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Original Article

On The Survival Curve of Repeated Nucleation of a Single Volume of Supercooled Water

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Abstract

If a sample of water is repeatedly cooled into the supercooled region until it eventually nucleates and freezes, the resulting data on nucleation temperatures can be plotted as a survival curve. We look here at the slope of such curves and suggest some reasons for both a changing slope, either with time or with more repeats, and also for the slope being related to multiple nucleation sites, either at the same time or perhaps subatomic particles, such as neutrons, passing through the sample and causing nucleation, rather than an impurity or defect on the container wall.

Introduction

Previously, while working at the University of Tsukuba in Japan, one of us (Wilson) looked closely at the shape and slope of survival curves for water repeatedly frozen and thawed [1]. Figure 1 shows a typical data set gathered from the device coined an automatic lag time apparatus (ALTA), which has been described many times by Haymet and his co-authors [1,2]. As the curve is asymptotic at both ends it is sensible to describe the 10-90 width as a measure of the spread/slope. It has been shown that for a single volume of clean water in a single container cooled and frozen and thawed many hundreds of times, that the 10-90 width seems to settle at about 0.7°C, as seen in Figure 1.

We report here results of several recent experiments. In the first set, carried out in Hastings, New Zealand, we have simply taken a commercial bottle of spring water, unopened, 250 mL, and attached a thermocouple to the outside and placed in a freezer. Figure 2 shows 14 runs of the same water, with warming to 20 °C between each run.

IIn the case of the data presented in Figure 2, by -2°C the bottle had never frozen and by -6 °C it had frozen each time. The 10-90 width is roughly 2°C. The larger 10-90 width than Figure 1 is due to the low number of repeats.

In a second set of experiments, carried out in Sydney, Australia, we looked at tube-dependence on nucleation temperature. We used five glass nmr tubes of 5 mm OD, each with 200 µl of distilled water, capped and suspended

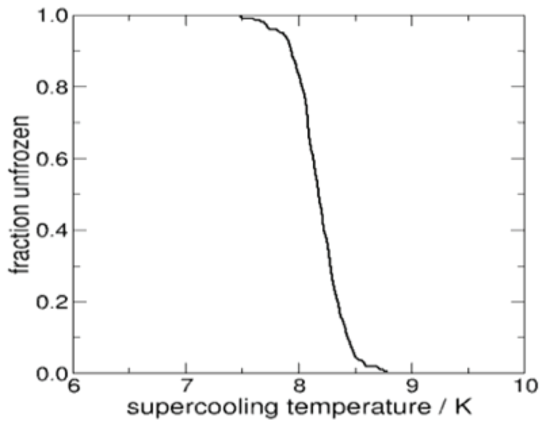

***Figure 1**. Data from an automated lag time apparatus, by the design of Haymet and Wilson [1,2], showing a typical survival curve for a single sample of clean water cooled to nucleation temperature some 350 times. In this case, the 'average' nucleation temperature, also known as the T50, the temperature by which the linearly cooling sample had frozen 50% of the time, is about -8.3°C and the 10-90 width is about 0.7°C.*

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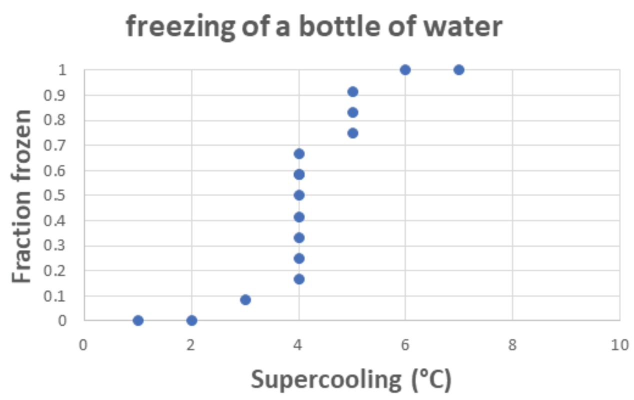

Figure 2. *Data which could, given sufficient repeats, form a survival curve for the nucleation of a bottle of commercial mineral water. The first and last data points are added only to show what an asymptotic curve would look like, as an aid to the eye.*

Table 1. *Six glass tubes of water, each cooled and warmed seven times. As the water bath was cooled linearly and the tubes became colder, the freeze events are in order, i.e. tube one always froze at the warmest temperature, and tube 4 at the slightly colder temperatures and so on.*

