



## Editorial

## Special Issue on “Ultrasound hybridized technologies: A new breathing for sonochemistry”



## 1. Overview

The term “hybrid” derives from Latin *hibrida* meaning offspring of a mixed union. According to the Encyclopaedia Britannica (<https://britannica.com/science/hybrid>), that offspring differs in genetically determined traits, such as species, genera, or rarely families. The process of hybridation increases the genetic variety within a species, which is required for evolution to occur. Although hybrids (animals in particular) may be sterile because of biological incompatibility, the concept of hybridation is invariably timely to illustrate the advantages of mixed approaches or technologies resulting in fruitful and synergistic, rather than sterile, achievements. In this context, the Special Issue on “Ultrasound hybridized technologies” was launched to show the cooperation of ultrasound with different strategies and activation methods, which assure higher performance than sonication alone. Such a hybridation/merging often closes the gap in an unsolved or poor solution.

Inevitably, submissions to this Special issue have been seriously hampered by the worldwide pandemic caused by SARS-Cov2, which interrupted abruptly both academic and research activities. Even so, 13 papers have been accepted to be published in this Issue, largely focused on environmental concerns in line with the ever-increasing attention to sustainability, waste reduction, as well as more benign and efficient processes.

Thus, Yentür and Dükkancı describe the sono-photocatalytic oxidation of carbamazepine, a widely sold anticonvulsant and analgesic drug, present in wastewater and aqueous effluents [1]. The process works well under visible LED irradiation combined with low (20 kHz) and high (850 kHz) frequencies, and is markedly influenced by other experimental conditions, including pH range, catalyst loading, acoustic power, and sonication mode; the latter being more efficient in continuous, rather than pulse, operation for degradation efficiency.

Perhaps, one of the most popular hyphenated technologies involves the simultaneous (or separate) application of ultrasound and microwave irradiation. A new reactor design, suitable for process intensification, is reported by Călinescu and coworkers [2]. The set-up enables the continuous dissipation of thermal energy, thereby making possible the use of high powers. The efficiency is illustrated through the degradation of *p*-nitrophenol along with biodiesel production by a transesterification reaction.

Ziylan-Yavas et al. describe in detail the decomposition and mineralization of caffeine, a substance commonly found in wastewater [3]. The authors have employed two well-known oxidative processes affording OH radicals, such as sonolysis (at 577 kHz) and UV-photolysis with H<sub>2</sub>O<sub>2</sub>. Both led to complete caffeine elimination, but they were insufficient for complete mineralization. A hybrid process consisting of

UV-H<sub>2</sub>O<sub>2</sub> and TiO<sub>2</sub> as catalyst was the most efficient at minimum doses of the reagents. Sonication largely enhanced the rate of caffeine decomposition, but paradoxically it reduced the overall degradation as well. From a mechanistic viewpoint, this result was interpreted in terms of additional H<sub>2</sub>O<sub>2</sub> formation and cavity bubbles that impeded the efficient transmission of UV light.

In what is probably the first study addressing the combination of high-intensity ultrasound and supercritical CO<sub>2</sub> for fungal and bacterial inactivation, Gomez-Gomez and associates report the inactivation kinetics at different pressures and temperatures of *Aspergillus niger* and *Clostridium butyricum* spores in oil-in-water emulsions [4]. Results are preliminary, yet promising, and need further exploration. However, the team has demonstrated that ultrasound intensified the sc-CO<sub>2</sub> inactivation of *C. butyricum* by shortening the inactivation time, from 10 to 3 min at 85 °C, while sonication had little or no effect on the sc-CO<sub>2</sub> inactivation of fungi (*A. niger* spores).

Choi et al. report an efficient ultrasonic desorption of heavy metal (Cu, Pb, and Zn) using HCl and EDTA in real contaminated soils [5]. Compared with conventional soil washing through mechanical mixing, the combined US/mixing process was more efficient for the removal of such polluting species, especially in the less extreme washing conditions, i.e. at lower HCl or EDTA concentrations and lower liquid:soil particle ratio.

