

Formation of uranium-thorium-rich bitumen nodules in the Lockne impact structure, Sweden: A mechanism for carbon concentration at impact sites

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Abstract—The Ordovician Lockne impact structure is located in central Sweden. The target lithology consisted of limestone and black unconsolidated shale overlaying a Precambrian crystalline basement. The Precambrian basement is uranium-rich, and the black shale is both uranium- and organic-rich. This circumstance makes Lockne a good candidate for testing the occurrence of U-Th-rich bitumen nodules in an impact structure setting. U-Th-rich bitumen nodules are formed through irradiation; hence the increase in the complexity of organic matter by a radioactive (uranium- and thorium-rich) mineral phase. U-Th-rich bitumen nodules were detected in crystalline impact breccia and resurge deposits from the impact structure, but samples of non-impact-affected rocks from outside the impact structure do not contain any U-Th-rich bitumen nodules. This implies that in the Lockne impact structure, the nodules are associated with impact-related processes. U-Th-rich bitumen nodules occur throughout the geological record and are not restricted to an impact structure setting, but our studies at Lockne show that this process of irradiation can readily occur in impact structures where fracturing of rocks and a post-impact hydrothermal system enhances fluid circulation. The irradiation of organic matter by radioactive minerals has previously been proposed as a process for concentration of carbon on the early Earth. Impact structures are suggested as sites for prebiotic chemistry and primitive evolution, and irradiation by radioactive minerals could be an important mechanism for carbon concentration at impact sites.

INTRODUCTION

Aim of Study

Irradiation causes the polymerization of simple organic molecules, generating an increase in complexity and concentration of carbon. This has been proposed as a mechanism that could have occurred on the early Earth, involving irradiation from radioactive uranium- and thorium-rich minerals (Parnell 2004). Is this a process that can occur in impact structures, which have been suggested as sites for prebiotic chemistry and primitive evolution (Cockell et al. 2006)? Uranium- and thorium-rich bitumen nodules formed through this irradiation process are detected throughout the geological record (e.g., Rasmussen et al. 1989; Parnell et al. 1990; McCready et al. 2003; Robb et al. 1994), but are here

for the first time studied in an impact structure setting. This study aims to test the occurrence of U-Th-rich bitumen nodules in the Lockne impact structure, central Sweden. Lockne comprises both uranium and organic-rich target rocks.

Geological Setting

Target Rocks and Impactites

The ~455 Ma Lockne impact structure (Lindström et al. 1992) is located on the fringe of the Caledonian front, ~20 km south of the town of Östersund (Fig. 1). The Lockne impact structure has a concentric morphology with an inner crater diameter of 7 km and an ejecta rim of crushed basement rocks up to 2.5 km wide (Lindström et al. 2005). The target rocks consisted of a Proterozoic granitic basement overlain by a sedimentary cover of mainly black unconsolidated shale,

