and close to secular equilibrium for $^{234}\text{U}/^{238}\text{U}$ in some of the near surface fractures and also in a couple of deeper fractures. Indicated U-leaching in the talik within the last 200 ka is proposed to be the result of talik-related discharge of water with a capability of keeping U in solution. Circulation of oxidative water in the deep system beneath the permafrost is indicated only in a few fractures and solely by U-series disequilibrium ($^{230}\text{Th}/^{238}\text{U}$ activity ratios up to 2.97 at 431 m depth), probably due to restricted, perhaps sporadic infiltration of oxidative water, potentially during the Holocene. In these fractures, the conditions have in general been more reducing than in the near surface system where oxidising conditions have prevailed and penetration of oxygenated waters may have been continuous.

1. Introduction

Several countries are planning for deep geological repositories designed to keep spent nuclear fuel isolated from mankind and the environment for more than one hundred thousand years (e.g.

☆ This paper has been recommended for acceptance by Philippe Negrel.

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http://dx.doi.org/10.1016/j.apgeochem.2016.12.001

Gunnarsson et al., 2006). Within this time frame areas in e.g. northern Europe, will experience climate induced changes such as the growth of ice sheets and permafrost will influence and change the sub-surface environment, including its hydrology and hydrochemistry, and thus influence repository safety. Due to lack of direct observations of such processes from areas with existing ice sheets, models of glacial influence on the hydrology/hydrochemistry have to-date been based on simplified and conservative assumptions, which do not reflect the complexity of natural systems and processes. Hydrogeological models indicate that glacial meltwater can intrude to considerable depth (up to several hundreds of metres) in crystalline bedrock during glacial advance and retreat and components of glacial meltwater have been documented in the deep bedrock aquifers in the Baltic shield; e.g. Laxemar/Äspö (Laaksoharju et al., 2009) and Forsmark (Smellie et al., 2008) in Sweden and Olkiluoto in Finland (Pitkänen et al., 2004) in accordance with hydraulic modelling (Vidstrand et al., 2010). Some theoretical models (Glynn et al., 1999) suggest that the glacial water may have relatively high content of dissolved oxygen due to its low temperature and high pressure below the ice. Rapid transport of glacial water into the bedrock can therefore introduce oxic conditions also at great depth, which may lead to oxidation of the copper canisters planned to encapsulate the spent nuclear fuel and in the worst case also oxidise the U(IV) in the fuel. There are, however, models showing that oxygen will be consumed along the flow pathways or already in the overburden (Guimerà et al., 1999). In order to increase the understanding of processes associated with glaciations/deglaciations and their impact on the long term safety of a deep geological repository, the Greenland Analogue Project (GAP), a field and modelling study of the Greenland ice sheet and sub-surface conditions, was initiated collaboratively by the Swedish Nuclear fuel and Waste Management Co. (SKB, Sweden), Posiva Oy (Finland) and Nuclear Waste Management Organization (NWMO, Canada). The study site at the margin of the western Greenland Ice Sheet, east of Kangerlussuaq, is an appropriate analogue of the conditions that are expected to prevail in much of Canada and Fennoscandia during future glacial cycles (Harper et al., 2016; Claesson Liljedahl et al., 2016). Pioneering *in situ* investigations of some of the parameters and processes needed to achieve a realistic understanding of how an ice sheet may impact a deep repository have been conducted. Although the Kangerlussuaq area is reasonably well investigated from a glacial hydrological point of view (e.g. Mernild et al., 2012), the permafrost hydrology and sub-permafrost groundwater system are not well known. To increase the knowledge of sub-surface processes deep drilling and downhole surveys were performed.

Groundwater sampling is very challenging in these boreholes due to extensive permafrost and because water conducting fractures below the permafrost can only be accessed by running hot sample tubes through the permafrost, which will give a short time slot for sampling. These circumstances do not allow potentiometric redox measurements to be carried out. Furthermore, analyses of redox sensitive elements like Fe, Mn and S may also be hampered by drilling water contamination and by the disturbance created by the borehole itself in this pristine environment. Instead, fracture coatings with different stored markers of water-rock interaction can be used to overcome the shortages of the groundwater studies. Fracture coatings have been successfully used to provide information e.g. about hydrologic responses to climate changes (Paces et al., 2010, 2013) and about redox conditions and transitions in deep groundwater aquifers (MacKenzie et al., 1992; Perez del Villar et al., 2002; Drake et al., 2009; Dideriksen et al., 2010).

