

Evidence for seismogenic hydrogen gas, a potential microbial energy source on Earth and Mars

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Abstract

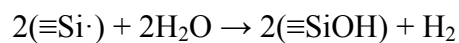
The oxidation of molecular hydrogen (H_2) is thought to be a major source of metabolic energy for life in the deep subsurface on Earth, and could likewise support any extant biosphere on Mars, where stable habitable environments are probably limited to the subsurface. Faulting and fracturing may stimulate the supply of H_2 from several sources. We report the H_2 content of fluids **present** in terrestrial rocks formed by brittle fracturing on fault planes (pseudotachylites and cataclasites), along with protolith control samples. The fluids are dominated by water and include H_2 at abundances sufficient to support hydrogenotrophic microorganisms, with strong H_2 enrichments in the pseudotachylites compared to the controls. Weaker and less consistent H_2 enrichments are observed in the cataclasites, which represent less intense seismic friction than the pseudotachylites. The enrichments agree quantitatively with previous experimental measurements of frictionally driven H_2 formation during rock fracturing. We find that conservative estimates of current martian global seismicity predict episodic H_2 generation by Marsquakes in quantities useful to hydrogenotrophs over a range of scales and recurrence times. On both Earth and Mars, secondary release of H_2 may also accompany the breakdown of ancient fault rocks, which are particularly abundant in the pervasively fractured martian crust. This study strengthens the case for the astrobiological investigation of ancient martian fracture systems.

Key words: Deep biosphere, faults, fault rocks, seismic activity, hydrogen, Mars

1. Introduction

The oxidation of molecular hydrogen (H₂) is an important energy source and carbon fixing process in anaerobic ecosystems, and may have powered the earliest life on Earth (e.g. Kral *et al.*, 1998; Schulte *et al.*, 2006; Sleep *et al.*, 2011; Lane & Martin, 2012). Methanogenesis, acetogenesis and sulphate reduction are among the hydrogenotrophic metabolic pathways highlighted as the most plausible strategies for life on Mars (e.g. Nixon *et al.*, 2013), partly because terrestrial volcanic aquifers analogous to those thought to occur in the martian subsurface are populated by such hydrogenotrophs (Stevens & McKinley, 1995; Chapelle *et al.*, 2002).

On both planets, water–rock reactions and groundwater radiolysis may generate high enough concentrations of H₂ in the subsurface to sustain microbial hydrogenotrophy (e.g. McCollom, 1999; Chapelle *et al.*, 2002; Lin *et al.*, 2005; Onstott *et al.*, 2006; Blair *et al.*, 2007; Nixon *et al.*, 2013). Faulting and fracturing may locally accelerate these processes by exposing fresh rock surfaces to groundwater. It may also both generate and liberate hydrogen from minerals; a number of studies have demonstrated experimentally that H₂ evolves during the crushing or grinding of rocks and minerals. Among these, Freund *et al.* (2002) report a passive role for cracking and fissuring in accelerating the outwards diffusion of H₂ from interstitial sites in crystalline rocks, where it can form during cooling from the redox conversion of OH[−] into peroxy defects and H₂ (*ibid.*, and references therein). Kita *et al.* (1982) described an active role for frictional grinding (on a fault plane) in generating silicon radical species that react rapidly with water to yield H₂, a process represented by the summary reaction:



Experimental support for this “mechanoradical” mechanism was furnished by Saruwatari *et al.* (2004), while Hirose *et al.* (2011) showed that frictional grinding liberates H₂ from a range of silicate and non-silicate rocks (as a function of the friction applied), and that even dry basalt (maintained at 100°C for two weeks to expel intergranular water) generates H₂ under friction, probably derived from crystallographic water. In support of these experimental results, an association between earthquake activity and high fluxes of H₂ has been observed in natural fault zones (Wakita *et al.*, 1980; Wiersberg & Erzinger, 2008). Both Freund *et al.* (2002) and Hirose *et al.* (2011) have suggested that this earthquake-generated (or earthquake-liberated) H₂ might fuel pockets of microbial activity around faults on Earth; Michalski *et al.* (2013) extrapolated this proposal to Mars.

The present study aimed to determine: (1) whether the unique rock-types produced by faulting contain molecular hydrogen, and if so: (2) whether the H₂ and H₂O contents of these rocks, if representing original pore fluids, would imply a hydrogen concentration sufficient to support microbial hydrogenotrophy, and (3) whether the empirical model of frictionally controlled H₂ release during earthquakes furnished by Hirose *et al.* (2011) is quantitatively appropriate to suggest an explanation for the measured enrichments. We then asked (4) whether biologically significant quantities of hydrogen are predicted by this model when applied to the present-day subsurface of Mars.

1.1 Measurement of H_2 in fault rocks

Movement on fault planes generates two unique rock types: cataclasites and pseudotachylites. Cataclasites are fault breccias, comprising angular clasts of wall rock in a comminuted fine-grained matrix; these rocks are produced by brittle fracturing and grinding during seismic activity. Pseudotachylites (also spelled “pseudotachylytes”) are glassy or very fine grained fault rocks formed under more intense friction by melting; the molten rock forms under pressure along fractures and other planes of weakness, often flows considerable distances, and quenches rapidly to form dyke- or vein-like structures (Macloughlin & Spray, 1992). Pseudotachylites on faults represent planes of weakness along which new fractures propagate to accommodate tectonic strain, and typically show complex, fractured microstructures indicative of brittle deformation (e.g. Wenk, 1978; Kirkpatrick & Rowe, 2013).

Faulted rocks commonly contain healed microfractures bearing ancient fault-zone fluids (e.g. Lespinasse & Cathelineau, 1990; Anders *et al.*, 2014). In the present study, cataclasites and pseudotachylites were crushed to evacuate fluids for sampling by a mass spectrometer. The cataclasites were collected from Cambrian quartzites associated with the Moine Thrust Belt in Scotland. Impact-related pseudotachylites were collected from two very large Precambrian crater structures—the Sudbury Crater in Canada (granite host) and the Vredefort Crater in South Africa (quartzite host)—and non-impact-related pseudotachylites were recovered from Permian granite in the Isles of Scilly (UK) and Precambrian gneiss in the Outer Hebrides of Scotland. Control samples representative of the original pre-faulting lithologies were collected from the Outer Hebrides, the Moine Thrust Belt, and the Isles of Scilly, with an additional Precambrian quartzite sourced from Kiruna in northern Sweden.

We analysed the bulk fluid content of our samples using the CFS mass spectrometry technique previously used by our team to liberate and analyse volatiles from serpentinites, mineral veins, basalts, and most recently martian meteorites (Parnell *et al.*, 2010; McMahon *et al.* 2012; McMahon *et al.* 2013; Blamey *et al.*, 2015). This method involves crushing the sample and immediately passing the volatiles released through a quadrupole mass spectrometer, and offers very low detection limits.

2. Methods

2.1 Materials

The fault-rock samples are tabulated in **Table 1**. Two of the pseudotachylite samples were collected from Earth's two largest known impact structures, the 1.85-Ga Sudbury crater in Canada and the 2.02-Ga Vredefort structure in South Africa. Control samples were unfortunately not recovered from the crater localities. The remaining pseudotachylites and appropriate control samples were collected from the Outer Hebrides and the Gairloch region in Northwest Scotland, and the Isles of Scilly, all in the UK.

