

## Real foundations and background of Sonochemistry and Ultrasonic Metallurgy

Let us start with generally applicable comments, such as:

1. All (acoustically linear) liquids, single and multi-phase liquid solutions, simple and composite liquid mixtures on low and moderate temperatures, including liquid metals on temperatures (being on not very close to solidification temperature) behave on a similar way regarding acoustic, mechanical, and ultrasonic vibrations, cavitation, degassing and micro-crystallization. We also apply the same mathematical models to describe such vibrations, oscillations, and waves in different liquids. Exceptions are liquids behaving as acoustically or mechanically non-linear, and/or anisotropic, such as “biologically-charged liquids”, sea water with salts, minerals, and biological-ingredients, high viscosity, and high-density liquids... Clean water and most of liquid metal alloys (within a temperature range sufficiently higher than relevant solidification temperatures) dominantly behave as acoustically or mechanically linear materials (until certain maximal temperature). If we consider M. Planck Black body electromagnetic radiation as an “**external radiation**” (radiating from a black-body cavity towards surrounding, external, and empty space), then we could understand and conceptualize effects within heated liquids, as being influenced by an “**internal black-body radiation**”. *Here are grounds for opening a research domain related to extended thermodynamics applicable on all fluids.*
2. Ultrasonic and acoustic vibrations in liquids are very much like thermal or thermodynamic activity of atoms and molecules in fluids. Both are creating mechanical oscillations and associated heat related effects in liquids. The difference is that an ordinary (non-ultrasonic) heating source is producing spatially chaotic, randomized, non-isotropic and non-laminar heating-related motions and radiations (of atoms, molecules, and other small particles), and other specific-technology designed ultrasonic sources, transducers and sonotrodes are producing structurally and spatially organized flow of vibrating and radiating energy. Both, thermal and ultrasonic waves, and radiations can be measured using almost the same “heat flow sensors” (for instance based on Peltier effect). This is the reason why ordinary heating (or thermal activity and radiation) is strongly interfering with ultrasonic waves.
3. Liquid aluminum, from the point of view of ultrasonic vibrations (when we agitate it with certain sonotrode or ultrasonic radiator), behaves like water on a room temperature. This is the reason why we often use water as the liquid-aluminum acoustic-load simulator, or as testing media (it has similar acoustic impedance like water, similar viscosity like water on a room temperature, and its density is also in the same order of magnitude as water, meaning 2.7 times higher...). When we realize a well operating ultrasonic regime in water (using ultrasonic radiators or emitters, or systems designed for ultrasonic liquid aluminum processing), we will have very similar operating regime like ultrasonically treating a liquid aluminum (with the same tools/emitters/radiators/sonotrodes). Of course, this is valid only if a liquid aluminum is on temperatures sufficiently higher than solidification temperature (for instance, higher for 20, or 50 or 100°C). If liquid aluminum is cooled and approaching closer to its solidification

temperature, its density, viscosity, and other fluids-related properties, including acoustic impedance, will significantly change, and we will not have very good acoustic coupling with implemented sonotrode/radiator/emitter, as it was before in a very fluidic, water-similar phase. In fact, by cooling liquids, acoustic waves attenuation will increase, cavitation will decrease, and applied vibrating tools will gradually stop oscillating. This is related to the fact that ultrasonically produced degassing is much stronger when liquid aluminum is on certain high temperature, and when melt temperature is decreasing, we create conditions for increasing acoustic (or ultrasonic) micro-crystallization (of course this about increased crystallization works better around added impurities (such as master alloys), when combined with vibrations). Acoustically induced micro and nano crystallization is also (analogically) applicable for all other liquid mixtures (and water solutions) on low and moderate temperatures.

4. Now, let us address mutual dependence of liquid temperatures and acoustic or ultrasonic agitation. For instance, we start sonication when certain liquid is on certain temperature that is close to solidification. For water on temperature close to  $+2^{\circ}\text{C}$ , when externally introduced sonication starts (but water is still fluidic), we produce immediate and total micro-crystallization and freezing of water. This is very similar to liquid metals (only relevant and comparable temperatures are shifted up). Now, imagine that we start heating and sonicating water (kinetic energy and randomly distributed velocity of involved molecules is increasing). Mobility of water molecules is proportionally increasing with increased temperature, and overall acoustic activity, cavitation and degassing are also increasing until approx.  $57^{\circ}\text{C}$ , following certain parabolic or bell-curve, also associating on a Black-Body radiation curve). Maximum of acoustic activity and cavitation in water is known to be on  $57^{\circ}$ . Increasing water temperature higher than  $57^{\circ}\text{C}$  will start reducing ultrasonic activity and cavitation in water... For other liquids we will also have similar effects, "but maximal acoustic activity temperatures" will be differently shifted (again following parabolic or bell-shaped curves). That means, if liquid aluminum temperature is still within the rising zone of its parabolic curve, acoustic activity and cavitation will also increase until certain maximum, and we will produce very good degassing and cavitation. If we continue increasing liquid temperature (passing parabola maximum point), cavitation, degassing, and ultrasonic activity will start decreasing. All of that is analogically valid for all linearly behaving liquids. Organic liquids and sea salt water are behaving as non-linear liquids, but most of liquid metal alloys and composites are behaving like acoustically linear liquids (in described temperature interval).
5. Now, to say something about sonicating and oscillating amplitudes of ultrasonic energy sources. Ultrasonic or acoustic energy is directly proportional to the contact surface between certain liquid and applied or submersed ultrasonic source/tool/sonotrode/radiator/emitter (here, all mentioned terms are synonyms). That means, we need to realize maximal (as possible) contact surface to produce and transfer maximal ultrasonic energy (analogical to electromagnetic waves emitters, antennas, receivers...). If we increase amplitudes of applied ultrasonic waves, we will also increase acoustic energy emission until certain maximal amplitude of ultrasonic source. If we continue increasing amplitude of oscillations, we will start braking molecular, cohesion,

adhesion and Van der Waals forces between ultrasonic emitter and a surrounding liquid state. High amplitudes agitation of sonicated liquids will gradually produce non-linear acoustic effects (coming closer to plastic deformation effects). This is creating acoustic decoupling (meaning that ultrasonic energy transfer will stop). Submersible ultrasonic transducer is this what we consider as ultrasonic waves emitter/radiator/sonotrode..., which will be immersed or submersed in certain liquid (this way we vibrate liquid from inside). For instance, in ultrasonic cleaning and Sonochemistry applications we often apply ultrasonic agitation from outside.

As I know, nobody integrally formulated such simple, analogical, generally valid (for all liquids) conclusions in one place, related to ultrasonic liquid processing, or to ultrasonic energy transfer, degassing, cavitation, influence of operating temperatures, acoustic coupling, and decoupling between ultrasonic sonotrodes and treated liquids, linear and non-linear behaviors of liquids during ultrasonic processing... This what is summarized here is essential for understanding Sonochemistry and Ultrasonically assisted Metallurgy...

Without considerations and facts as just mentioned we will not have a basic conceptual understanding about what is happening during ultrasonic processing of liquids. Of course, all that what exposed here is separately and sporadically known until certain level, and on some way stated or published, but nobody made such generally valid and mutually related comments and conclusions. We need to address such problematic much better and systematically ...

This what is briefly elaborated here is our competitive advantage regarding understanding Ultrasonic Metallurgy. Maybe it is still not recommendable to explain to others what we are doing (and what the relevant background behind all of that is), before we create something much more significant ...

*PS. Read more about matter waves, oscillations, vibrations... here:*

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