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Fuel movement along grain boundaries in ice

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Abstract

Knowledge about movement of hydrocarbon-based fuels in ice is relevant to marine and terrestrial fuel spills in polar regions, and to the dispersion of hydrocarbon-based drilling fluids used to maintain lithostatic pressure in deep ice coring projects. The latter is of concern from both environmental and sample integrity aspects, particularly for projects that plan to sample deep subglacial environments such as Lake Vostok. These concerns prompted us to examine the mobility of a liquid hydrocarbon fuel in non-fractured polycrystalline ice near the melting point in darkened conditions characteristic of deep glacial ice. Our experiments revealed rapid fuel propagation along intergranular tubes, at velocities up to 1.6 m h^{-1}, in ice with an overall impurity concentration ranging from 7 to 10 μM. This fuel movement, herein referred to as fuel tunneling, was absent in ice grown from distilled water with two- to three-fold fewer impurities, suggesting a role of dissolved contaminants which tend to concentrate along grain boundaries. Fuel tunneling commenced when ice temperatures were within 0.2 °C of melting and water vein diameters about 0.5 mm. Our results show that bubble free, non-fractured polycrystalline ice near its melting point is highly permeable to liquid hydrocarbon fuel via grain boundaries, a result that should be considered when sampling deep subglacial lakes, particularly those with overlying accretion ice.
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1. Introduction

Ice near the melting point is inherently a two-phase material consisting of solid crystals separated by films and channels of intergranular liquid water (Nye and Frank, 1973; Raymond and Harrison, 1975). Impurity solutes in ice tend to concentrate along grain boundaries (Renaud, 1949), particularly the grain boundary junctions which form the intersections between three or more grains (Harrison and Raymond, 1976). The combined effects of solutes in grain boundary junctions and high surface energy along grain edges (Ketcham and Hobbs, 1969) result in a depression of the phase equilibrium temperature known as undercooling (Rempel et al., 2002). This phenomenon results in the thermodynamic stability of intergranular liquid water channels known as water veins. The cross-sectional areas of water veins adjust to changes in the environment, including temperature and intergranular solute concentration among others (Lliboutry, 1971; Raymond and Harrison, 1975). These adjustments are

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linked to the permeability of polycrystalline ice near the melting point and may contribute to the infiltration processes of a hydrocarbon fluid.

Water veins are believed to be exceptionally large in accretion ice frozen to the ceiling of Antarctic subglacial Lake Vostok due to the presence of large crystals and warm temperatures (Price, 2000). Over 145 of these subglacial lakes are known to be concentrated at the base of the Antarctic ice sheet where the ice thickness exceeds 3 km (Siegert et al., 2005; Bell et al., 2006). Although these lakes have yet to be sampled directly, biological studies of accretion ice over subglacial Lake Vostok imply that these lakes, and their overlying accretion ice, provide a habitat isolated from the Earth's surface for hundreds of millenia (Priscu et al., 1999; Karl et al., 1999; Price, 2000; Siegert et al., 2001). Subglacial lake sampling will require a deep borehole drill utilizing a compensation fluid to prevent closure resulting from plastic deformation (Gosink et al., 1991; Wumkes, 1994; Talalay and Gundestrup, 2002). Kerosene-based liquid hydrocarbon (LH) is a primary component of many borehole fluids because of its desirable properties of low freezing temperature and ice-like density. Sixty tons of LH-based drilling fluid is now present in the deep borehole at Vostok Station, which is planned to be used as the initial sampling site for Lake Vostok (Inman, 2005). Care must be taken to maintain sample integrity when using LH drilling fluid, particularly when sampling for extant life, because the fluid can be a carbon source for certain heterotrophic psychrotolerant microbes (Whyte et al., 2001; Aislabie et al., 2004). Recent work has shown that LH fluid in the Vostok borehole contains bacterial cells in quantities several orders of magnitude greater than the ice itself (Christner et al., 2005), making conclusions about the actual nature of the ice difficult without stringent sample decontamination once the ice is returned to the surface. Accidental contamination of a subglacial lake with drilling fluid could disrupt the natural ecosystem both geochemically and biologically, making conclusions about the actual nature of these unique lakes equivocal, not to mention the ethical issues that surround contamination of these pristine systems (Inman, 2005). Similar conclusions apply to the proposed biological and geochemical sampling of other frozen worlds such as Mars and Europa (Rummel, 2001; Christner et al., 2005; National Research Council, 2006). Fuel and oil spills are also a significant anthropogenic source of pollution in polar regions (Kennicutt et al., 1990; Tumeo and Wolk, 1994; Simpson et al., 1995). Such pollution is a seemingly inevitable consequence of activities associated with fuel storage, transportation and borehole drilling.