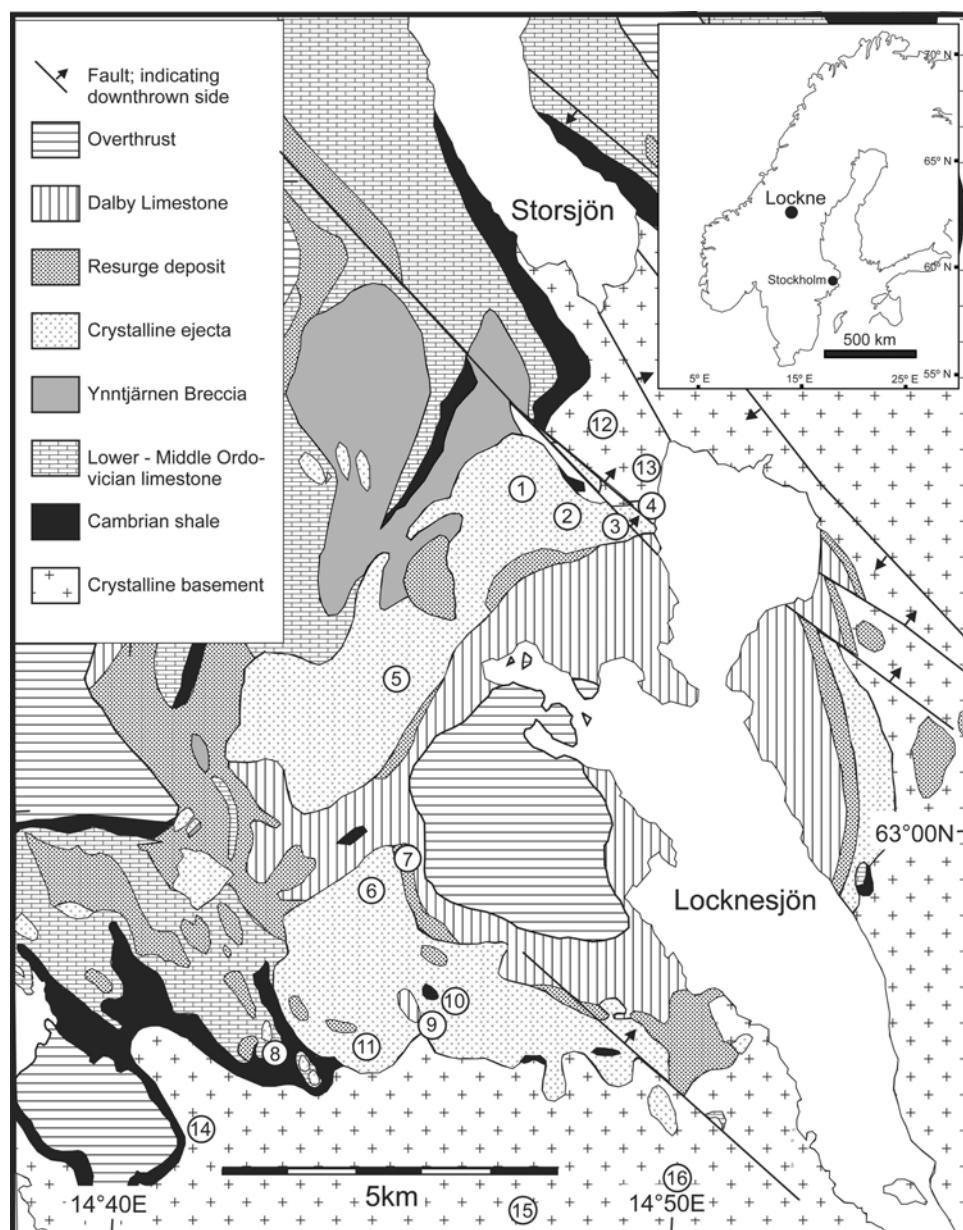


Fig. 1. Geological map of the Lockne impact structure, after Lindström et al. (2005). Sample localities 1–16 are marked on the map. See Table 1 for sample description and occurrence of U-Th-rich bitumen nodules.

limestone, and seawater (Sturkell 1998). The basement was extensively fractured and brecciated as a result of the impact (Lindström et al. 1992; Sturkell 1998). This crystalline impact breccia is referred to as the Tandsbyn breccia, and consists of crystalline basement clasts set in a fine-grained and dark lithic matrix. The matrix is mainly composed of finely crushed granitic material with traces of carbonaceous matter (Sturkell 1998). There are three main varieties of the Tandsbyn breccia (Lindström et al. 2005): matrix-supported breccia, clast-supported breccia, and carbonaceous breccia. The carbonaceous breccia has a dark, sooty, or asphalt-like matrix derived from the carbon-rich shales. Other impactites at

Lockne include the resurge deposits (Sturkell 1998), Lockne breccia, and Loftarstone. The Lockne breccia is a coarse polymictic breccia with limestone clasts, shale clasts, and crystalline clasts set in a muddy matrix. The Loftarstone is a fine-grained deposit overlying the Lockne breccia, and is composed of grains and fragments of crystalline rock, limestone, and shale. The Loftarstone contains quartz grains with planar deformation features and microscopic particles of melted rock (Therriault and Lindström 1995). An autochthonous monomictic limestone breccia, referred to as the Ynnjtjärnen breccia (Lindström et al. 2005), is located below the resurge deposits.