The Sudbury crater spans about 70 km and is tectonically distorted into an elliptical shape (**Figure 1a**). Pseudotachylites occur as the matrix in the “Sudbury breccia”, which forms a 15-km-wide collar around the Sudbury Igneous Complex (SIC). The sample analysed here was collected by Alison Wright from granite host rock near Creighton in the Creighton Pluton, adjacent to the SIC. The Vredefort structure, which occurs within the Witwatersrand Basin, is much larger, spanning 250 km. Pseudotachylites occur here as veins and dykes on scales from mm to tens of m within the central Vredefort Dome, which represents the central uplift of the original crater (Riller *et al.*, 2010). The sample analysed in the present study was donated by Roger L. Gibson, who collected it from a quartzite unit in the Witwatersrand Supergroup, at a locality about 10 km NNW from the town of Parys, towards the edge of the Vredefort Dome (**Figure 1b**). It has been suggested that the Sudbury and Vredefort pseudotachylites are impact melts injected across large distances rather than local frictional melts derived from the fault walls (Riller *et al.*, 2010). However, both impact-related and non-impact-related pseudotachylites are rich in healed microfractures and other microstructural features indicative of brittle deformation

and cataclasis and as such may contain H₂ and other fluids trapped during fault motion (e.g. Wenk, 1978; Boullier *et al.*, 2001; Lafrance & Kamber, 2010).

The samples from the Outer Hebrides were collected on the islands of Barra, South Uist and North Uist, and represent pseudotachylites of the Outer Hebrides Fault Zone (OHFZ), which runs about 170 km ~N–S down the eastern seaboard of the islands (**Figure 2**). Pseudotachylites in this region are hosted by the Late Archean/Early Palaeoproterozoic Lewisian Gneiss, and appear to have been formed during several different fault reactivation events during the Precambrian, including one at ~1250 Ma and one at ~700 Ma (Sherlock *et al.*, 2008). Pseudotachylites recovered from coastal exposures on Barra are illustrated by **Figure 3** and were found to contain healed microfractures probably representing brittle deformation during fault reactivation.

Pseudotachylites in the Gairloch region on the northwest Scottish mainland are also hosted by Lewisian Gneiss but are associated with a different fault system and have been dated to around 1 Ga (Sherlock *et al.*, 2008). The samples analysed here were collected from the Loch Tollie Quarry, west of Loch Tollie, and from a second locality to the east of Loch Tollie, where the control gneiss sample was also collected. The loch lies between two “crush belts” (shear zones) about 1 km distant to the NE and SW (Sherlock *et al.*, 2008; **Figure 4**). Each of the Scottish pseudotachylite occurrences consists of glassy black vein networks up to metres in width, containing fragments of unaltered gneiss country rock. The pseudotachylites are locally cut by planar quartz veins, representing brittle reactivation of the fault system. Samples from the Loch

Tollie localities contain pyrite up to 1 mm size, particularly around healed microfractures (**Figure 5**). The analyses were made of glassy material free of country rock, quartz or pyrite.

The Isles of Scilly are located about 50 km WSW from the westernmost point of mainland Cornwall, England. They are dominated by a large Hercynian (Permo-Carboniferous) granite pluton, with superficial sedimentary cover. As far as we can discover, pseudotachylite has not previously been described in the Isles of Scilly, although Barrow & Flett (1906) refers to “greisen veins” in the granite, by which the pseudotachylite may be intended. The samples analysed here were collected from float material of granitic rock cut by centimetres-width black, glassy veins on a beach on St. Agnes Island where the pseudotachylite can also be seen *in situ* (**Figure 3B**). The control granite (predominantly quartz and feldspar, identical to the pseudotachylite host rock) was also collected at this locality.

Cambrian cataclasites (originally quartzites) were sampled from faults associated with the Moine Thrust Belt in the vicinity of Ullapool in western Scotland (**Figure 4**). Each of the Cambrian samples was originally a quartz arenite (almost pure quartz sandstone), consisting of rounded grains of medium-coarse quartz sand, cemented by quartz overgrowths, such that there is negligible residual porosity. This large and structurally complex thrust zone developed during the Caledonian orogeny around 450–430 Ma (Goodenough *et al.*, 2011), and extends for nearly 200 km. Samples were collected by the present authors from a beach outcrop near Achiltibuie, a roadcut at Skiag Bridge, the mountain Conival near Inchnadamph and a roadcut at Corry Bridge near Ullapool. Skiag Bridge is a well-known locality close to the shore of Loch Assynt, where the Cambrian Pipe Rock quartzite is offset by a minor thrust fault, with “damage zones” on both

sides (Lloyd, 2000). The Conival sample is from a crushed zone in which brecciated quartzite fragments are cemented by a matrix rich in iron oxide and kaolinite. The Corry Bridge exposure (on the A835 road) is also a well-known locality, where fractured Cambrian quartzite outcrops along a fault where rocks of the Proterozoic Torridonian Supergroup are thrust over limestone of the Ordovician Durness Group (Allison *et al.*, 1988). Five control samples for the cataclasite analyses were selected from a range of Cambrian quartzites from western Scotland and an additional specimen was sampled from the Palaeoproterozoic Hauki Quartzite at Kiruna, Sweden. The control sample from Coulags is conspicuously veined by quartz as a result of Moine Thrust deformation, but is not a cataclasite (and the quartz veins themselves were excluded from the analysed portion). The sample from Ord, Isle of Skye, was obtained from close to a thrust fault, but is not a cataclasite.

2.2 Crush-fast-scan mass spectrometry

Samples were extracted from the interiors of rock specimens using a water-lubricated diamond circular saw to avoid (as far as possible) superficially weathered and contaminated material. Match-head-sized samples of about 250 mg were obtained by crushing specimens with a stainless steel fly-press scrubbed clean with acetone. Samples were rinsed in hydrogen peroxide, potassium hydroxide and deionized water to remove superficial organic matter and dried in a laminar flow hood before sealing in sterile plastic bags for shipping.

Volatiles were extracted and analysed at the New Mexico Institute of Technology using the crush-fast-scan (CFS) mass-spectrometry technique (Norman & Moore, 1997; Moore *et al.*, 2001; Parry & Blamey, 2010; Blamey, 2012; Blamey *et al.* 2015). Samples were crushed incrementally under an ultra-high vacuum ($\sim 10^{-8}$ Torr), with each crush liberating a swift burst of mixed volatiles. Each crush increment may liberate fluids from multiple sites. A typical sample size of about 250 mg released four to ten bursts of volatiles into the vacuum chamber, which remained there for 8–10 analyser scans (~ 2 seconds) before removal by the vacuum pump. This method does not require a carrier gas and volatiles are not separated from each other but released simultaneously into the chamber. The crushing area and crushing bellows were cleaned with potassium hydroxide and swabbed with de-ionized water and isopropanol prior to analysis. Crushing of fused silica cuvettes with low gas-content (“blanks” used to support the analysis of geological samples) produces a burst of hydrogen three-to-four orders of magnitude smaller than that released from fluid inclusions in rocks; these bursts are likely to represent trace gases already present in the blanks.

3. Results

The main constituents of the evacuated fluids were H₂, N₂, CO₂, and H₂O (**Table 2**), with most samples dominated by water (typically >90%). CO₂ was the most abundant of the non-water phases. All pseudotachylites with local country rock controls were enriched in H₂ compared to the controls. For comparison with model predictions and microbial requirements expressed as aqueous concentrations, we report H₂ relative to H₂O (mol dm⁻³) using the H₂O and H₂ percentages; however, the measured values generally exceed the solubility of H₂ in water under standard conditions (~0.8 mM), indicating that some hydrogen is actually present in a free gas phase coexistent with water. (An almost identical overall distribution of results is found if H₂/N₂ or %H₂ is considered.) Results for the pseudotachylites are shown in **Figure 6A**. H₂ abundance per volume of water in the pseudotachylites ranged from 1.9×10^{-2} mol dm⁻³ (an outlier) to 8.3 mol dm⁻³, with most values between approximately 0.1 and 1.0 mol dm⁻³. The high value at Barra Quarry (1.1% H₂ or 6.2 mol dm⁻³) should be interpreted with caution because the fluids from this sample were uniquely dominated by CO₂ rather than water. Country-rock controls ranged from 0.4 to 5.3×10^{-2} mol dm⁻³. The average enrichment in H₂ enrichment in the pseudotachylites compared to the controls was 1.3 mol dm⁻³ H₂O in the Outer Hebrides gneiss (0.4 mol dm⁻³ if Barra Quarry is excluded), 0.13 mol dm⁻³ in the gneiss west of Loch Tollie, 1.1 mol dm⁻³ in the gneiss east of Loch Tollie and 4.6 mol dm⁻³ in the granite from St Agnes Island. No significant difference in H₂ abundance was observed between the tectonic and the impact-crater pseudotachylites, although suitable controls were not available for comparison with the latter. The two pseudotachylites from the western locality at Tollie, Gairloch are over an order of magnitude discrepant; the higher of the two samples is closer to the pseudotachylites from other localities.