Past studies of LH fuel movement through ice have considered the effects of contaminant surface adhesion (Liukkonen et al., 1997) and the vertical and horizontal spreading rates (Tumeo and Larson, 1994). However, many questions remain concerning the fundamental physical mechanisms involved. An understanding of how contaminants would migrate through the cryosphere is essential for informed decision-making during mitigation and basic research efforts in cold regions. The purpose of our study was to examine the mobility of LH fuel in near-melting ice, via the intergranular water vein network, in darkened conditions characteristic of deep glacial ice. This mode of contaminant transport has particular relevance in climatological, geochemical and biological studies of deep ice and associated subglacial systems beneath the Antarctic and Greenland ice sheets.

2. Methods

Three experiments were carried out in a cold laboratory to observe the mobility of one type of LH fuel, JP-8, in clear, bubble free, non-fractured ice blocks near the melting point. This fuel is in standard use for military and commercial aircraft and is very similar to the compensation fluid used in deep ice core drilling operations (Victor, 2003). The measured specific gravity of the JP-8 was 0.805, with a standard deviation of 0.006 for five samples. In order to approach the environment of subglacial lake accretion ice, the experimental conditions included darkness, temperatures within several degrees of melting, and ice structure lacking fractures and bubbles. Blocks 1, 2 and 3, each measuring 16×16 cm horizontally and 13 cm vertically, refer to the ice samples tested in each respective experiment. The ice was grown in an upward direction within a reservoir of water circulated by a mechanical pump. The resulting ice was clear, free of both bubbles and fractures, and contained vertically-elongated grains with horizontal dimensions of 5 to 70 mm and vertical dimensions of 90 to 280 mm. Crystallographic orientations were examined using a Rigsby stage, an instrument equipped with a four-axis stage and cross-polarized filters. Most crystallographic c-axes were found to be aligned near the horizontal plane, but otherwise random, thus resembling type S2 columnar ice. Major ion concentrations in the ice were measured using ion chromatography (see Table 1). The ice in experiments 1 and 2 was grown from tap water and contained 7 and 10 μM of bulk impurities, respectively, while the ice in experiment 3 was grown from distilled water and contained 3.5 μM of bulk impurities (Table 1). The experiments were conducted within a temperature-controlled environmental chamber ventilated by a carbon air filtration system. The ice temperature, measured with Type-T thermocouples (accuracy ~0.2 °C), was varied by adjusting air temperature settings. A digital camera with a

Table 1
Major ion concentrations (micromoles per liter [µM] meltwater) in samples taken from ice composed of tapwater (Tap) and distilled water (Distilled)

Ice	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Total
Tap (Exp 1)	5.35	0.50	0.06	1.76	1.94	0.32	0.21	10.14
Tap (Exp 2)	2.29	0.71	0.07	1.77	1.65	0.11	0.10	6.71
Distilled (Exp 3)	0.35	0.25	0.05	1.61	1.04	0.06	0.07	3.45

The meltwater samples were obtained from ice sections cut from the top of the ice blocks (see Fig. 1). The experiment (Exp) is given in parentheses after the ice type.

1:1 macro lens on manual focus was used to document visual developments. Before each experiment, a cm-thick horizontal section was cut from the top of the ice block and photographed between cross-polarized film to determine the locations of grain boundaries relative to the planned placement of fuel (Fig. 1). These horizontal sections were then submitted for chemical analyses using ion chromatography, the results of which are listed in Table 1. After removing a horizontal section from the ice, a cylindrical-shaped pocket, 55 mm wide and 25 mm deep, was melted into the upper surface of the remaining ice. Each experiment began at a reference time of 0 h when 25 ml of JP-8 fuel, brought to ice temperature, was added to the cylindrical pocket in the ice. At the conclusion of each experiment, the ice was cut into cm-thick horizontal sections and examined between cross-polarized films for the residual traces of fuel movement relative to the crystal structure.