Organic Matter in Lockne Impactites

Organic matter in the Lockne impact structure is widespread in the crystalline-impact breccia. There it occurs as fracture-fillings of carbon-rich shale and bitumen, as well as hydrocarbon fluid inclusions (Sturkell 1998). Organic matter is also extensively dispersed in the dark bituminous matrix of the carbonaceous Tandsbyn breccia (Sturkell 1998). In the resurge deposits, organic matter is present predominantly as carbon-rich shale fragments in the Lockne breccia and Loftarstone, and in the muddy matrix of the Lockne breccia.

Furthermore, both the crystalline breccia and the resurge deposits contain organic matter in the form of bitumen nodules that contain inclusions of uranium- and thorium-rich radioactive minerals. These nodules are referred to as U-Th-rich bitumen nodules.

Occurrence and Formation of Uranium-Thorium-Rich Bitumen Nodules

Bitumen coatings around radioactive mineral grains are present throughout the terrestrial geological record. They are generally reported in sandstones and conglomerates (e.g., Parnell and Eakin 1989; Parnell et al. 1990; Parnell 1996a, 1996b; McCready and Parnell 1998). Formation of U-Th-rich bitumen nodules in sedimentary rocks is often linked to the existence of plutonic rocks supplying thorium and uranium in the vicinity of a sedimentary basin (Parnell et al. 1990). Uraniferous and thoriferous bitumen nodules are also present (although less well-known) in granitic and other basement rocks, including around the Witwatersrand basin, South Africa (Klemd and Hallbauer 1987; Robb et al. 1994), the Rum Jungle Mineral Field, Australia (McCready et al. 2003) and the Athabasca Basin, Canada (McCready, personal communication 2007). In some cases, U-Th-rich bitumen nodules may be the only evidence for hydrocarbon migration through the rocks, and they are therefore a valuable source of information for the palaeofluid record (Parnell 1994).

The formation of the nodules can occur where hydrocarbon passes through a permeable rock immediately adjacent to radioactive minerals. The hydrocarbons form coatings around the radioactive mineral, or digest and replace it so that only small grains of the original mineral are preserved (Parnell 2004). Ellsworth (1928) was among the first to describe carbonaceous matter containing thorium and uranium, and referred to this as thucolite. Laboratory work by Charlesby (1954) showed that radiation causes the breaking of C-H bonds in hydrocarbons, releasing hydrogen and forming free carbon, which crosslinks with another hydrocarbon molecule. As crosslinking continues, hydrogen gas evolves, and a complex organic polymer molecule of increased molecular weight develops, which rapidly becomes insoluble. During continued irradiation, unaltered hydrocarbon molecules are drawn into the cross-linked network. In nature, this is a process that will concentrate carbonaceous matter where it otherwise might be undetectable.

METHODS

Detection of U-Th-Rich Bitumen Nodules

Samples of crystalline impact breccia and resurge deposits collected from 11 localities across the impact structure (Fig. 1; Table 1), including core samples of crystalline impact breccia, were prepared as blocks with a polished surface area of $\sim 1 \times 2$ cm. The polished surface areas of the blocks were examined using an ISI-ABT55 scanning electron microscope (SEM) equipped with an energy dispersive X-ray analyzer at the University of Aberdeen for detection of U-Th-rich bitumen nodules (Table 1). Nonimpact-affected granitic basement from 5 localities outside the impact structure (Fig. 1; Table 1), and 5 samples of fault-brecciated crystalline basement related to Caledonian shear zones (Högdahl et al. 2001) ~ 20 km southwest of the impact structure were prepared in the same way and examined for comparison (Table 1).

Reconstruction of Fluid Entrapment

Hydrocarbon migration conditions were determined through analysis of 14 fluid inclusions in one sample of a quartz-bearing crystalline impact breccia containing veins of solid bitumen (sample 6.1 in Table 1). Polished fluid inclusion wafers were prepared and characterized using a calibrated (error ± 1 °C) Linkam TH-600 fluid inclusion stage, with a heating rate of 10 °C/min. The standard methods of Shepherd et al. (1985) were employed. Inclusions that showed evidence of stretching were excluded from the study. Due to the coeval entrapment of aqueous and hydrocarbon fluid inclusions, the aqueous fluids are gas-saturated and therefore require no pressure correction (Hanor 1980). Homogenization temperatures are therefore indicative of the minimum temperatures of fluid entrapment. Final ice melting temperature measurements were determined using a heating rate of 1 °C/min. Salinities were determined using the methods of Bodnar (1993).

