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Short Communication

# Up to which temperature ultrasound can heat the particle?

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ABSTRACT

Crystallographic property such as crystallite size has been used for evaluation of the temperature up to which high intensity ultrasound can heat metal particles depending on physical properties of sonication medium and particle concentration. We used >100 μm metal particles as an in situ indicator for ultrasonically induced temperature in the particle interior. Based on powder X-ray diffraction monitoring of $Al_3Ni_2$ crystallite sizes after ultrasound treatment the average minimum temperature $\left(\bar{T}^{min}_{particle}\right)$ of sonicated particles in various sonication media was estimated. Additionally, it was found that crystallite size in ultrasonically treated metal particle depends on the frequency of interparticle collision. Through the adjustment of particle concentration, it is possible to either accelerate the atomic diffusion or force the melting and recrystallization processes. Overall, the energy released from collapsing cavitation bubble can be controllably transferred to the sonication matter through the appropriate choice of sonication medium and the adjustment of particle concentration.



## 1. Introduction

The cavitation phenomenon has been intensively studied for several decades unraveling the details on the mechanism of cavitation bubble formation, oscillating growth, and consequent collapse [1,2]. Great number of research work has been dedicated to investigation of cavitation bubble physical properties such as internal temperature and pressure [3]. Experimentally measured values of temperature inside cavitation bubble were reported in the range of ~5000 K depending on vapor pressure of the liquid that is being ultrasonicated [4,5].

The acoustically generated "hot spots" – localized areas of extremely high temperatures and pressures [6] can release significant amount of energy to initiate various bottom-up synthesis of colloids at highly non-equilibrium conditions [7,8]. For example amorphous iron, carbon and nanoalloys were sonochemically prepared from organometallic compounds that were decomposed inside the collapsing bubble [9–11].

Besides sonochemical processes that use collapsing bubble as a microreactor [12] for high-temperature synthesis, sonomechanical processes can also trigger highly non-equilibrium nanostructuring [13–17]. In suspensions of particles bubble collapse induces shock waves, intensive turbulent flow, and interparticle collisions. It was shown that in concentrated (20 wt.%) suspensions of ~10 μm particles the surfaces of sonicated particles can locally melt in the temperature range of 2600–3400 °C and form fused agglomerates of particles [18]. However, which temperature is inside ultrasonically treated particles is still unknown. Weather acoustic cavitation can affect microstructure of both the surface and the interior of μm-sized particles and can be used for top-down synthesis of nanomaterials is an interesting question.

Energy release/transfer from collapsing cavitation bubble depends on sonication medium [1,2]. Varying sonication medium it is possible to adjust ultrasonic conditions for a particular synthesis. For instance, the thermal impact of continuously imploding cavitation bubbles on the surrounding liquid or sonicated matter itself depends on the physical properties of sonication medium such as vapor pressure or viscosity [19]. The main obstacle in precise evaluation of energy transfer from collapsed bubble is the absence of reliable characterization methods or a material which would serve as an indicator for such transition.

Recently, we showed that high intensity ultrasound (HIUS) treatment of AlNi (50 wt.% Ni) alloy particles suspension in ethanol led to noticeable changes in crystallographic properties [20]. Specifically, we observed present in AlNi alloy $Al_3Ni_2$ intermetallic crystallite growth and microstrain reduction due to thermally accelerated atomic diffusion. As oppose to metallurgical industry where temperature forced grain growth is leading to undesirable metal materials softening [21], in catalysis, for example, growth of crystallites may be quiet beneficial [22].

Since it is undoubtedly clear that grain boundary reduction is forced thermally, induced by temperature gradient crystallite growth to a particular value requires a defined amount of energy,

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thus, the crystallite size may serve as an indicator of the energy transfer efficiency from collapsing cavitation bubble with respect to sonication medium. In other words, we suggest that through powder X-ray diffraction (PXRD) based monitoring of the crystallite size in metal particles it is possible to estimate the average minimum temperature $\left(\bar{T}^{min}_{particle}\right)$ to which metal particles can be heated in the cavitated medium. These findings might be of a great interest in the areas of sonochemical synthesis, ultrasonic food processing, or ultrasonically induced nanostructuring for various applications.

In present study we report on the temperature effect of acoustic cavitation during HIUS treatment of $Al_3Ni_2$ phase in AlNi (50 wt.% Ni) alloy particles depending on physical properties of sonication medium. Based on PXRD monitoring of $Al_3Ni_2$ intermetallic crystallite sizes after ultrasound treatment we made an attempt to estimate the average minimum temperature $\left(\bar{T}^{min}_{particle}\right)$ of sonicated AlNi alloy particles in various sonication media. $Al_3Ni_2$ intermetallic phase identification was performed by Rietveld refinement of PXRD patterns. Crystallographic parameter such as crystallite size was evaluated using uniform deformation model (UDM) based Williamson–Hall (WH) method [23–25]. Keeping in mind that pressure can influence the crystal structure of solids; we plan to address the aspects of ultrasonically induced pressure effects on metal particle in our ongoing studies.

## 2. Experimental section

Aluminum–Nickel (50 wt.% Ni) alloy powder with average particle size of 140 μm was purchased from Fluka. Anhydrous ethanol, ethylene glycol, and decane were purchased from Sigma Aldrich. All chemicals were the highest purity grade available and were used as received without further purification. The water was purified before use in a three stage Millipore Milli-Q Plus 185 purification system.