The format of the first experiment differed from that of experiments 2 and 3. In experiment 1, Sudan IV red solvent dye was added to the JP-8 to improve its visual detection. Observations were made as the ice temperature was gradually increased from an initial value of

Table 2
Summary of (a) ice temperatures and rates of meltpool growth in ice, and (b) fuel tunneling, initiation times and fuel tube propagation rates

(a) Ice temperatures and meltpool growth rates:

Exp	T_i (°C)	t_{-1} (h)	$t_{-0.2}$ (h)	t_{mg} (h)	v_{vg} (mm h^{-1})	v_{hg} (mm h^{-1})
1	−4	74	99	0–104	0.07	0.1
2	−2	2	4	0–24	1	0.6
3	−2	<1	2	0–72 (end)	1	1.6

(b) Fuel tunneling, initiation times and fuel tube propagation rates:

Exp	F.T.?	Tubes observed	Initiation time of F.T. (h)	v_{ft} (mm h^{-1})
1	Yes	Tube 1-1	104	n/m
2	Yes	Tube 2-1	24	>160
		Tube 2-2	32	1600
3	No	None	n/a	n/a

Abbreviations: T_i=initial ice temperature, t_{-1}=time ice temperature reached −1 °C, $t_{-0.2}$=time ice temperature reached −0.2 °C, t_{mg}=time range of meltpool growth only, v_{vg}=average rate of vertical meltpool growth, v_{hg}=average rate of horizontal meltpool growth, F.T.=fuel tunneling, v_{ft}=fuel tube propagation rate, Exp=experiment, n/m=not measured, n/a=not applicable.

−4 °C. The ice temperature had exceeded −1 °C at approximately 74 h, and was within thermocouple accuracy (∼0.2 °C) of the melting point by approximately 99 h (Table 2a). Images of the ice and fuel were obtained manually two times a day. An evaluation of the experiment 1 protocol led to several improvements in the format of the two experiments that followed. In these, no dye was added to the JP-8 because it was unnecessary to track the fuel movement of interest. The ice temperature in experiments 2 and 3 was increased from an initial value of −2 °C to a value near the melting point (>−0.2 °C) in under 4 h (Table 2a). Images were
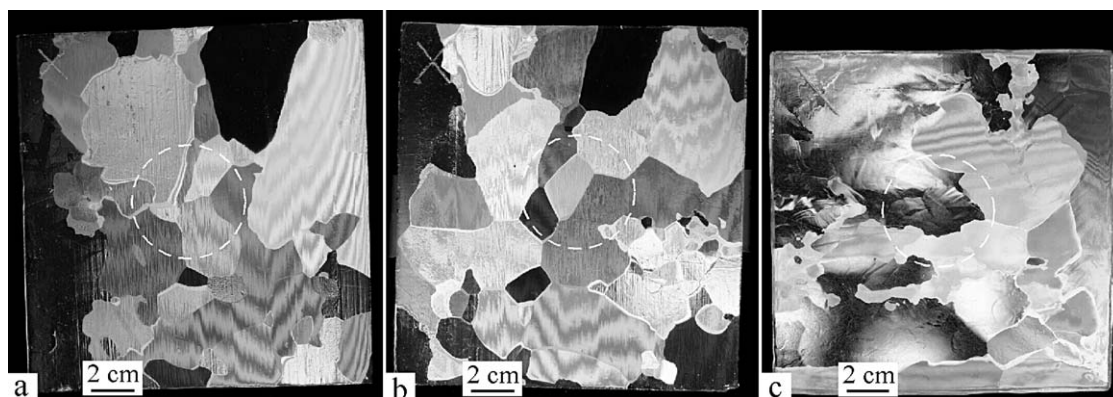

Fig. 1. Cross-polarized images of cm-thick, horizontal ice sections removed from the top of ice blocks tested in (a) experiment 1, (b) experiment 2 and (c) experiment 3. White circles are vertical projections of the initial fuel pocket boundaries from the ice beneath the sections shown.

acquired using remote capture software programmed in accordance with the speed of visual changes.