RESULTS

Distribution of U-Th-Rich Bitumen Nodules

A total of 55 U-Th-rich bitumen nodules were detected in the crystalline breccia and resurge deposits from the Lockne impact structure. The nodules are present in 17 of 20 impact structure samples investigated (Table 1). In addition to the Lockne impactites, we also examined 5 samples of local granitic basement from outside the impact structure (Fig. 1; Table 1), and 5 samples of Caledonian fault-brecciated crystalline basement from ~ 20 km southwest of the impact structure (Table 1). Both the crystalline basement and the Caledonian fault breccias from outside the impact structure contain uranium and thorium-rich minerals, particularly Th

Table 1. The occurrence of U-Th-rich bitumen nodules and fracture filling bitumen in samples from the Lockne impact structure and adjacent areas.

Locality	Sample	Fracture-filling bitumen	Number of U-Th-rich bitumen nodules
1 Skanska quarry	1.1 Matrix-supported Tandsbyn breccia	–	–
	1.2	Yes	–
	1.3	Yes	2
	1.4	Yes	2
2 Golf course car park	2.1 Carbonaceous Tandsbyn breccia	Yes	1
3 Lockne church	3.1 Carbonaceous Tandsbyn breccia	Yes	2
4 Stensjö railway section	4.1 Clast-supported Tandsbyn breccia	Yes	1
	4.2	Yes	4
5 Tand	5.1 Clast-supported Tandsbyn breccia	–	1
6 Tandsbyn railway section	6.1 Matrix-supported Tandsbyn breccia	Yes	12
	6.2 Matrix-supported Tandsbyn breccia	Yes	10
7 Crater-track Tandsbyn	7.1 Loftarstone	–	1
8 Klocksåsen	8.1 Ejecta block of matrix-supported Tandsbyn breccia	Yes	12
9 Southwest of Blisterloken	9.1 Loftarstone	–	1
	9.2 Lockne breccia	–	2
10 Southwest of Blisterloken	10.1 Tandsbyn breccia	Yes	2
11 Drillcore 6986072/1446487	11.1 Carbonaceous Tandsbyn breccia 13.40 m	–	–
	11.2 Carbonaceous Tandsbyn breccia 22.90 m	Yes	1
	11.3 Matrix-supported Tandsbyn breccia 27.75 m	Yes	1
	11.4 Clast-supported Tandsbyn breccia 30.50 m	Yes	2
12 Local crystalline basement	12.1 Granite	–	–
13 from outside Lockne impact structure	13.1	–	–
14	14.1	–	–
15	15.1	–	–
16	16.1	–	–
17 Samples from Caledonian shear zones, immediately south of the village of Hackås, ~20 km southwest of Lockne impact structure.	17.1 Crushed basement	Yes	–
18	18.1	Yes	–
19	19.1 Fault breccia	Yes	–
20	20.1 Coherent basement	No	–
21	21.1	No	–

Localities 1–11 are samples of crystalline impact breccia and resurge deposits from the Lockne impact structure. Localities 12–16 are samples of local crystalline basement from outside the impact structure. Localities 1–16 correspond to the sampling localities marked on the map in Fig. 1. Localities 17–20 are from Caledonian fault zones ~20 km southwest of the Lockne impact structure. Each sample is a block with a polished surface area of $\sim 1 \times 2$ cm.

silicates. The Caledonian fault breccias from outside the impact structure also contain bitumen, but no U-Th-rich bitumen nodules were detected in either the fault breccias or in the granitic basement from outside the impact structure (Table 1).