The use of metallic nanoparticles (NPs) activated by ultrasonic fields currently represents a major portion of so-called sonocatalytic applications, as witnessed by literature data. In this context, magnesium diti-tanate (MgTi<sub>2</sub>O<sub>5</sub>) NPs were prepared by Selvamani *et al.* using a simple hydrothermal annealing method and thoroughly characterized by structural elucidation methods [6]. Their catalytic properties (sonocatalytic, photocatalytic, and sono-photocatalytic) were assessed using a few common triphenylmethane dyes, like crystal violet and fuchsin. Notably, the sono-photocatalytic activity of the aforementioned NPs were found to be much higher than the photo- and sonochemical processes taken individually.

The hybrid combination of cavitation (20 kHz, 100 W) with oxidative processes (Fenton’s chemistry and ozone) has also been applied to the treatment of dye-containing industrial effluents, as reported by Gujar and Gogate [7]. By merging sonication with Fenton oxidation (H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> reagent) followed by lime treatment, maximum COD reductions could be achieved leading to colorless and reusable effluents. Addition of ozone did not result in significant improvements, although it promoted a faster treatment.

Alfonso-Muniozguren et al. provide a comprehensive review on pharmaceutical removal from urban and industrial wastewater, a major concern and threat to human and animal health [8]. Numerous drugs

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end up in aquatic effluents, where they undergo further biotransformations, thereby inducing toxicity and antibiotic resistance. Such mechanisms are documented by the authors, who highlight the importance of combining ultrasonic activation with membrane filtration and membrane bioreactors, two usual treatment methods for wastewaters. Considerations of enhanced efficiency under sonication and scale-up capabilities are discussed by the authors as well.

The extraction of bioactive substances from natural matrices aided by ultrasound has become a well-established domain of modern sonochemistry, which competes favorably with other technology-assisted extractions. Baião Dias and coworkers have reviewed the subject of ultrasound-promoted extraction of a variety of natural products using pressurized liquids, both sub- and supercritical fluids [9]. The combination of these techniques lie in the realm of green chemistry and opens up new applications. The extraction mechanisms are discussed in detail and the authors highlight the influence of some operational parameters. Obviously, the combination itself faces some limitations and challenges, which should be overcome for broad applicability.

There has been an increasing use of piezoelectric materials (either bulk solid or nanoparticles), for which mechanical activation affords charge separation through the piezoelectric effect. Clearly, ultrasonic vibration as mechanical energy has been harnessed by Meroni and associates, to activate a piezoelectric solid (ZnO), which enhances the sono-photocatalytic degradation of diclofenac, a common anti-inflammatory agent [10]. While only a slight enhancement in drug degradation was observed, a strong effect could be obtained for the mineralization process, reaching up to 70% of complete degradation. The morphologies of the piezoelectric material also influenced the extent of mineralization.

A sono-chemical (in strict sense indeed) advanced oxidation process, involving no more than sonication (600 kHz) and chlorine, is disclosed by Hamdaoui et al. for the degradation of persistent organic contaminants, such as textile dyes [11]. The protocol can compete with other oxidation processes such as sono-ozonation and UV/chlorine. As inferred from radical probes, the synergistic effect can likely be associated with the generation of chlorine and oxy-chlorine radicals. This sono-chemical transformation is, however, limited by other parameters like the presence of inorganic anions and organic matter (humic acids for instance) that decrease the overall efficiency.

Muñoz-Almagro and coworkers have reviewed the effect of high-intensity ultrasound and microwaves on food treatment [12]. This hybridization in agri-food industry is of paramount importance, not only as accelerating tool that avoids side degradation steps caused by prolonged irradiation, and lowers energy consumption, but also to preserve the bioactivity of food components. Both pros and cons, and experimental designs should be of broad interest.

Sono-ozonation has also been explored by Siddique et al. for the degradation of pesticides present in fresh fruit and vegetables [13]. Although the study concentrates on a local market in Pakistan, methods and conclusions can be of general application, and hence benefit. Significant reductions in pesticide content (five common chemicals were assessed to this end) could be achieved in short US/O<sub>3</sub> treatment (<10 min), while leading to good food safety.

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