3. Results

The JP-8 fuel preferentially melted the lower pocket surfaces to form meltpools with bulbous and scallop-shaped boundaries (region in Fig. 2a above tube) during the initial phases of the experiments (Table 2a). These meltpools consisted of an aqueous solution formed by melting of ice, overlain by a floating layer of water-immiscible fuel. The fuel pockets in experiments 2 and 3 grew deeper at an average rate of about 1 mm h^{-1} and wider at average rates of between 0.6 and 1.6 mm h^{-1} (Table 2a). These melt rates in experiment 1 were much slower due to the lower imposed temperatures (Table 2a). Unexpectedly, the fuel drained through the ice of experiments 1 and 2 at elapsed times of 104 and 24 h,

respectively (Fig. 2a, b; Table 2b). The tubes which channeled fuel through blocks 1 and 2 are referred to as tube 1-1 and 2-1, respectively (Fig. 2a, b). These drainage events initiated 30 and 22 h after the temperatures of ice blocks 1 and 2, respectively, had risen above −1 °C (Table 2). The presence of surficial liquid water and stationary thermocouple readings near initiation of fuel tunneling indicated ice temperatures within 0.2 °C of melting. Tube 1-1 was first observed at the end of experiment 1, within 4 h of its formation based on times between successive, manually-acquired images (Fig. 2a). At this time, the tube had a diameter of 2 mm and branched into three segments near the bottom of the block (Fig. 2a). This branch structure delineated a water vein node (discussed below), the point where multiple grain boundary junctions intersect. Tube 2-1 of experiment 2 had a diameter of 1 mm and propagated downward at a velocity of about 160 mm h^{-1} (Fig. 2b). All fuel drained
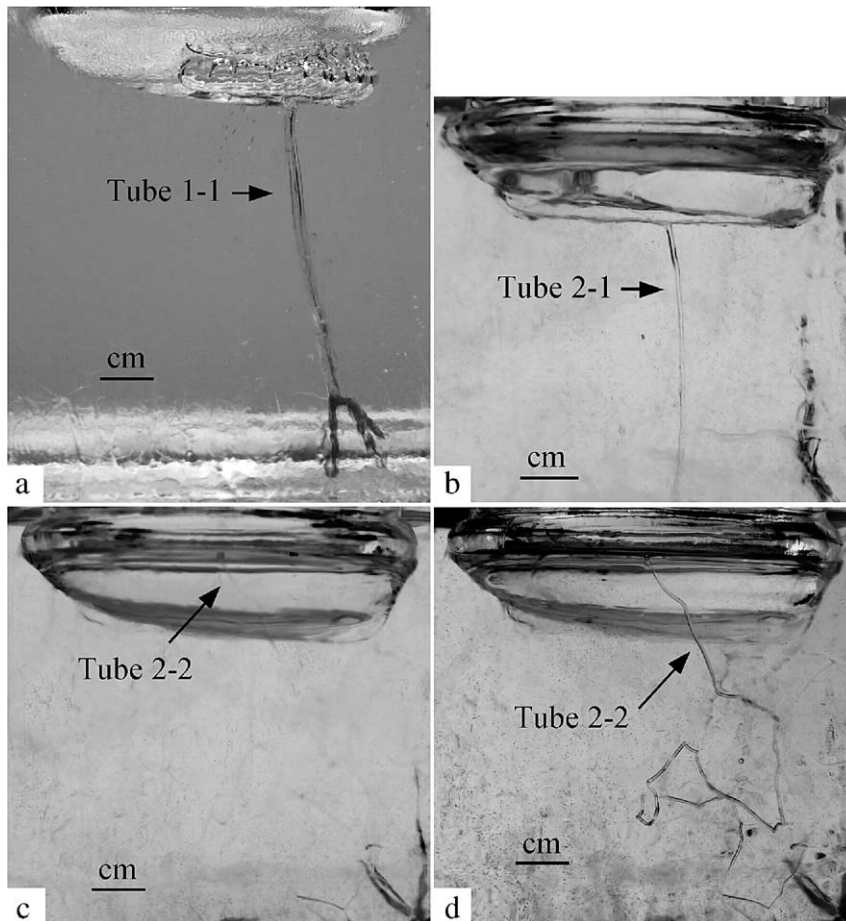

Fig. 2. Meltpools and fuel tubes in experiments 1 and 2. (a) Fuel tube 1-1 in block 1, 106 h into experiment 1; (b) fuel tube 2-1 in block 2, 24 h into experiment 2; (c) first visible movement of fuel in tube 2-2, 32 h into experiment 2; (d) tube 2-2 three minutes after first visible fuel movement shown in Fig. 2c.

through this tube during a time interval lasting between 0.5 and 1.5 h, yielding an average flow rate of between 17 and 50 ml h^{-1}. After this draining event, tube 2-1 was no longer visually apparent and had presumably frozen closed.

