Supplement of

A pyroelectric thermal sensor for automated ice nucleation detection

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1 BET Measurements

Table S1: Individual measurements and mean values of sample surface area.

| Glassy K-feldspar (m$^2$g$^{-1}$) | Crystalline K-feldspar (m$^2$g$^{-1}$) |
|-----------------------------------|----------------------------------------|
| 1.430                             | 4.062                                  |
| 1.710                             | 5.195                                  |
| 2.204                             | 5.769                                  |
| 1.781                             | 5.009                                  |

2 Fitting Liquid Proportion Curves

The liquid proportion curves were fitted using the assumption that the probability of a droplet remaining liquid at time t is described by a non-homogeneous Poisson process,

$$P_{liq}(t) = \exp\left[-\int_{t_0}^{t} J(T(t')) dt'\right],$$  \hspace{1cm} (1)

where $t_0$ is the time at which the temperature drops below the melting point of ice. To convert $P_{liq}$ to a function of $T$ substitute $dT = -\alpha dt$, since linear cooling ramps were used in this experiment.

$$P_{liq}(T) = \exp\left[-\int_{T'}^{T_m} \frac{J(T')}{\alpha} dT'\right],$$  \hspace{1cm} (2)

where $T_m$ is the melting temperature of ice. At this point it is necessary to assume a functional form for $J(T)$. Here we assume for simplicity that $\log(J)$ varies linearly with $T$. Typically in ice nucleation experiments the majority of droplets freeze in a narrow temperature region. Defining the centre of this region as $T_0$ and linearly expanding $\log(J)$ around it we obtain

$$J(T) = A \exp[-\omega(T - T_0)],$$  \hspace{1cm} (3)

where $A$ is $J(T_0)$ and $\omega = -\frac{d \log(J(T))}{dT}|_{T_0}$. This choice is justified by the quality of the resultant fit in the region of interest.

Substituting equation 3 into equation 2 and integrating we obtain

$$P_{liq}(T) = \exp\left[\frac{-A}{\alpha \omega} [e^{-\omega(T - T_0)} - e^{-\omega(T_m - T_0)}]\right].$$  \hspace{1cm} (4)

This function can be fitted to the liquid proportion data using standard techniques, with $T_0$ fixed at the temperature at which half of the droplets are frozen. Here Scipy’s optimize.curve_fit function was used, the optimized parameters are in Table S2.

3 Error Calculations

Stochastic errors were estimated using the Wilson score confidence interval on each temperature bin with more than one nucleation event. An example is shown in Figure S1. The errors in the liquid
Table S2: Fitting parameters for Figure 5A using equation 4.

| Sample                  | $T_0$ (K) | $A$  | $\omega$ |
|-------------------------|-----------|------|----------|
| Crystalline K-Feldspar 0.1 wt% | 259.6     | 0.009 | 0.915    |
| Crystalline K-Feldspar 0.05 wt% | 257.6     | 0.014 | 1.020    |
| Crystalline K-Feldspar 0.025 wt% | 256.9     | 0.014 | 1.046    |
| Crystalline K-Feldspar 0.0125 wt% | 256.9     | 0.006 | 0.664    |
| Glassy K-Feldspar 1.0 wt%     | 254.3     | 0.005 | 0.391    |
| Glassy K-Feldspar 0.1 wt%     | 250.8     | 0.008 | 0.607    |
| Background               | 245.6     | 0.005 | 0.433    |

Proportion were calculated based on how the minimum and maximum number of freezing events would effect the liquid proportion at that temperature bin, assuming the mean number of events were seen in all higher temperature bins. These errors were then combined with the errors in INP area in the calculation of $n_s$ and $J$ using standard propagation techniques.

Figure S1: Data from 0.1%wt crystalline K-feldspar binned in 0.5°C wide bins. The error bars are from the Wilson score at a 95% confidence interval.
4 Background Removal

Each liquid proportion curve $P(T)$ is the probability that a droplet is still liquid at temperature $T$. The measured curve, $P_{\text{meas}}(T)$ is the product of the probability that there has been no ice nucleation event caused by the ice nucleating agent of interest $P_{\text{INP}}(T)$, and the probability that there has been no background event $P_{\text{back}}(T)$. Hence the background can be removed by dividing $P_{\text{meas}}(T)$ by $P_{\text{back}}(T)$. The influence of the background can be seen in Figure S2, where only the 0.1wt% glassy sample shows a noticeable change, although it is a maximum of 0.2°C.

Figure S2: The background adjusted liquid proportion curve fits for pure water, crystalline K-feldspar and glassy K-feldspar. The colours are the same as Figure 5 in the manuscript.