Run	Order of nucleation form first(left) to last (right)					
1	Tube 1	Tube 4	Tube 6	Tube 2	Tube 5	Tube 3
2	Tube 1	Tube 4	Tube 6	Tube 2	Tube 5	Tube 3
3	Tube 1	Tube 4	Tube 6	Tube 5	Tube 2	Tube 3
4	Tube 1	Tube 4	Cracked	Tube 2	Tube 5	Tube 3
5	Tube 1	Tube 4	Cracked	Cracked	Tube 5	Tube 3
6	Tube 1	Tube 4	Cracked	Cracked	Tube 5	Tube 3
7	Tube 1	Tube 4	Cracked	Cracked	Tube 5	Tube 3

into a refrigerated water/alcohol bath, with stirring, allowing for subzero temperature, largely isothermal, fluid to surround the tubes. Thermocouples were attached to the outside of each tube and data recorded electronically. These results (Table 1) show the very clear tube-dependence of heterogeneous nucleation, occurring most likely at the inside of the tube wall, as opposed to a piece of dirt, or other impurity. If the most efficient site does not change, then neither does the nucleation temperature, beyond what is expected from heterogeneous nucleation theory, which, as mentioned, would have a typical 10-90 width for a defined nucleator of some 0.7°C. We are not aware of such a tube dependence analysis being done since the seminal work of Dorsey in 1948 [3].

It is significant that, in seven runs, on six nmr tubes, only once did the order of freezing change, indicating very clearly the tube dependence and the well-defined existence of a nucleation site inside each tube, be that the tube wall imperfections, or perhaps a piece of dirt.

In July 2024, in a third set of experiments, carried out in Sydney, Australia and in Utqiaġvik, Alaska, we examined the supercooling ability of a 5 mm diameter glass nmr tube with 200 µl of doubly-distilled, deionised water sealed within. It was repeatedly cooled, supercooled, frozen and warmed. The cooling was in a dedicated aluminium block sandwiched between two 65 W Peltier thermoelectric cooling units and cooling was set

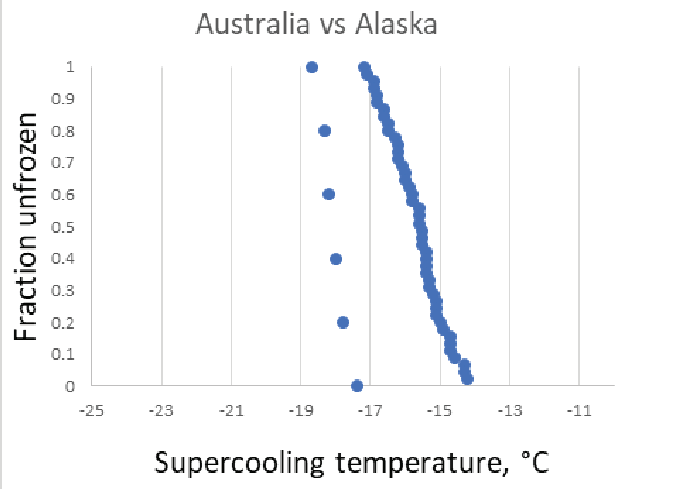

Figure 3. *Data from Alaska (rhs data set), Sydney (six points).*

electronically at 3°C per minute. Freezing was determined optically by the lack of transmission of a light beam through the tube and temperature recording was by a thermocouple placed outside, but adjacent to, the tube. This experimental set-up is well described in the various works of Haymet and Wilson [1,2]. The same equipment and same tube was used in both sets of measurements. The results are presented in Figure 3.

The six blue dots colder than -17°C are the Sydney freezing data and the slope of those data is very typical, albeit fairly narrow 10-90 for such few data points. This does suggest a well-defined nucleation site not changing with time, or with each warming/cooling cycle. As mentioned above, typically a larger number of repeats provides a tighter survival curve and so the higher number in Figure 3 showing more spread is interesting and worthy of comment and investigation. We argue that the larger 10-90 width of the Utqiaġvik data suggests one of two things. The sample is changing, or at least the nucleation site is changing, with time, over the three days it took to gather the data. Or, there are multiple nucleation sites, i.e. not the same for each event, or at least not the same efficiency for each event. This second option would be the case if it were, say, traversing neutrons causing the events.