After all fuel drained through tube 2-1, the surface pocket was refilled with 25 ml of JP-8. Interestingly, the original tube was not reactivated with the addition of this fuel. Instead, four minutes after adding the fuel, a new fuel tube became visible in the upper front side of the pocket (Fig. 2c, d). This tube, referred to as tube 2-2, had an initial diameter of 0.5 mm and propagated downward (compare Fig. 2c, d). Within three minutes of the tube's appearance, the fuel had traversed the entire 80 mm vertical distance of ice beneath the pocket (Fig. 2d) indicating a propagation rate of about 1.6 m h^{-1}. The diameter of the tube increased with time, reaching 2 mm one hour after its initial detection (Fig. 3a). The tube also turned upward and bifurcated at several points, branching out in the direction of fuel movement (Fig. 3a). The contents of the tube consisted of a distinct, alternating sequence of immiscible phases resembling bubbles in a fluid. This "bubble-like" structure had also been observed in a fuel tube during a separate, earlier experiment

during which simulated solar radiation had been applied (Fig. 3b).

One of the most significant observations was that fuel tunneling never occurred during the third experiment testing ice grown from distilled water with an overall impurity concentration of 3.5 μM, a factor of two to three lower than that of the ice grown from tap water. At the end of experiment 3 (72 h), the melt pool remained and had grown to a depth of 28 mm and width of 97 mm in ice which had been maintained near the melting point (>-0.2 °C) for 70 h (Fig. 3c). Visual inspections near the end of experiment 3 revealed numerous water veins, estimated to be 0.1 mm in diameter, intersecting the lower surface of the meltpool. The intersection of water veins with the fuel pocket is also supported by the grain boundary locations in ice cut from the top of the block prior to adding fuel (Fig. 1c). It should also be noted that the horizontal area of the fuel pocket had increased from that indicated in Fig. 1c by about 350% during experiment 3.

Examinations were made of the fuel tube locations relative to crystal boundaries in the ice of experiments 1 and 2. The dyed fluid in experiment 1 collected in grain
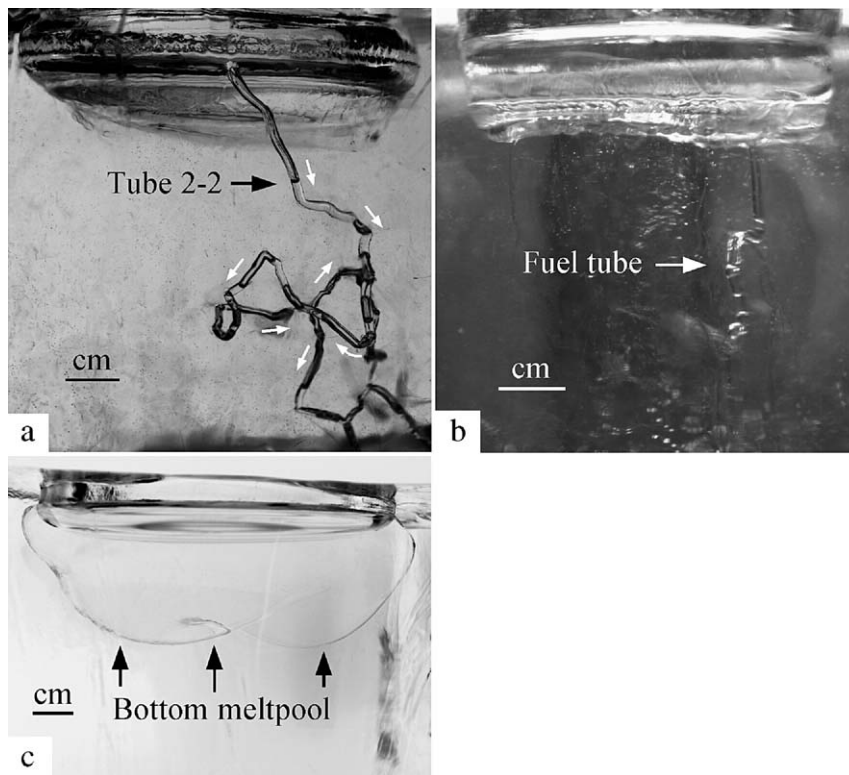