Textural and Morphological Features of U-Th-Rich Bitumen Nodules

In the crystalline impact breccia, the U-Th-rich bitumen nodules occur within microfractures in crystalline clasts as well as in the matrix. The diameter of the nodules varies between ~50 to 400 μm . They vary from being rounded and massive (Fig. 2a) to highly irregular (Fig. 2b); the former are the more common textural variety. The highly irregular nodules could represent veining of bitumen by the U-Th minerals, or possibly many smaller bitumen nodules joined together. A rim of calcite may partially surround the nodules

(Figs. 2a and 2c), which may represent the infilling of space produced by hydrocarbon contraction during the latter stages of nodule formation. Fractures emanating from the calcite rim are a product of radiation damage termed “radioactive blasting halos” (Schidlowski 1966). The nodules locally exhibit lobate and crenulated margins, which is suggestive of a replacement origin (Figs. 2c and 2d) (McCreedy et al. 2003). The uranium-thorium phase often occurs as small inclusions in the bitumen, which implies digestion of a pre-existing mineral (Figs. 2a, 2c, and 2d). Locally, the inclusions are larger (Figs. 2e and 2f), and in some places the bitumen has enveloped discrete radioactive minerals (Fig. 2e). The uranium-thorium phase occurs as uranium-thorium silicates, uranium oxides, and uranium-rich phosphates. The most common nodule-related minerals are thorite, coffinite, uraninite, and monazite. The mineral phases were identified qualitatively by acquiring X-ray spectrum of the mineral phases during SEM examination.