In the present study, fracture coatings were sampled from three cored boreholes. These boreholes were drilled with the aim to define the depth of permafrost at the ice sheet margin and to sample deep groundwater beneath the ice sheet and permafrost, as well as to confirm the presence of a talik structure beneath a lake (a layer of year-round unfrozen ground in permafrost areas, SKB, 2010). A set of open fractures from the ground surface down to 650 m depth was analysed, including both fractures in bedrock within current permafrost and hydraulically conductive fractures below the permafrost. The aim was to gain information about redox transitions in the bedrock fractures, and consequently about the depth of intrusion of oxygenated waters into the bedrock aquifer. Emphasis was on redox sensitive minerals providing indication of oxidising conditions (Fe-oxydroxides; FeOOH, and Mn-oxide/hydroxides) or reducing conditions (e.g. pyrite), and on redox sensitive elements Ce, Mn and U supplemented with U-series disequilibrium technique (USD). Because U is a redox sensitive element its mobilisation from fracture surfaces can indicate a change in redox conditions within the time-scale provided by the half-lives of the ^{238}U-decay chain nuclides ^{238}U, ^{234}U and ^{230}Th (MacKenzie et al., 1992; Osmond and Ivanovich, 1992; Suksi, 2001; Gascoyne et al., 2002). It is important to keep in mind that the different elements studied do not reflect the same redox interval, e.g. oxidation of U can occur at conditions where Fe does not oxidise, i.e. U can exist in its oxidised form also at mildly reducing conditions, especially when occurring as carbonate complexes. Moreover, because redox fronts may fluctuate spatially and temporally it is important to use several independent parameters to obtain a conclusive model of the redox conditions. Here we use, for the first time in a permafrost region, a methodology proven useful for redox front studies in other crystalline rock settings, e.g. Laxemar, Sweden (Drake et al., 2009). At that site, a redox transition front was detected at c. 20 m, but a few deeper indications of oxidizing conditions also existed in highly conductive fractures (Drake et al., 2009). This study increases the understanding about deep groundwater flow, and potential intrusion of oxygenated glacial meltwater to great depth, e.g. beneath permafrost, in crystalline rock fractures.

2. Setting and borehole locations

The region around Kangerlussuaq, located in the dry continental area of central west Greenland is dominated by the ice sheet, but with an ice-free coastal zone of 200 km. This zone comprises a hilly tundra landscape with limited vegetation and a large portion of exposed bedrock. Bedrock ridges and stream valleys trending E-NE to W-SW dominate. These major valleys are sub-parallel to the ice movement and provide a continuous drainage system from the ice sheet to the fjords. Bedrock depressions are normally filled with sandy till and fresh, striated bedrock surfaces are common (Aaltonen et al., 2010). The highest summits close to the ice margin are about 600 m.a.s.l. (meters above sea level). Typically, the stream valley-hill relief is a few tens of meters up to 300 m. The same kind of relief seems to exist under the ice sheet, where the ice bed is locally depressed hundreds of meters below the sea level (Lindbäck and Pettersson, 2015). The area in focus for the deep boreholes is located in between the glacial outlets, Russel's Glacier to the south and Isunnguata Sermia to the north (Fig. 1).

Three boreholes named GAP01, GAP03 and GAP04 were drilled within the project, and the drilling and mapping are reported in Pere (2014). Borehole GAP01 is 222 m long (191 m of vertical depth, ground surface elevation 374 m.a.s.l.), and drilled 20 m from the shoreline of a 1200 m long, 300–400 m wide and 40 m deep lake overlying a talik, about 1 km from the nearest ice margin. The NE-SW trending elongated lake basin is located in a lineament. The GAP03 borehole (ground surface elevation 484 m.a.s.l.) was drilled about 1 km from the margin of Isunnguata Sermia, 6 km NE from the talik lake. Drilling of GAP03 was terminated at 341 m length