Fig. 3. Fuel tubes and meltpools in ice. (a) Fuel tube 2-2, 33 h into experiment 2, white arrows indicating the direction of fuel movement. (b) Fuel tube in a separate, earlier experiment which applied simulated sunlight. (c) View of meltpool in distilled water ice, 70 h into experiment 3, in which no fuel tubes formed.
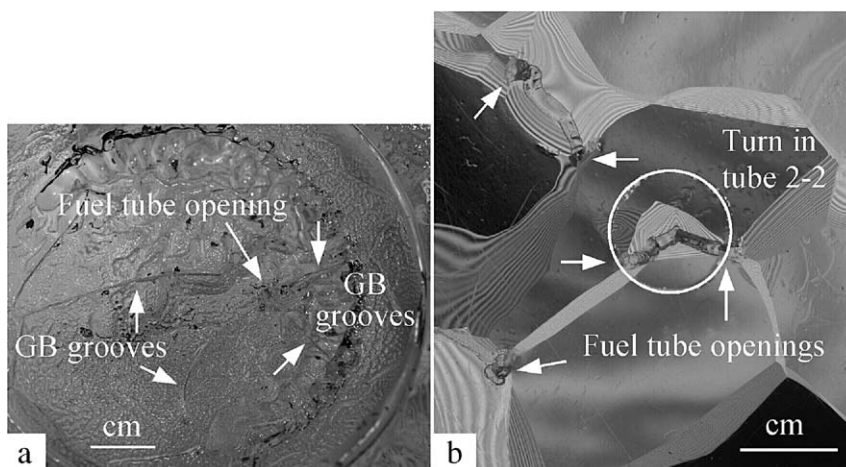
Fig. 4. Locations of grain boundaries (GB) relative to fuel tubes. (a) Intersection of GB grooves at the opening of fuel tube 1-1, bottom of empty fuel pocket. (b) Cross-polarized image of horizontal section cut from block 2 showing coincidence of fuel tubes and triple junctions. The arrows indicate fuel tube openings, the circle demarcates a ∼180° turn in tube 2-2.

boundary grooves in the bottom of the empty fuel pocket (Fig. 4a). Four of these grooves intersected at the opening of tube 1-1, indicating that the tube coincided with a four-grain junction. At the end of experiment 2, a polaroscopic examination of horizontal ice sections confirmed that all segments of tube 2-2 followed grain boundary junctions (Fig. 4b). At one location, the tube turned to follow the apex of a crystal rather than continuing on through a simple grain boundary (circle in Fig. 4b), indicating the preference of a triple junction over a simple grain boundary for the path of fuel. Visual examinations during experiments 1 and 2 indicated circular to oval fuel tube cross sections, not the triangular shape that might be expected based on thermodynamic considerations (Nye and Frank, 1973) and previous experimental observations (Ketcham and Hobbs, 1969).

4. Discussion and conclusion

According to past estimates addressing diffusion through water veins, hydrocarbon-based drilling fluid would migrate through 1 m of ice in about 10^3 years (Victor, 2003). This estimate did not consider the possibility of vertical fluid advection in association with phase changes along grain boundary junctions (developed below). Our experiments revealed that the fuel–water mixtures moved through ice at rates reaching 1.6 m h^{-1} via fluid advection associated with internal phase changes and not just diffusion. The rapid fuel movement occurred in ice made from tap water, but not in ice made of distilled water. We interpret this finding as related to two contrasting properties of the ice tested: (1) an overall impurity concentration in the distilled water ice two to three times

lower than in the tap water ice, and (2) the serrated grain boundary texture in the distilled water ice (Fig. 1c) which would act to increase the surface area of each grain and possibly reduce the quantity of impurities allocated to triple junctions. Other factors being equal (e.g., grain size, grain structure, temperature), fewer impurities per unit length of grain edge would decrease water vein cross-sectional areas (Raymond and Harrison, 1975; Nye, 1991a,b). This may in turn lower the "availability" of liquid channels for fuel tunneling.