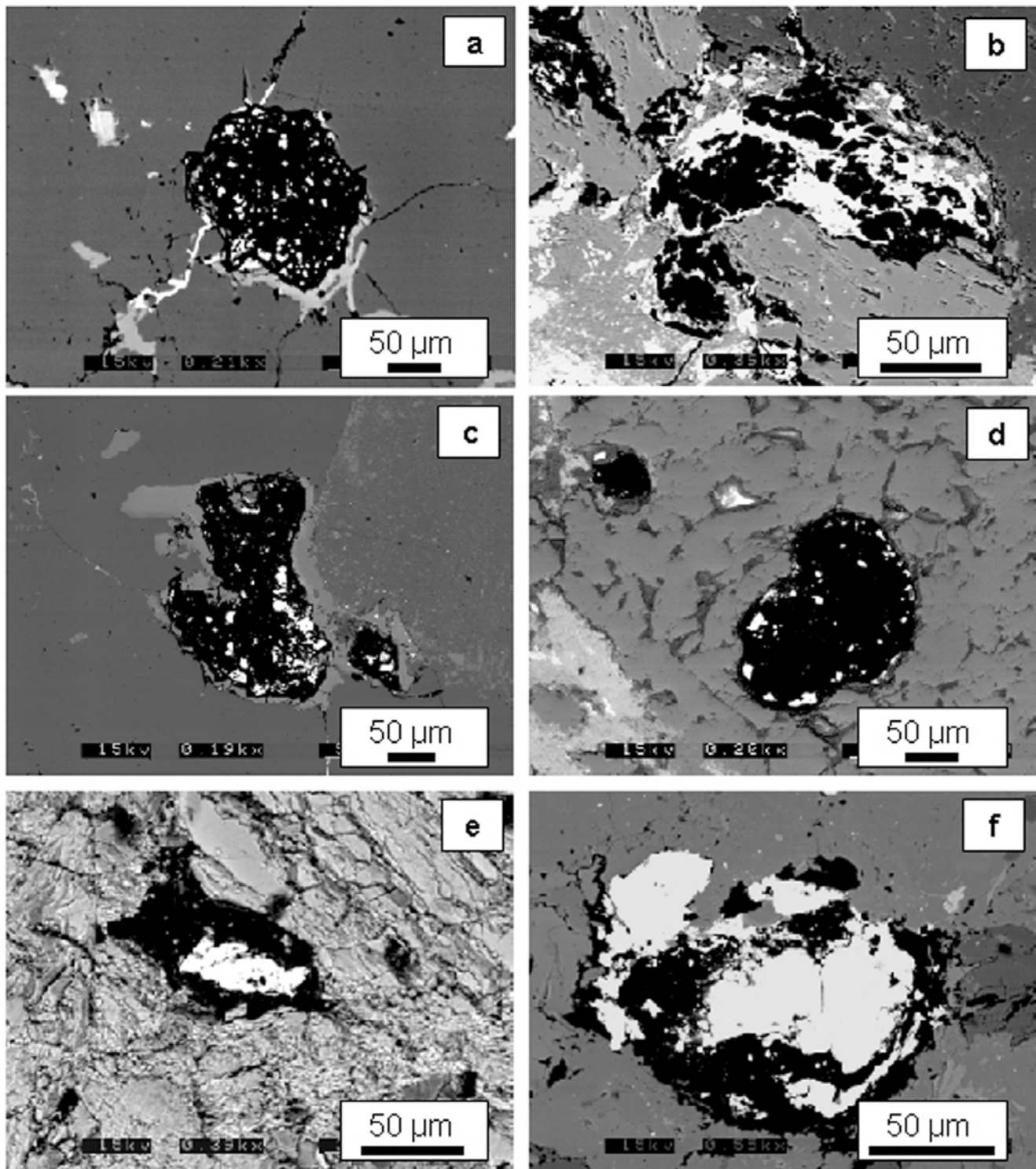


Fig. 2. Backscattered electron photomicrographs of U-Th-rich bitumen nodules in the crystalline impact breccia from the Lockne impact structure. The black components are bitumen; brighter inclusions are the U-Th-rich phase. a) A rounded and massive U-Th-rich bitumen nodule hosted by quartz and surrounded by a rim of calcite (light gray). Fractures emanating from the rim are the products of radiation damage (sample 4.1). b) Highly irregular U-Th-rich bitumen. The bitumen is either veined by the U-Th phase, thorite, or many smaller bitumen nodules joined together. The nodule is hosted by quartz (dark gray) and chlorite (light gray) (sample 6.1). c) Solid U-Th-rich bitumen nodules with crenulated margins, indicative of replacement, surrounded by a rim of calcite (light gray) and hosted by quartz (sample 4.2). d) Solid U-Th-rich bitumen nodules containing scattered grains of coffinite. The nodules are hosted by quartz. The larger nodule could represent two different nodules that have joined together (sample 4.2). e) A nodule where the bitumen occurs as mantle around a monazite grain. The host is potassium feldspar (sample 6.1). f) Bitumen coating and partially digesting a monazite grain (sample 6.1).

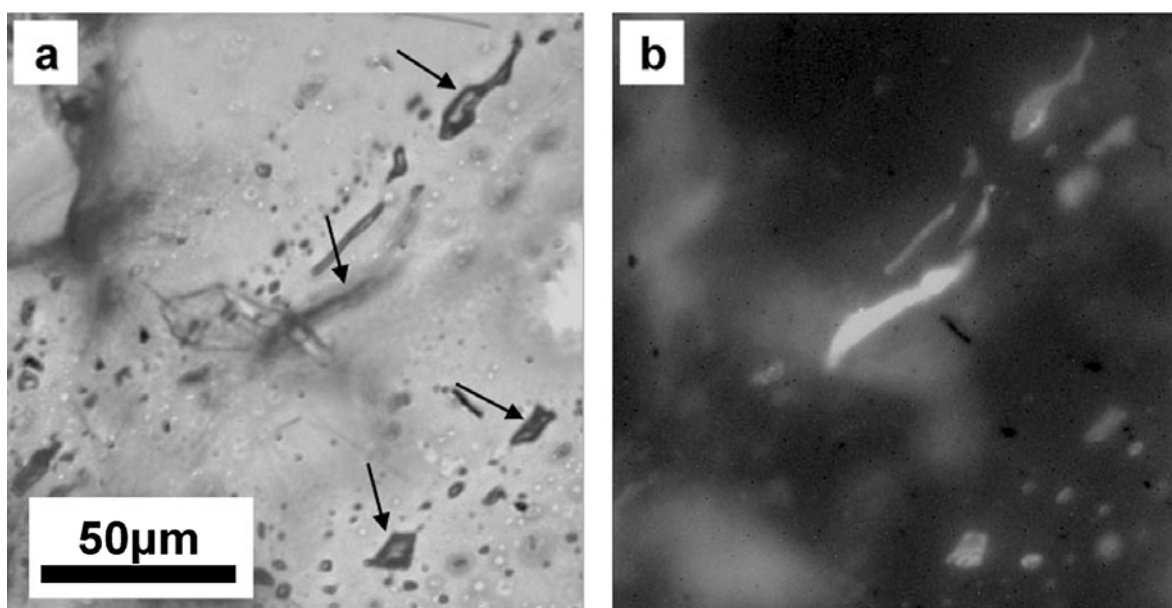


Fig. 3. Fluid inclusion photomicrographs of hydrocarbon fluid inclusions in quartz within the crystalline impact breccia from locality 6; sample 6.1 (Fig. 1; Table 1). a) Transmitted light image showing fluid inclusion trails of monophase hydrocarbon inclusions. b) Corresponding UV-light image showing fluorescence of the hydrocarbon inclusions.