To explain further; in 2021, it was shown in a laboratory in Albany, NY, USA, that over many repeats, a beam of neutrons from a Cf source could nucleate a small volume of deeply supercooled water [4]. Figure 4. reproduces these published results of probability of freezing of supercooled water, with and without a neutron beam being turned on. The so-called T50 has been shifted to warmer temperatures with the neutrons turned on, and with a 5.2 σ difference, but perhaps more importantly, the slopes are different for the same volume of water, in the same container.

SSince the 1980s it has been known that cosmic rays, mainly reported as atmospheric neutrons, can penetrate the natural shielding of buildings and equipment and produce errors in integrated circuits. The high-energy neutron fluxes range between 10 particles $cm^{-1}\cdot h^{-1}$ at sea level and some 103 particles $cm^{-1}\cdot h^{-1}$ at 10 km altitude [5]. Atmospheric neutrons are end-products of the interactions of cosmic rays with our atmosphere. The cosmic rays interact with air nuclei to generate particles comprising protons, neutrons, etc. The radiation intensity is maximum at 20 km and then drops off to sea level at which

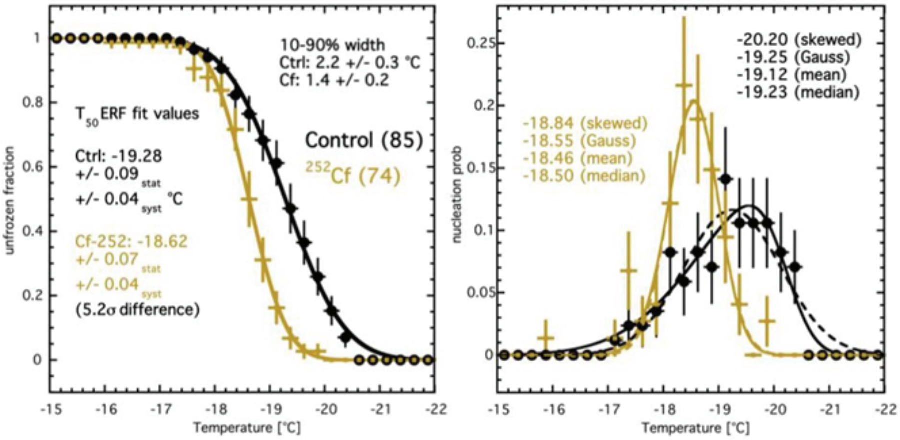

Figure 3. *Figure reproduced from ref. 5 of survival curves for a sample of water exposed to, and not exposed to, a beam of neutrons.*

the neutron flux remains significant. Particle physicists have been utilising bubble chamber for many years, relying on the phase transition of a metastable substance. In recent years, the search for dark matter has led researchers to re-examine the idea of metastable fluids, specifically to determine if a deeply supercooled volume of water can be nucleated by the action of a subatomic particle. Szydagis and colleagues [4,6] have been using supercooled water in a device nicknamed Snowball Chamber and, as above, indeed shown that a beam of neutrons can nucleate a volume of deeply supercooled water. The dark matter community use neutrons as an analogue for dark matter, having no better proxy at this time.

Separately, it is known that the numbers of cosmogenic neutrons at high latitudes are many times that at the equator [7,8] These are muon-induced and reach as low as 5 km altitude, if not lower. Szydagis and colleagues have recently looked at the energy requirements for nucleating water at deep supercooling [6]. Although speculative, their arguments suggest the order of 1keV and this barrier would also put energy for various neutron sources within reach, even cosmogenic neutrons.

Discussion

It seems apparent that when a single nucleation site of present (it is very hard to achieve homogeneous nucleation where there is no site at all) then the survival curve is confined to a 10-90 of about 0.7°C. However, if the so-called 'best' nucleation site changes over time or, if indeed at any given time, the best site is a passing sub-atomic particle we would expect the 10-90 to be significantly wider. What is interesting is the likelihood of a tight 10-90 in the case of only a few measurements on the same volume of water, clearly a function of probability as much as anything. Further, what do we expect the 10-90 to do in the case of additives such as solutes? If we add an insoluble crystal to aid nucleation the width remains the same [9], if we coat the tube walls with a hydrophobic layer, the width also does not change [10]. Overall, if a survival curve is found from a repeated series of freeze-thaw events and if the 10-90 is greater than around 0.7°C, the user may consider if the nucleation site is changing over the length of time of the experiments.

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