Results for the cataclasites are shown in **Figure 6B**. The H₂ enrichments are slightly higher on average in the cataclasites than the local quartzite controls but lower than those detected in most of the pseudotachylites. The average H₂ enrichment in the cataclasites compared to the local controls was 0.18 mol dm⁻³ (H₂O).

4. Discussion

The gas analysis shows that the pseudotachylites are consistently enriched in hydrogen compared to suitable controls, while an enrichment in the cataclasites is more equivocally suggested. Notably, there is no evidence of hydrogen enrichment in the cataclasite sample from the minor thrust at Skiag Bridge compared to the non-cataclasite quartzite from the same locality. However, it may be significant that the Corry Bridge cataclasite, which is particularly well developed and has been described as “ultracataclasite” (Allison *et al.*, 1988), yields the highest H₂ abundance among the cataclasites we analysed.

4.1 Possible sources of measured hydrogen

The rocks are of low permeability and the exposures sampled are not close to mafic or ultramafic rocks, to serpentine- or talc-bearing units, or to organic-rich sediments from which H₂ might otherwise have been derived. Radiolysis and water-rock reactions are the most widely discussed mechanisms for generating H₂ within rocks. Radiolysis over long timescales has previously been suggested to account for most of the H₂ in Precambrian basement, although we find H₂ enrichments in the Phanerozoic pseudotachylites comparable to those in the more ancient samples, and the Palaeoproterozoic quartzite sample analysed (Kiruna, northern Sweden) is poorer in H₂ than most of the Cambrian samples. Water-rock reactions capable of generating H₂ during the oxidation of ferrous iron are well known in mafic and ultramafic igneous rocks but have not been widely described in gneiss or granite (which do nevertheless contain some ferrous iron, e.g. in biotite).

The sampled rocks must have lost some H₂ by diffusion over geological time, but the non-fault rocks should not have lost substantially more H₂ than the fault rocks; nor should the cataclasites have lost more than the pseudotachylites. Hence, the H₂ enrichment in the fault rocks compared to the controls implies a process of faulting-related H₂ production or release. The fact that pseudotachylites are found to record a greater H₂ enrichment than cataclasites supports this inference, since the former represent a greater intensity of fault activity and, being glassy or very fine-grained, are especially vulnerable to disruption, including microfracturing, during fault reactivation (Kirkpatrick & Rowe, 2013). However, the difference between the H₂ content of the two rock types may be less significant than the differing tectonic and lithological settings of the particular localities sampled, particularly the fact that the faults sampled for cataclasites had smaller displacements.

Faulting stimulates both water–rock reactions and radiolysis by exposing fresh rock surfaces to groundwater (e.g. Sherwood Lollar *et al.*, 2007; Sleep., 2012), but the addition of water through fractures would not necessarily increase the amount of H₂ per volume of water as recorded in our samples. Moreover, these are slow processes, whereas H₂ generated rapidly at the time of fault motion should be more abundantly trapped (in concurrently propagating microfractures). Such rapid fracture-enhanced processes include the diffusion of existing H₂ out of interstitial sites in crystals (Freund *et al.* 2002) and the mechanoradical mechanism described by Kita *et al.* (1982) and Hirose *et al.* (2011). The melting of the pseudotachylite would only have led to minimal H₂ formation because, given the absence of organic matter and graphite, conditions were insufficiently reducing. However, while a crystalline rock is cooling after crystallisation, the redox conversion of OH⁻ into peroxy defects and H₂ can occur (Freund *et al.* 2002 and references

therein). This may have contributed some portion of the H₂ we measured in the pseudotachylites, which would be retained interstitially independently of any fluid inclusions trapping original pore fluids from the fault zone. Additional fluids, including water, may have been exsolved during cooling after crystallization both of the pseudotachylites and of their precursor crystalline rocks, forming inter- and intragranular fluid-filled cavities.

Assuming that the fluids we measured in the pseudotachylites mostly represent original porewater trapped in microfractures during faulting activity, we now ask whether the measured H₂ enrichments are broadly consistent with the measurements made by Hirose *et al.* (2011) of gas liberated during the grinding-together of rock surfaces. From these measurements, Hirose *et al.* derived a semi-empirical equation describing the concentration of mechanically derived H₂ in fault-zone porewater immediately after an earthquake:

$$[H_2]_{local} = \frac{10^2 \alpha \sigma_{eff} \mu_d}{\phi} \quad \text{Equation 1}$$

where α is an empirical coefficient describing the slope of the graph of hydrogen production against frictional work; σ_{eff} is the effective stress acting on the fault surface, μ_d is the average dynamic friction coefficient during fault motion and ϕ is the average porosity. The use of this equation assumes that (1) all pore-spaces in the original fault zone are filled with fluid, and (2) there is negligible fluid flow over the recurrence time of the seismic motion. These variables—and therefore the estimate of H₂ concentration—are independent of event magnitude. It should be noted that the “concentration” calculated may exceed the actual solubility of hydrogen in water under the relevant conditions and that a separate gas phase may be generated; nevertheless, the

equation predicts the quantity of H₂ per volume of water. The protolith for pseudotachylite samples 1–7 was the famous Lewisian Gneiss. These pseudotachylites probably formed at depths of four to five kilometres (Sibson, 1975). At the present day, the porosity of Lewisian gneiss is reported by geophysical surveys as 0.07% at 1 km depth and 0.005% at 2 km depth (Hall, 1987). Substituting into **Equation 1** the value of the empirical coefficient α determined for granite by Hirose *et al.* (2011; 2012; these authors did not report a value for gneiss) therefore yields an estimated H₂ abundance (per volume of water) of 0.2 mol dm⁻³ at 1 km depth and 5 mol dm⁻³ at 2 km depth, with still higher concentrations at greater depths. Thus, it is realistic to explain the comparable H₂ values measured in our pseudotachylites (*Section 3*) as the product of ancient earthquakes in the Lewisian gneiss, generated mechanically and trapped in groundwater within fluid inclusions. Isotopic study of the H₂ (i.e. D/H ratio) would facilitate more rigorous testing of genetic hypotheses, but may be technically challenging for such small volumes.

4.2 Biological utility of measured hydrogen enrichments

Hirose *et al.* (2011) report that seismogenic H_{2(aq)} in fault zones may attain concentrations in excess of one millimolar, which they follow Takai *et al.* 2006 in regarding as the lower limit for the support of lithoautotrophic microbial communities. Both thermodynamic considerations and field measurements suggest that the true requirement may be much lower. In order to be metabolically viable, archaeal methanogenesis must yield free energies of at least -10 kJ mol^{-1} CH₄ (Hoehler *et al.*, 2001; Heuer *et al.*, 2009). In deep marine sediments, it has been shown that H_{2(aq)} concentrations of only 1 nM ($10^{-9} \text{ mol dm}^{-3}$) enable methanogenic CO₂ reduction to meet this criterion (Heuer *et al.*, 2009). The steady state H_{2(aq)} concentration measured in natural hydrogen-powered ecosystems provides an upper limit for the minimum viable concentration. Lovley & Goodwin (1988) estimate that the steady state H_{2(aq)} concentration in sediments with

hydrogenotrophic methanogenesis or sulphate reduction should be 13 or 1 nM respectively. More recently, H₂ concentrations of ~10 nM were measured in a 200-m deep igneous aquifer supporting hydrogenotrophic methanogens (Chapelle *et al.*, 2002). Hence, even H₂ concentrations of millimolar magnitude are probably far in excess of minimum biological requirements.