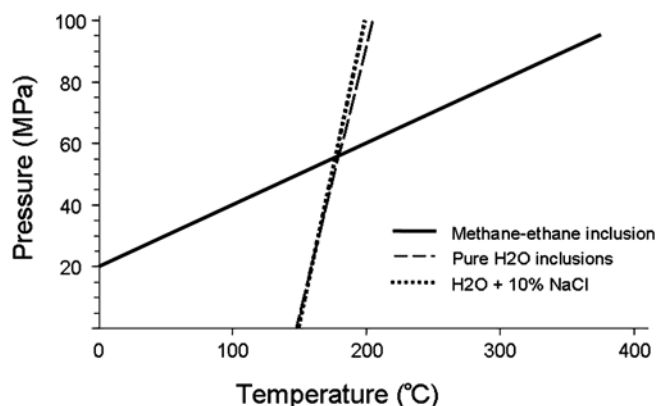


Fig. 4. Isochore model for coeval aqueous and methane/ethane inclusion entrapment. The model shows entrapment conditions of fluids, using the program Fluids (Bakker 2003). The isochores cross at a pressure of almost 60 MPa and a temperature of about 180 °C. Microthermometry indicates that the aqueous fluids have a salinity between 0% and 10% equivalent NaCl.

Conditions of Fluid Entrapment

Quartz in the crystalline impact breccia contains trails of secondary, monophase hydrocarbon fluid inclusions, which are identified by a distinct yellow fluorescence under ultraviolet light (Fig. 3). These inclusions homogenize to a liquid at -55.8 °C ($n = 1$, range -52.7 °C to -58.6 °C). The hydrocarbon inclusions are coeval with two-phase aqueous inclusions that homogenize at 149.7 °C ($n = 4$, range 147.6 °C to 151.9 °C) (Fig. 4). The hydrocarbon inclusions have previously been described by Sturkell et al. (1998) and shown by Raman spectroscopy to have an approximate composition of 95% methane and 5% ethane and

aromatic hydrocarbons (Sturkell et al. 1998). The co-entrapment of the hydrocarbon and aqueous inclusions allows use of the crossed isochore method to estimate the pressure and temperature of fluid entrapment (Goldstein and Reynolds 1994). This method relies on the construction of separate isochores for each of the hydrocarbon and aqueous phases, and using their point of intersection to define the conditions of entrapment. Isochores deduced from the microthermometry data (-55.8 °C for the hydrocarbon inclusions and 149.7 °C for the aqueous inclusions) and the composition data of Sturkell et al. (1998) using the program Fluids (Bakker 2003) are plotted in Fig. 4. The isochores cross at a pressure of almost 60 MPa and a temperature of about 180 °C.

DISCUSSION

Fluid Flow

The polymerization of bitumen around the radioactive minerals took place during the migration of a hydrocarbon front through the impact structure. The conditions of fluid entrapment (Fig. 4) are less elevated than those experienced during the Caledonian Orogeny (Sturkell et al. 1998), and therefore suggest that the fluid migration and entrapment was not related to the Orogeny. They are, however, consistent with a hydrothermal system circulating after the impact event (Sturkell et al. 1998). Although the pressures and temperatures were greater during the Caledonian Orogeny than during the impact-generated hydrothermal system, the fluid inclusions were not reset. The survival of pre-orogenic inclusions has been demonstrated elsewhere in the Caledonides (Baron et al. 2003).