The first visible fuel movement in ice blocks 1 and 2 occurred 30 and 22 h, respectively, after the ice temperature rose above −1 °C. The temperature of block 2 appeared stationary during the 20 h period preceding the first detectable fuel movement (Table 2). In comparison, the distilled water ice, through which no visible fuel movement occurred, remained in this thermal state for 70 h (Table 2). During this quiescent period, we contend
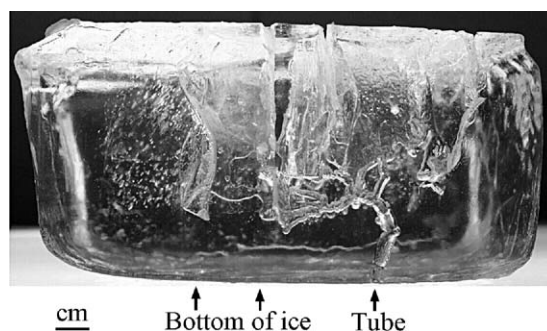

Fig. 5. Tube extending up into an ice block after floating in a near-freezing, 39 mM NaCl solution. The tube has a maximum diameter of 4 mm, tapers and branches out in an upward direction, reverse to that observed in the experiments using fuel.

that aqueous hydrocarbons diffused from the meltpools downward into the ice through water veins. These hydrocarbons would have included the lighter aromatics, the most water soluble components of JP-8, comprising roughly 20% of the fuel (Arco, 1998; Talalay and Gundestrup, 2002; Gerasimoff, 2003). The influx of aqueous hydrocarbons into water veins would cause them to expand in order to approach an equilibrium solute concentration. Based on two separate observations in experiment 2 (one image in Fig. 2c–d, other not shown), fuel tunneling initiated when water vein diameters reached ~0.5 mm. These diameters are noted to be four times larger than those reported in a microscopic study on water veins in *well-rotted* ice grown from distilled water, where the temperature was maintained very close to the melting point (Mader, 1992a). Our results led us to hypothesize that at a critical water vein diameter or expansion rate, the transport process of diffusion shifts suddenly to a different process driven by phase change. The specific volume decrease upon melting around water veins would induce localized pressure gradients. At a critical water vein diameter or expansion rate, these pressure gradients may become sufficient to suction the non-aqueous fuel downward along water veins in a non-gravitationally driven process. Just such a mechanism has been predicted to cause fluid flow along water veins (Nye, 1991a) and has been experimentally observed, and referred to as "pumping," by Mader (1992b). Our hypothesis was tested in an additional experiment to observe ice, of the type used in experiments 1 and 2, floating in a near-freezing, aqueous 39 mM NaCl solution. This concentration is an order of magnitude less than is typical for seawater (Michel, 1978, p. 38). Based on our previous experiments, we hypothesized that the dissolved solutes would diffuse upward into the ice, eventually leading to upward suctioning of the solution. About 5 h into this experiment, two large tubes were observed extending upward into the ice block from its lower surface, one of which is shown in Fig. 5. These tubes had a maximum diameter of 4 mm and branched out into smaller tubes in an upward direction, opposite to that observed in earlier experiments 1 and 2 (Figs. 2a and 3a). These findings confirmed our hypothesis that the saltwater would travel upward into the ice.

Fuel tunneling may represent a significant mechanism of contaminant transport in non-fractured ice near the melting point, particularly in ice containing large water veins. These conditions likely exist in accretion ice which forms the ceiling of Lake Vostok (Jouzel et al., 1999; Price, 2000), as well as those of other deep subglacial lakes (e.g., Price et al., 2002; Siegert et al., 2004). The best documented accretion ice system occurs above Lake Vostok, Eastern Antarctica, which lies ~4 km beneath the surface of the ice sheet (Siegert et al., 2001). Water vein diameters larger than 0.5 mm have been predicted in the lower 35 m of the Vostok accretion ice, with values increasing toward the lake water (Price, 2000). A critical assumption of the Price (2000) work is that all impurities are concentrated in liquid water solutions following triple junctions. We observed water veins with a diameter of 0.5 mm at the initiation of fuel tunneling (Fig. 2c, d) in the laboratory ice grown for the experiments described in this paper. Our results, in concert with those reported for the Vostok accretion ice (Price, 2000), indicate that fuel tunneling from the LH fluid reservoir in the Vostok borehole to the underlying lake water will become more likely as the residence time of fluids in the borehole increases and as the borehole distance from lakewater decreases.

Acknowledgements

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