If the measured fluids are indeed representative of groundwater trapped during fault motion, then they therefore support the hypothesis that fault-generated gas (from all sources) could supply energy to microbes in fracture zones. Moreover, while most fault-related H₂ presumably escapes into pore and fracture waters where it could support microbial hydrogenotrophs, these results may indicate that another (no doubt far smaller) portion of it is trapped and retained by fault rocks, producing the strong H₂ enrichments we observed. Once preserved in fault rocks, the trapped H₂ may be subsequently liberated by reactivation of the fault, along with any newly generated gas. For example, the numerous reactivation events in the Outer Hebrides Fault Zone may have repeatedly released H₂ from pseudotachylites into local groundwater (e.g. Osinski *et al.*, 2001). Pseudotachylites are particularly vulnerable to alteration and destruction in a variety of other ways that could liberate H₂ (Rabinowitz *et al.*, 2011; Kirkpatrick and Rowe, 2013). As noted, concentrations of H₂ as low as 10⁻⁹ M can probably support hydrogenotrophic methanogens. Our samples liberated on average about 2 × 10⁻¹¹ mol H₂ per gram of pseudotachylite (with a range from 5 × 10⁻¹³ to 9 × 10⁻¹¹); hence, the H₂ contents of one gram of pseudotachylite are sufficient to stimulate methanogenesis in ten millilitres of water, at least instantaneously. The CO₂ necessary for methanogenesis is also available from groundwater (and detected in our pseudotachylites; **Table 2**).

4.3 Implications for martian habitability

It is widely recognised that the most hospitable environments for life on present-day Mars are likely to occur in the deep subsurface (e.g. Boston *et al.*, 1992; Fisk & Giovannoni, 1999; Westall *et al.*, 2013; Cockell, 2014). There is good evidence that liquid water was abundant at the martian surface for long periods of geological time, and most of this water is expected to remain on Mars in the form of ground ice and sub-permafrost liquid aquifers, which potentially achieve thicknesses of up to several hundred metres (Cockell, 2014; Clifford *et al.*, 2010). If present, these aquifers are likely to be buried, on average, ≥ 5 km below the surface, although local variation may be considerable (Clifford *et al.* 2010).

To assess whether frictionally controlled H₂-generating processes on fault planes could generate biologically significant quantities of hydrogen in present-day subsurface aquifers on Mars, we adapt the model of Hirose *et al.* (2011, 2012) for the martian geological context (compaction curve, rock type and gravitational field strength).

Equation 1 yields the approximate H₂ concentration in a fault zone immediately after a seismic event as a function of porosity, dynamic friction coefficient and effective stress (Hirose *et al.*, 2011). Both porosity and effective stress (lithostatic pressure minus fluid pore pressure) vary with depth below the martian surface. **Figure 7** shows the modelled concentrations of seismogenic H₂ as a function of Mars-quake depth according to the porosity–depth relationships given by Athy’s formula ($\varphi_z = \varphi_0 e^{-kz}$ where φ = porosity, φ_0 = surface porosity, k = the compaction coefficient and z = depth; Athy, 1930) supplied with the martian parameters of Clifford *et al.* (2010). Fracture porosity and cataclasis in fault zones could either increase or

decrease bulk porosity away from this background curve. Higher porosities yield smaller concentrations because the volume of fluid into which the H₂ is dispersed is greater. It is assumed that the dynamic friction coefficient is 0.25, pore pressure is hydrostatic, and fluid does not flow away from the fault during seismic activity.

It was suggested in *Section 4.2* that hydrogenotrophic communities require H_{2(aq)} concentrations of at least 10 nM (10⁻⁸ mol L⁻¹). The modelled seismogenic H₂ concentrations below a few km depth are five or six orders of magnitude above this threshold and therefore eminently capable of supporting life. The volume of fluids in which these high H_{2(aq)} concentrations are actually present depends on the magnitude of the seismic event as well as the availability of liquid water; if all the pores are filled with water, it is simply the product of the average displacement D(M) on the fault, the surface area S(M) on which displacement occurs, and the porosity (Hirose *et al.*, 2011). Following Hirose *et al.* (2011), we derive both D(M) and S(M) from the magnitude by the empirical formulae of Utsu, 2001: D(M) = 10^{½M-3.1}; S(M) = 10^{M+2}. Under martian conditions, if the fluids are trapped within the fault zone and all pores are filled with fluid, the groundwater volume varies from only 5.0 m³ for a magnitude-2 event at 5 km depth to 3.7 × 10⁷ m³ for a magnitude-7 event at 10 km depth, if porosity follows the higher curve in **Figure 7** and the empirical coefficient α in Equation 1 is given the experimental value determined for water-saturated basalt under terrestrial air (Hirose *et al.*, 2011).

At the present day, events of these magnitudes or greater occur somewhere on Mars on average every 34 days and every 4,500 years respectively according to the most conservative model of martian seismicity proposed by Knapmeyer *et al.* (2006). This model is based on a fault

catalogue compiled from Mars Global Surveyor altimetry data; direct measurement of seismic activity on Mars awaits the arrival of the SEIS planetary seismometer to be carried on the Insight mission (Mimoun *et al.*, 2012). Combined with the results of Hirose *et al.* (2011, 2012), it predicts that average annual cumulative H₂ production is less than 10 tonnes over the entire planet. Hence, although seismic H₂ production could sporadically fuel pockets of microbial activity on Mars, it would be highly spatially and temporally restricted to fault activity and its aftermath, with small total biomass. Nevertheless, faulting and fracturing during seismic activity on Mars (Mars-quakes and impact events) can improve subsurface habitability in at least two other ways. Firstly, permeability and potentially porosity are increased in the fracture zone, creating new habitable space and enabling fluid flow and cell transport (Sleep, 2012). Secondly, fresh rock surfaces may be brought into contact with circulating water, enabling life-supporting water–rock reactions and H₂ production by radiolysis (Sleep & Zoback, 2007). It is interesting to note that, like radiolysis, the crushing of quartz in water has also been found to produce hydroxyl radicals (e.g. Shi *et al.*, 1988). Thus, there is a previously unrecognised possibility that pyrite is abiotically oxidised to sulphate in the vicinity of active faults, supplying another electron acceptor of key interest on Mars. Experimental work would be worthwhile to determine whether this phenomenon could generate the anomalies in the sulphate concentration of groundwater reported in association with earthquakes (e.g. Song *et al.*, 2006; Harabaglia *et al.*, 2008).

Mars may have had something more like a plate tectonic regime in the distant past (e.g. Yin, 2012). Numerous large faults are exposed at the martian surface, which is also heavily fractured and cratered by billions of years of impacts. It has been proposed that craters could have provided local refugia for microbial life on Mars (and early Earth) by increasing the temperature

and porosity of the target region, enabling hydrothermal circulation over geological timescales (Cockell, 2004). One might speculate that these conditions could also have stimulated the origin of life itself on either planet, providing “crucibles” in which the components necessary for the origin of life evolved through a slow cooling curve, favouring a sequence of prebiotic reactions in a long-lived hydrothermal system (Cockell, 2004; 2006). Our results suggest that a supply of seismogenic H₂ may have contributed to both the habitability and prebiotic chemical evolution of these systems.