The granitic rocks in the impact structure have been fractured and brecciated by the impact, and trails of hydrocarbon fluid inclusions in the quartz within crystalline clasts demonstrate that microfracturing, providing pathways for hydrocarbon migration, and fracture healing can take place on this scale. The bitumen-coated radioactive minerals at Lockne are in good agreement with the aforementioned known occurrences in granitic settings in other parts of the world. Hydrocarbon flow in basement rocks as a result of flow through fracture systems is more widespread than commonly believed (e.g., P'an 1982; Petford and McCaffrey 2003). Gaseous fluids are more likely to flow in low-permeability basement rocks. The composition of the fluid inclusions at Lockne (95% methane, 5% ethane and aromatic hydrocarbons [Sturkell et al. 1998]) indicates that gas-rich fluids were involved. Methane flow has also been encountered elsewhere in the Swedish Caledonides (Grip and Ödman 1944), indicating that a source for methane was widespread. Methane has the potential to polymerize in the same manner as higher hydrocarbons and is the assumed precursor of solid polymers in several cases where carbonaceous matter is found within uranium deposits (Křibek et al. 1999; Court et al. 2006).

The granitic basement outside the impact structure is much less extensively fractured and brecciated than the impact-brecciated granite and therefore it is less likely to accommodate fluid flow. However, rocks in Caledonian shear zones about 20 km southwest of the impact structure are fractured and brecciated. The fault breccias contain both fracture-filling bitumen and U-Th-bearing minerals, but no U-Th-rich bitumen nodules were detected in these samples. The lack of nodules in the fault breccias might be a result of more viscous fluids flowing in these shear zones, which were not able to interact with a uranium-thorium source (i.e., uranium-thorium-bearing minerals in the host rock).

Implications for Carbon on Mars

This study raises the question of whether radioactivity is a viable mechanism for the fixation of organic carbon in impact structures on other rocky planets such as Mars. The availability of uranium and thorium at the necessary levels to form radioactive minerals requires their concentration in the crust. Uranium and thorium are incompatible and will not be readily incorporated into high-pressure minerals that form in deep magmas, so they will be concentrated in the more differentiated rocks of the Earth's crust. The SNC meteorites from Mars have lower contents of uranium and thorium than in mean values of crustal terrestrial samples (Dreibus and Wänke 1985), but the meteorites do not represent the more differentiated rock types in which radioactive minerals are likely to be present in higher concentrations. The recent discovery of granitic-like rocks on Mars (Bandfield et al. 2004) suggests that some radioactive minerals may occur

there. Gamma ray spectrometry data from Phobos 2 (McLennan 2001) and Mars Odyssey (Taylor et al. 2003) also indicates higher levels of radioelements (K, U, Th) in Martian soils than recorded in the SNC meteorites, but still at lower concentrations than mean values of crustal material on Earth. Given that methane has been recorded in the Martian atmosphere, and that it needs replenishment to maintain its presence (Formisano et al. 2004; Onstott et al. 2006), it is conceivable that the interaction of a methane flux and radioactive minerals could result in a concentration of carbon.

CONCLUSIONS

U-Th-rich bitumen nodules are widespread in the brecciated granites of the Lockne impact structure, but have not been detected in bedrock outside the impact structure. This strongly implies that in the case of the Lockne impact structure, the U-Th-rich bitumen nodules are associated with impact-related processes. These processes include brecciation and fracturing of the bedrock, providing pathways for migration of fluids, and a post-impact hydrothermal system allowing circulation of fluids. Although this case study is based on occurrences within an impact structure, the phenomenon of U-Th-rich bitumen nodules occurs in a variety of settings, including sedimentary basins (Rasmussen et al. 1989; Parnell and Monson 1990), ore deposits (England et al. 2001), and crystalline basement rocks (Robb et al. 1994; McCready et al. 2003). The nodules themselves are just one variety of a wide range of intermixtures of U-Th minerals and organic matter, which occurs in settings as diverse as pegmatites (Parnell 1990) and hydrocarbon reservoirs (Parnell and Eakin 1987).

Where evidence for organic matter may otherwise be very limited, enhanced fluid circulation through impact fracturing and impact-induced hydrothermal system will create a greater chance for organic molecules to be polymerized and concentrated through the interaction with U-Th-rich minerals. It is also reasonable to assume that this mechanism could take place on Mars, therefore enhancing the possibilities of detecting organic matter in Martian impact structures.

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