0.1 g $ml^{-1}$ Al/Ni alloy powder was sonicated in ethanol, ethylene glycol, decane, or ethanol/water and ethylene glycol/water mixtures for 60 min with a Hielscher UIP1000hd, (Hielscher Ultrasonics GmbH, Germany) operated at 20 kHz with a maximum output power of 1000 W. The apparatus was equipped with an ultrasonic horn BS2d22 (head area of 3.8 $cm^2$) and a booster B2-1.8. The maximum intensity was calculated to be 140 W $cm^{-2}$ at mechanical amplitude of 106 μm. To avoid overheating during sonication the experiment was carried in a homemade thermostatic cell connected to a thermostat (Huber GmbH, Germany). The temperature was monitored during the treatment and kept at 333 K after HIUS treatment metal particles were separated from supernatant by centrifugation at a speed of 10,000 rpm for 1 h and washed with absolute ethanol followed by drying under vacuum at room temperature. To investigate the effect of particle concentration, various amounts (0.025–0.125 g $ml^{-1}$) of AlNi particles were sonicated in decane for 60 min. For evaluation of ultrasonic irradiation time effect, 4 g of AlNi particle suspensions in decane were ultrasonically treated during 5, 10, 15, 20, 25, 30, 40, 60, and 90 min. As a reference experiment, to ensure the effect of acoustic cavitation on crystallographic properties, AlNi particle suspension was subject to 60 min of intensive stirring (13,000 rpm). No change in crystallographic parameters was observed.

For the annealing procedure 4 g of AlNi (50 wt% Ni) alloy powder was placed in the preheated to required temperature muffle oven (Heraeus, Germany) for 2 h and kept there for 60 min. The procedure was carried for six AlNi alloy powder samples at the following temperatures: 500, 550, 600, 650, 700 and 1000 °C.

Powder X-ray diffraction (PXRD) analysis of the samples was performed using Stoe STADI P X-ray transmission diffractometer ($CuK_\alpha$ radiation from the copper target using an in built nickel filter, $\lambda$ = 1.54056 Å).

Crystallographic parameter such as crystallite size was calculated using Scherrer and Williamson–Hall methods [20]. Present in AlNi alloy particles $Al_3Ni$ phase was not used for the calculations since it can be easily oxidized in aqueous sonication medium [22].

Scherrer method relies on utilizing the following equation:

$$D = \frac{k\lambda}{\beta_D \cos\theta} \tag{1}$$

where $k$ is shape factor (a constant equals to 0.94), $\lambda$ is the X-ray wavelength (1.54056 Å for $CuK_\alpha$ radiation), $\beta_D$ is the instrumental corrected peak width at half-maximum intensity, $\theta$ is the peak position, and $D$ is the effective crystallite size normalized to the reflecting planes. It is important to note that to avoid any misleading; only not overlapping peaks were chosen for data processing.

According to WH method strain-induced broadening arising from crystal imperfections and distortion are related by:

$$\varepsilon \approx \frac{\beta_s}{\tan_\theta} \tag{2}$$

Assuming that the size and strain contributions to the line broadening are independent of each other, the observed line breadth can be written as the sum of the two terms:

$$\beta_{hkl} = \beta_s + \beta_D \tag{3}$$

Substitution of Eqs. (1) and (2) into Eq. (3) results in the following:

$$\beta_{hkl} = \left(\frac{k\lambda}{D\cos\theta}\right) + (4\varepsilon\tan\theta) \tag{4}$$

After rearranging, Eq. (4) becomes:

$$\beta_{hkl}\cos\theta = \frac{k\lambda}{D} + (4\varepsilon\sin\theta) \tag{5}$$

The WH equation represents uniform deformation model (UDM) where the strain is assumed to be uniform in all crystallographic directions, thus considering the isotropic nature of the crystals, where all the material properties are independent of the direction along which they are measured. Plotting values of $\beta_{hkl}\cos\theta$ as a function of $4\varepsilon\sin\theta$ allows estimating microstrain $\varepsilon$ (slope of the fitted line) and the average crystallite size $D$ ($y$-intercept of the fitted line).

## 3. Results and discussion

Fig. 1 schematically represents $Al_3Ni_2$ crystallites growth in AlNi (diameter is approx. 140 μm, 50 wt.% Ni) alloy particles that is induced by temperature gradient. In 10 wt.% suspensions of metal particles cavitation induced shock waves and interparticle collisions can create the temperature gradient [26] that propagates in a particle and forces crystal growth. Since it is known that intensity of cavitation depends on sonication medium, different crystallite size can be expected in different sonication media. Thus, change in crystallographic properties might serve as an indicator of energy transfer between collapsing cavitation bubbles and sonicated matter. To provide the information on the heat transfer from collapsing cavitation bubbles to the sonicated matter we used sonication media with various physical properties – vapor pressure and viscosity. Namely, we treated AlNi alloy particles with HIUS in water/ethanol and water/ethylene glycol mixtures and in decane.

In order to calibrate thermal effect of acoustic cavitation on crystallographic property such as crystallite size we annealed the