5. Conclusions

This study has shown that fault rocks contain elevated quantities of hydrogen gas. The hydrogen concentrations implied by the measured H₂ and H₂O contents are sufficient to support hydrogenotrophic microorganisms, with implications for the habitability of fault zones, if the fluids measured represent trapped fault-zone porewaters. Although fluids derived from other sources may also be present, the enrichment in the fault rocks compared to controls is consistent with faulting-related H₂ production, for which several possible mechanisms have been described. Extending to Mars the empirical model of Hirose *et al.* (2011) for frictionally controlled H₂ production on fault planes, we find that the low level of martian seismicity is sufficient by itself to support spatiotemporally limited hydrogenotrophy in the subsurface. These considerations together with many others suggest that fault rocks or fault-related mineral vein networks deserve consideration as a target for biosignature sampling by future Mars missions.

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7. Author Disclosure Statement

No competing financial interests exist.

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TABLE 2

Location / Locality	%H₂	%H₂O	%N₂	%CO₂
Outer Hebrides, Scotland, UK				
Cleat, Barra	1.7×10^{-1}	9.2×10^1	2.9×10^{-1}	6.9×10^0
Barra Quarry, Barra	1.1×10^0	1.0×10^1	2.0×10^0	8.6×10^1
Allasdale Beach, Barra	1.3×10^{-1}	6.7×10^1	4.5×10^{-1}	3.3×10^1
Castlebay Pier, Barra	1.7×10^{-1}	9.9×10^1	1.3×10^{-1}	4.4×10^{-1}
Druim Reallasger Quarry, North Uist	1.2×10^0	7.8×10^1	7.2×10^0	1.2×10^1
Market Stance, South Uist	1.4×10^0	8.7×10^1	1.6×10^0	8.6×10^0
North Uist (control)	9.3×10^{-2}	9.7×10^1	1.5×10^0	4.2×10^{-1}
Near Loch Tollie, Northwest Scotland, UK				
West locality	4.8×10^{-1}	9.9×10^1	9.6×10^{-2}	3.7×10^{-1}
	3.4×10^{-2}	1.0×10^2	1.9×10^{-1}	1.5×10^{-1}
East locality	2.2×10^0	9.7×10^1	5.3×10^{-1}	4.3×10^{-1}
	1.6×10^0	9.8×10^1	4.2×10^{-1}	1.3×10^{-1}
West locality (control)	1.8×10^{-2}	9.9×10^1	2.3×10^{-1}	1.7×10^{-1}
East locality (control)	7.3×10^{-2}	1.0×10^2	2.7×10^{-1}	4.9×10^{-2}
Isles of Scilly, UK				
St. Agnes Island	1.1×10^1	7.5×10^1	8.3×10^0	4.0×10^0
	1.7×10^0	9.2×10^1	2.4×10^0	3.0×10^0
St. Agnes Island (control)	6.3×10^{-3}	9.7×10^1	1.1×10^0	1.8×10^0
	8.3×10^{-2}	9.8×10^1	8.3×10^{-1}	4.4×10^{-1}
Sudbury Crater, Canada				
Creighton Pluton, near Sudbury	2.0×10^0	9.6×10^1	1.6×10^0	7.7×10^{-2}
Vredefort Crater, South Africa				
Parys	4.3×10^{-1}	9.2×10^1	2.2×10^0	1.6×10^0
West Scotland, UK				
Achiltibuie	1.4×10^{-1}	9.7×10^1	3.1×10^{-1}	2.4×10^0
Skiag Bridge	8.8×10^{-2}	9.1×10^1	5.8×10^{-1}	8.2×10^0
Conival, Inchnadamph	5.7×10^{-2}	9.5×10^1	3.3×10^{-1}	4.6×10^0
Corry Bridge	1.1×10^0	9.7×10^1	4.0×10^{-1}	1.0×10^0
Coulags, Glen Carron (control)	6.0×10^{-2}	9.4×10^1	5.0×10^{-1}	5.0×10^0
Skiag Bridge (control)	8.8×10^{-2}	9.1×10^1	5.8×10^{-1}	8.2×10^0
Benmore Track (control)	1.2×10^{-2}	9.4×10^1	1.9×10^{-1}	5.3×10^0
Ord, Skye (control)	4.7×10^{-3}	9.8×10^1	1.4×10^{-1}	1.8×10^0
Northern Sweden				
Kiruna (control)	1.2×10^{-2}	8.9×10^1	5.2×10^{-1}	1.1×10^1

TABLE CAPTIONS

TABLE 1

Fault-rock and control samples used in hydrogen measurements. Samples included (a) gneiss- and granited-hosted pseudotachylites, (b) gneiss and granite controls for the pseudotachylites, (c) quartzite-hosted cataclasites, and (d) quartzite controls for the cataclasites. All pseudotachylite host rocks are Precambrian except for those from the Scilly Isles granite, which are about 290 Ma old (Chen *et al.*, 1993). All cataclasites and quartzite controls are Cambrian except for the Palaeoproterozoic control from Kiruna, Sweden. **Figure 1, 2, and 4** present the sample localities in context.

TABLE 2

Results of CFS analysis of major fluids in fault-rock and control samples. Each row represents the average of several fluid bursts released by one sample.

