Supplementary Information for Antisolvent Sonocrystallisation of Sodium Chloride and the Evaluation of the Ultrasound Energy Using Modified Classical Nucleation Theory

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The turbidity of the solution was measured as a function of time (data collected at 1 second intervals) using a fibre-optic turbidity probe (Crystal Eyes, H.E.L. Limited) and the induction time determined by the time lapse between the addition of the NaCl-water solution and the onset of turbidity (Fig.S1a). The turbidity can be affected by the initial degassing of ethanol with the addition of concentrated salt solution and cavitation bubbles from ultrasound. Therefore, the onset of turbidity was determined by taking the intercept of the slope of the initial linear rise in the turbidity and the initial blank turbidity reading of ethanol solution prior to the addition of salt solution (Fig.S1b). A data table for all the induction times measured at different supersaturation ratios and experimental conditions are tabulated in Table S1-8.

To validate the assumption that γ is constant, the data in **Error! Reference source not found.** was fitted with Eq. 6 and keeping γ as a variable along with A and Δ Wu. The data is presented in Table S9 and show γ increased to 49.7 mN m⁻² at 2 W sonication followed by a decrease to 15.1 mN m⁻² at 5 W then an increase to 23.2 mN m⁻² at 15 W. Similar fluctuations was also observed for the pre-exponent A. For Δ Wu, a decrease in the absolute value with increasing sonication power was obtained which would result in a decrease in the induction time. However, experimental observation shows a decreasing induction time with increasing sonication power. With these inconsistencies, it was assumed that γ is constant and the effect from sonication is largely relating to its impact on homogeneous nucleation.



Figure S1:An example of turbidity as a function of time data obtained and how induction time was determined. (a) a typical data and (b) interference from degassed bubbles when superaturation is first established.

Table S1:Induction time for different supersaturation ratios in the absence of ultrasound. A stirring speed of 100 rpm was applied.

| S | Induction time [s] | Standard deviation [s] | |
|-----|--------------------|------------------------|--|
| 1.7 | 148.9 | 1.38 | |
| 1.9 | 80.1 | 3.67 | |
| 2.1 | 77.3 | 1.87 | |
| 2.3 | 66.3 | 1.54 | |
| 2.4 | 50.9 | 1.38 | |
| 2.6 | 45.9 | 1.17 | |
| 2.8 | 31.3 | 1.72 | |
| 3.5 | 15.5 | 1.58 | |
| 3.8 | 9.3 | 2.11 | |
| 4.2 | 6.5 | 1.43 | |
| 4.5 | 3.7 | 1.29 | |
| 4.8 | 3.4 | 1.69 | |
| 5.2 | 2.7 | 1.26 | |

Table S2:Induction time for different supersaturation ratios in the presence of 98 kHz and 2 W sonication. A stirring speed of 100 rpm was applied.

| S | Induction time [s] Standard deviation [s] | |
|------|---|------|
| 1.73 | 8.81 | 1.10 |
| 2.07 | 7.18 | 1.08 |
| 2.77 | 3.21 | 1.35 |
| 5.19 | 1.85 | 1.29 |

| S | Induction time [s] | Standard deviation [s] | | |
|------|--------------------|------------------------|--|--|
| 1.73 | 7.73 | 1.08 | | |
| 2.07 | 3.58 | 1.18 | | |
| 2.77 | 2.62 | 1.15 | | |
| 5.19 | 1.66 | 1.08 | | |
| 1.91 | 3.89 | 1.00 | | |
| 2.42 | 2.68 | 1.12 | | |
| 4.16 | 1.71 | 1.26 | | |

Table S3:Induction time for different supersaturation ratios in the presence of 98 kHz and 5 W sonication. A stirring speed of 100 rpm was applied.

Table S4:Induction time for different supersaturation ratios in the presence of 98 kHz and 15 W sonication. A stirring speed of 100 rpm was applied.

| S | Induction time [s] | Standard deviation [s] |
|------|--------------------|------------------------|
| 1.73 | 3.38 | 1.11 |
| 2.07 | 2.50 | 1.15 |
| 2.77 | 1.74 | 1.27 |
| 5.19 | 1.32 | 1.21 |

Table S5:Induction time for different supersaturation ratios in the presence of 22 kHz and 2 W sonication. A stirring speed of 100 rpm was applied.

| S | Induction time [s] | Standard deviation [s] |
|------|--------------------|------------------------|
| 1.91 | 6.64 | 1.12 |
| 2.42 | 4.41 | 1.09 |
| 3.46 | 2.71 | 1.15 |
| 5.20 | 1.74 | 1.50 |

Table S6:Induction time for different supersaturation ratios in the presence of 44 kHz and 2 W sonication. A stirring speed of 100 rpm was applied.

| S | Induction time [s] Standard deviation [s] | | |
|------|---|------|--|
| 1.91 | 4.91 | 1.08 | |
| 2.42 | 2.78 | 1.06 | |
| 3.46 | 2.63 | 1.14 | |
| 5.20 | 1.89 | 1.56 | |

Table S7:Induction time for different supersaturation ratios in the presence of 139 kHz and 2 W sonication. A stirring speed of 100 rpm was applied.

| S | Induction time [s] | Standard deviation [s] |
|------|--------------------|------------------------|
| 1.91 | 4.24 | 1.07 |
| 2.42 | 3.34 | 1.07 |
| 3.46 | 2.09 | 1.11 |
| 5.20 | 1.89 | 1.56 |

| S | Induction time [s] Standard deviation [s] | | |
|------|---|------|--|
| 1.91 | 51.24 | 2.34 | |
| 2.42 | 37.76 | 2.10 | |
| 3.46 | 15.13 | 2.16 | |
| 5.20 | 3.26 | 2.73 | |

Table S8:Induction time for different supersaturation ratios in the presence of 500 kHz and 2 W sonication. A stirring speed of 100 rpm was applied.

Table S9: Parameters γ , ΔW_U and A for different ultrasound powers, determined by fitting Eq. 6 to the data in Figure 6. It was assumed that the supersaturation ratio is unaffected by ultrasound. The pre-exponent A and γ obtained using Eq. 3 for absence of ultrasound, are also shown for comparison.

| Amplifier Power [W] | Surface tension [mN m ⁻²] | $\Delta W_{\rm U} [J \text{ m}^{-3}]$ | Α |
|---------------------|---------------------------------------|---------------------------------------|------|
| 0 | 31.0 | 0 | 1.22 |
| 2 | 49.7 | -3.1×10^{8} | 2.22 |
| 5 | 15.1 | -1.46×10^{8} | 0.69 |
| 15 | 23.2 | -1.02×10^{8} | 1.06 |