FIGURE CAPTIONS

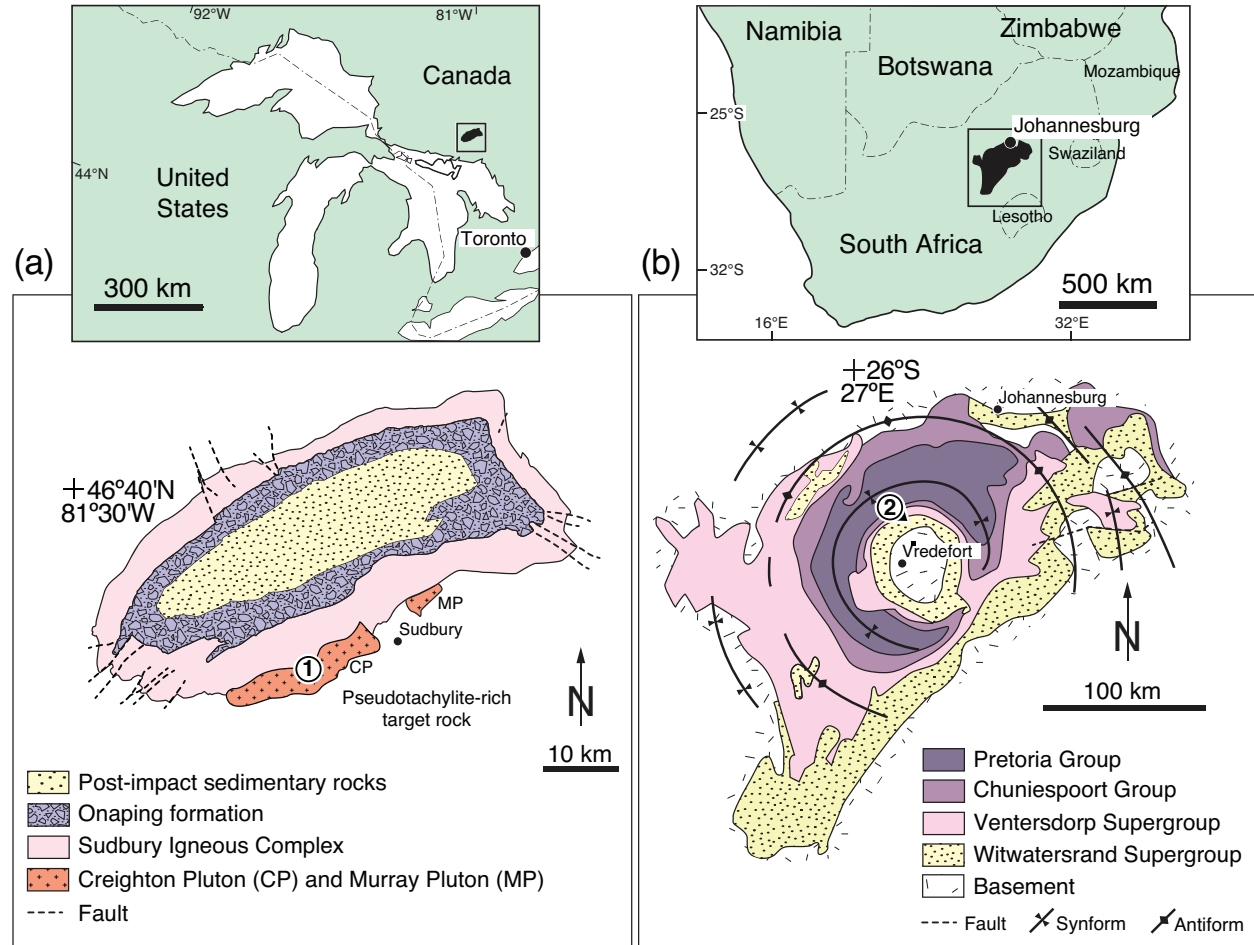


FIGURE 1

Sudbury (a) and Vredefort (b) impact structures: simplified geological maps and geographic context. In (a), (1) marks the location of Sample #1. MP = Murray Pluton; CP = Creighton Pluton. In (b), which indicates only pre-Karoo Supergroup exposure, (2) marks the location of Sample #2. Adapted from Riller *et al.* (2010), with details from Riller & Schwertdner (1997).

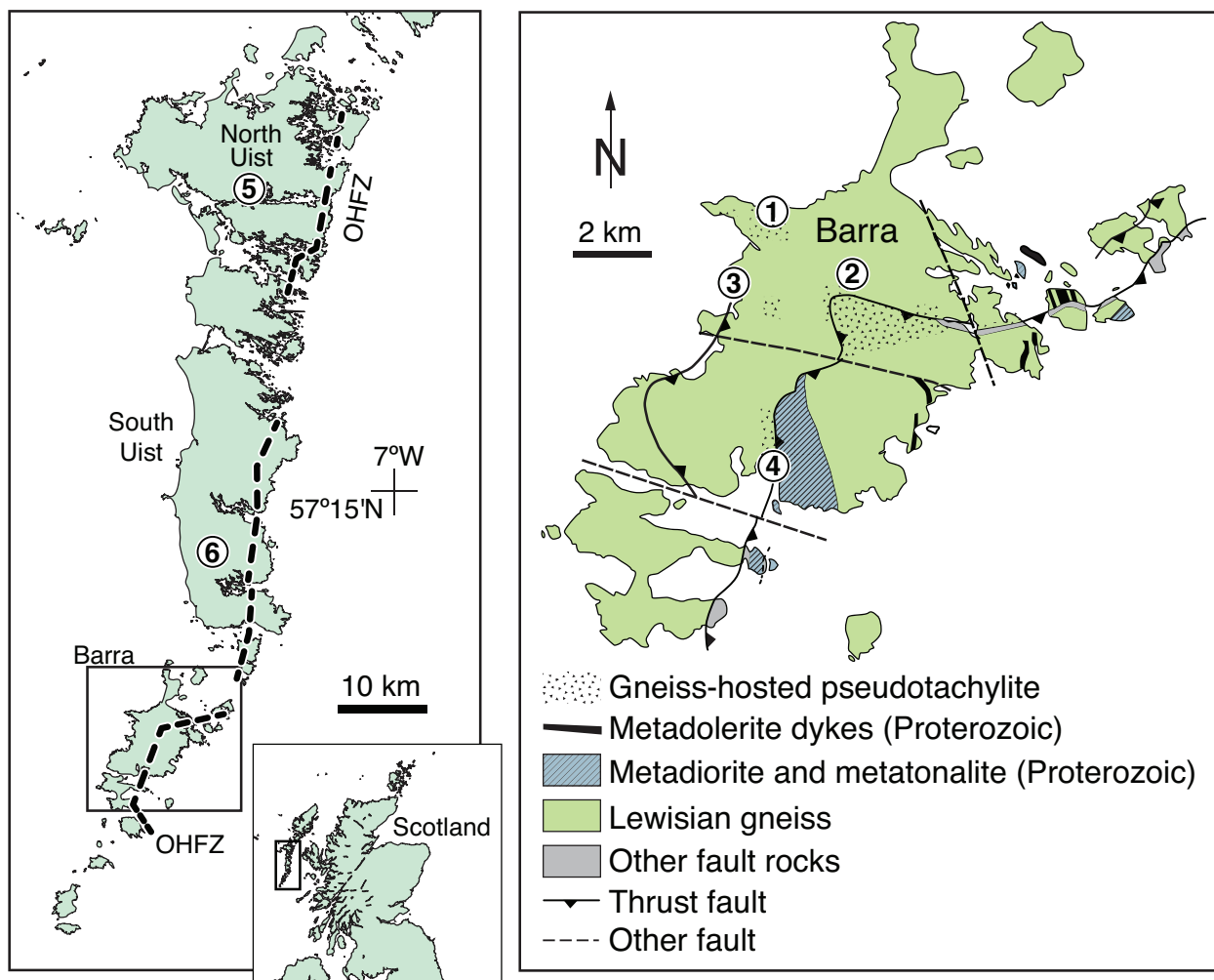


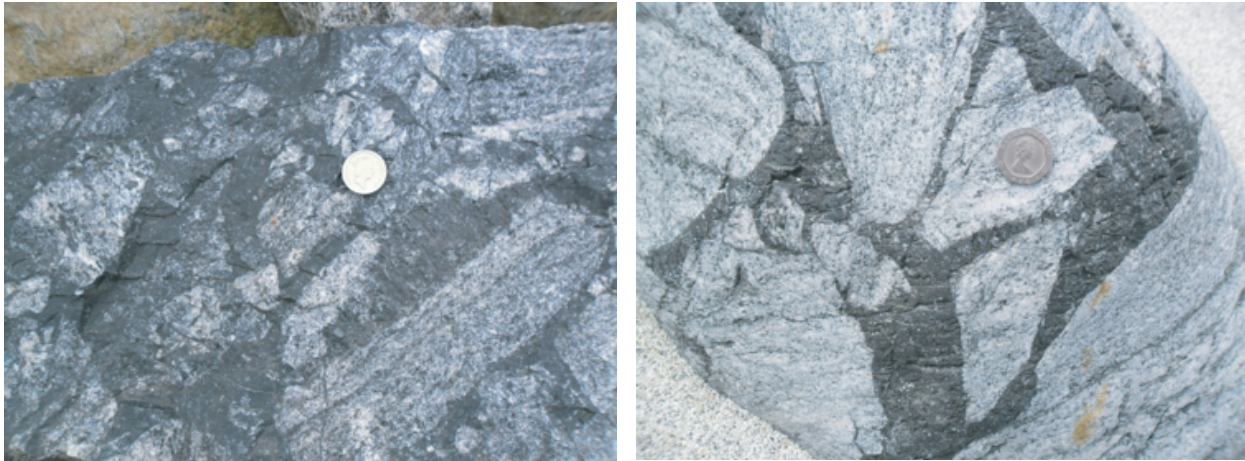
FIGURE 2

Figure 2: Pseudotachylite localities in the Outer Hebrides, and geological context on Barra.

OHFZ = Outer Hebrides Fault Zone. Adapted from Osinski *et al.* (2001) and Mendum *et al.*

(2009). See **Table 1** for key to sample localities.

A



B



FIGURE 3

Pseudotachylites from the British Isles. (A) Examples on the beaches of Barra, Outer Hebrides, UK. Pseudotachylites appear as the dark-coloured matrix between angular gneiss clasts. **(B) Examples on the beaches of St. Agnes Island, Scilly Isles, UK.** On the left, pseudotachylites appear as the dark-coloured matrix between eroded granite clasts. On the right, pseudotachylites occur *in situ* as thin dark veins through the granite (left to right). Coins on all images are about 20 mm across.

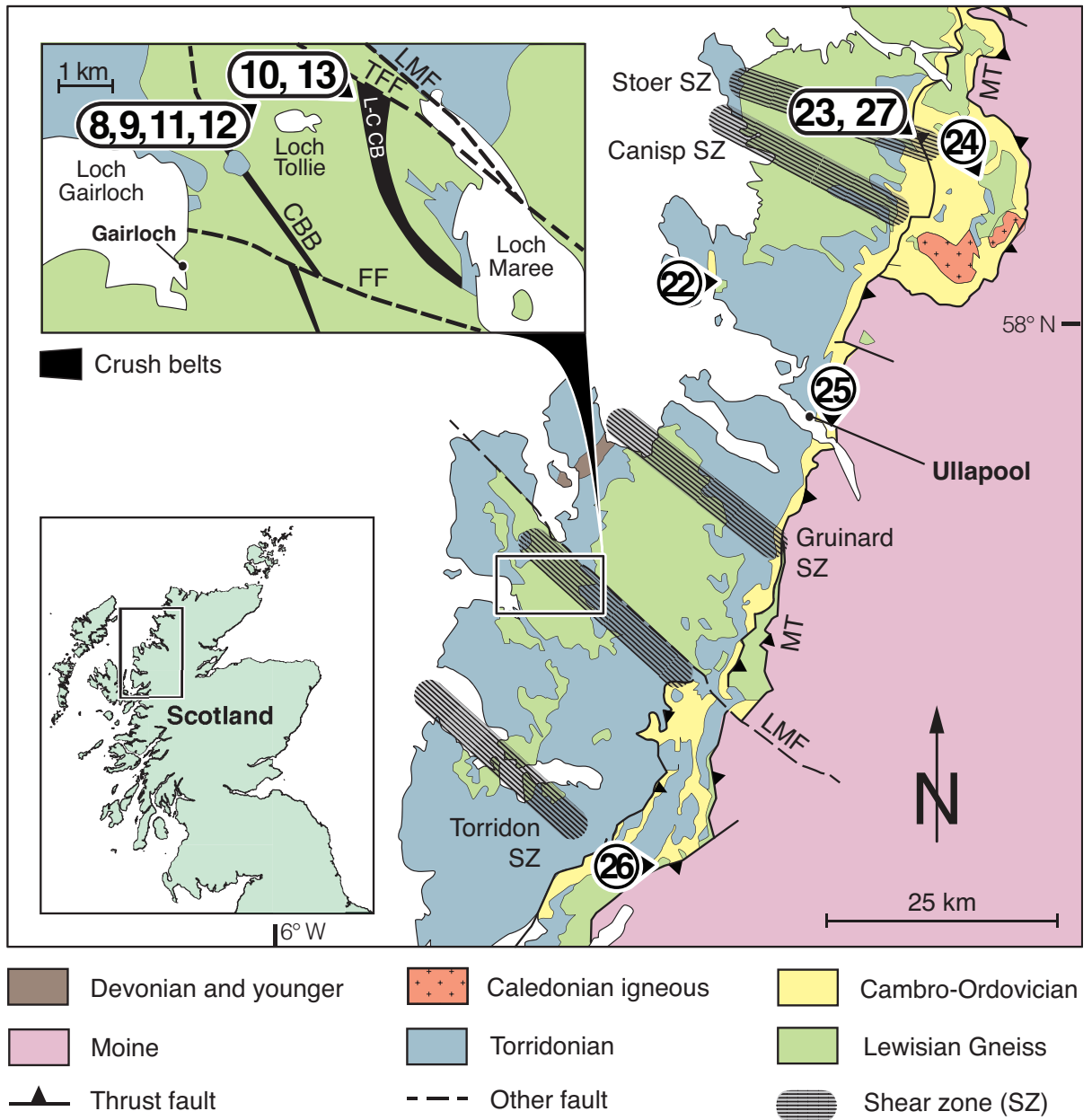


FIGURE 4

Sample localities in the Moine Thrust Belt, Northwest Highlands. Figure adapted from Krabbendam & Leslie (2010), with inset showing Gairloch region adapted from Sherlock *et al.* (2008). MT = Moine Thrust; LC = Langwell Culmination; DC = Dundonell Culmination; FH = Faraid Head; SZ = Shear zone. Details east of the Moine Thrust are not shown. LMF = Loch

Maree Fault; TFF = Tollie Farm fault; L-C CB = Leth-chreige crush belt; TA = Tollie antiform;
CBB = Creag Bahn belt; FF = Flowerdale fault. See **Table 1** for key to sample localities.

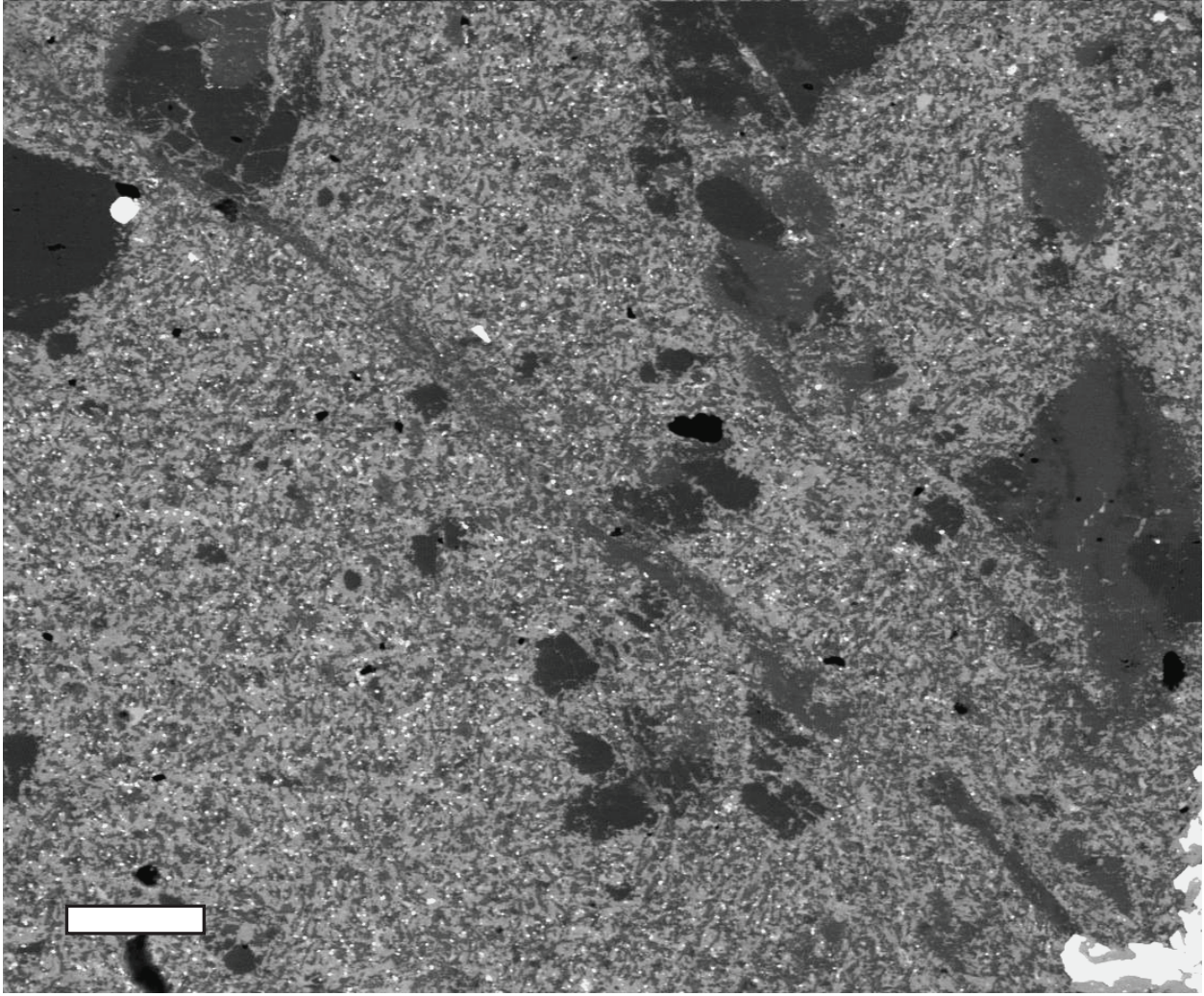


FIGURE 5

Scanning electron microscope image showing polished thin section of pseudotachylite from Loch Tollie in Northwest Scotland. Scale bar = 100 μm . Healed microfractures run from the upper left to the lower right of the field of view. Note large fractured and comminuted grains of quartz and feldspar suspended in the glassy groundmass. White areas (e.g. lower right) are pyrite; black grains are iron oxides.

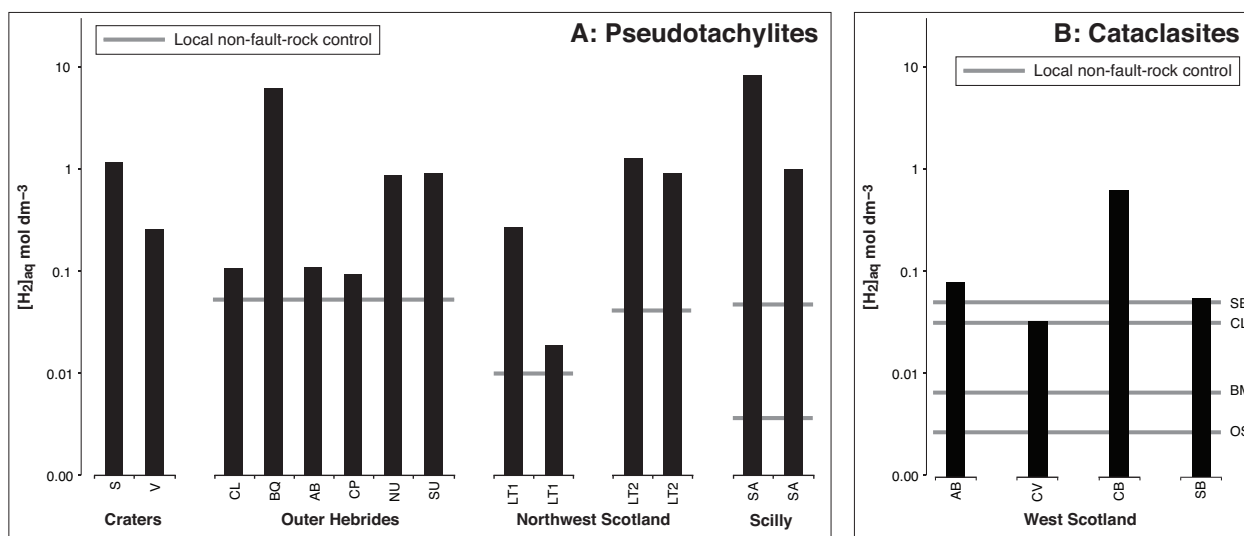


FIGURE 6

H₂ in pseudotachylites (A), cataclasites (B), and country rock control samples as reconstructed aqueous concentrations (mol dm⁻³). Note log axis. Craters: S = Sudbury, V= Vredefort. Outer Hebrides: CL = Cleat, Barra; BQ = Barra Quarry; AB = Allasdale Beach, Barra; CP = Castlebay Pier, Barra; NU = North Uist; SU = South Uist. Northwest Scotland: LT1 = Loch Tollie West; LT2 = Loch Tollie East. Scilly: SA = St Agnes Island. Cataclasites: AB = Achiltibuie; CV = Conival; CB = Corry Bridge; SB = Skiag Bridge; CL = Coulags; BM = Ben More; OS = Ord, Skye.

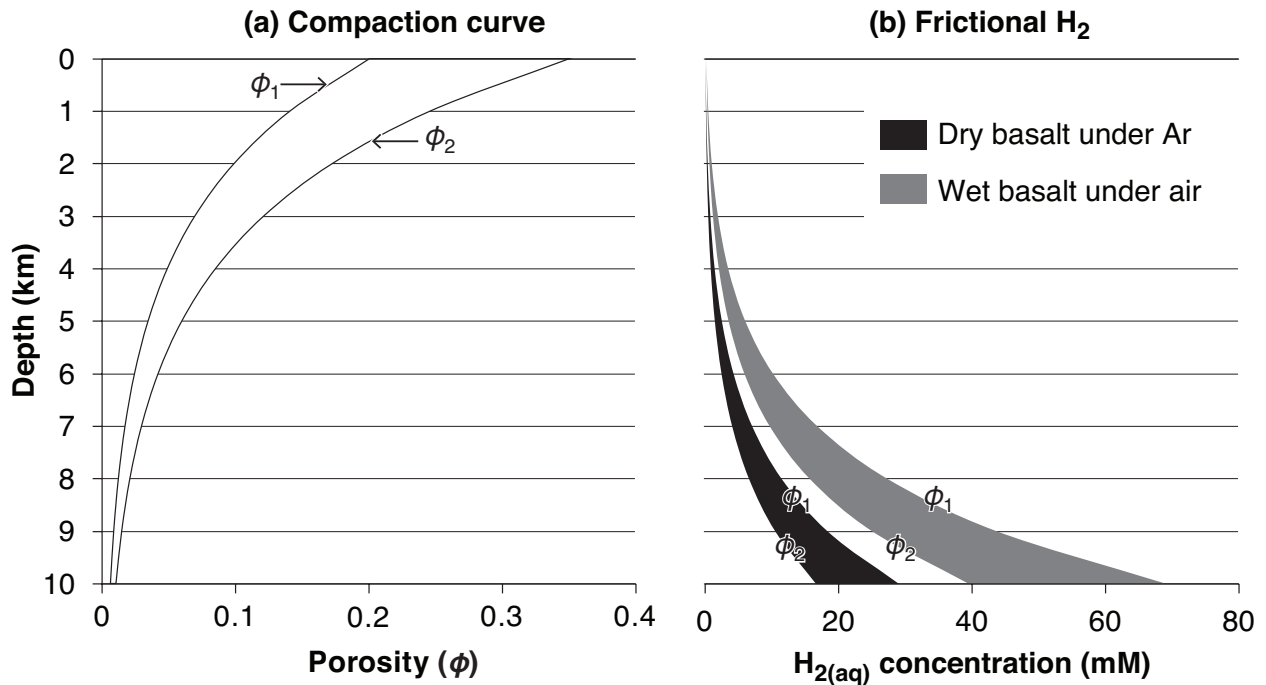


FIGURE 7

Approximate instantaneous seismogenic H_{2(aq)} concentrations after a Mars-quake as a function of depth and porosity below the surface. (a) Maximum (ϕ_2) and minimum (ϕ_1) porosity curves for martian crust proposed by Clifford *et al.* (2010). **(b)** Seismogenic (frictional) H_{2(aq)} concentrations in fault-zone groundwater according to the model from Hirose *et al.* (2011) and the porosity curves in (a), showing concentrations well above the threshold for microbial utility ($\ll 1$ mM). The experiments of Hirose *et al.* (2011, 2012) provide two values of α , an empirical coefficient used in the calculation (**Equation 1**); the black curve is obtained using α measured for dry (desiccated) basalt under argon gas; the grey curve is obtained using α measured for wet (water-saturated) basalt under air. Note that the saturation point of H₂ in water is about 1 mM under standard conditions.