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SONOCHEMISTRY AND ITS APPLICATION; A REVIEW

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ABSTRACT

Sequel to the pervasive nature of chemistry and the need to arouse renewed interest in some rare branches of chemistry, the field of sonochemistry and its applications was reviewed. The effects of ultrasonic waves on chemical reactions such as initiating disrupting or hastening chemical processes was highlighted. The use of sonochemistry in medicine, agriculture, pharmaceuticals and in industries were discussed. The application of ultrasound in analytical chemistry especially its use in sample preparation and acceleration of sample digestion were mentioned. It is hoped that this review will help in directing studies and research interest in this field of sonochemistry which may open up major breakthrough and major technological leap in science and technology in the future.

Keywords: Sonochemistry, Piezo-electric effect, ultrasonics, sonication, ultrasound, acoustics.

Introduction:

The foundation of the research in ultrasonics was laid down way back in 1880 following the discovery of piezo-electric effect (Piezozo-electric effect is the conversion of mechanical compression brought about by the compressing effect of travelling waves to electrical energy or electricity and the reverse is the re-stretching of this compressions and reconverting the electrical energy back to mechanical energy as in tranducers.) by Curies and its reverse phenomenon[1]. The following year in 1881, Langevin designed and fabricated the first ever ultrasonic based instrument in 1918. Later Pierce, Hubbord and Loomis[2] produced

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Ultrasonic instruments to measure ultrasonic velocities in liquid and gases. Parthasarthy was the first Indian scientist to measure and report ultrasonic velocities in a number of organic liquids, which are still taken as standard values. The initial activities of ultrassonics remained restricted to the measurement of ultrasonic velocity and attenuation, absorption of sound[3] in solutions. However, the inventions of improved electronics and better transducer materials such as quartz, ceramic and magnetostrictive devices to generate ultrasound from one hertz to several mega hertz range, led to the discovery of various applications of ultrasound in different fields of acoustics.[4] The list of applications of the ultrasound is too wide but as a chemist, any anticipation of the use of sound waves as a source of energy for initiating, completing, hastening or disrupting a chemical process must have remained only a very farfetched remote possibility,[5] until the discoveries in the later part of the twentieth century. Nevertheless, we would restrict our discussion to the basic principles of sonochemistry and only few important applications in chemistry, conveying the future role of this technology will be dealt with.

Theoretical Basis

The frequency of sound waves, beyond the human audible limits (16 Hz to 16 KHz), is called ultrasound,[6] which extends up to 500 MHz or even more with wavelength of 1.9cm or less. The upper limit of ultrasonic frequency for gases is 5MHz compared to 500MHz for liquids and solids. The use of ultrasound within this large frequency range may be broadly divided into two areas.[7] the first area involves low amplitude (higher frequency) propagation, which is concerned with the effect of the medium on the wave and is commonly referred to as low power and high frequency ultrasound. Besides the studies of the measurement of velocity and absorption co-efficient in this range (1-10 MHz), other important uses are in the fields of medical imaging, Chemical analysis and relaxation phenomenon. The second area involves high energy (low frequency) waves known as power ultrasound between 20 – 100 KHz, which is used for cleaning, plastic welding and effecting chemical reactivity.[8]

Definition of Sonochemistry

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The branch of chemistry dealing with the study of the effect of ultrasound waves (20 KHz–100 KHz), on chemical activity is known as sonochemistry.[9] In fact the observed chemical transformations generated through ultrasonic irradiation are not due to the interaction between ultrasound and the substances but a consequence of the effect of cavitation, which is (formation, growth and implosive collapse of bubbles) which the propagating ultrasound waves generate in a medium depending, of course, upon the power associated with ultrasound and the tensile strength of the solvent.[10] Cavitation generates short lived vapour filled cavities accompanied with an inward collapse when the size of the bubble is no longer tenable by the propagating ultrasonic waves. This implosion generate very high temperature[11] (≈ 5000 K), pressure (1500 atm) with a collision density of 1.5kg cm^{-2} and a pressure gradient of 2 Tpa cm^{-1} with life time shorter than 0.1μs, cooling rate above 10^{10} Ks^{-1} and a mass flow[15] of 500 ms^{-1}. These drastic conditions of temperature, pressure and mass flow make the tiny space of the micro-bubble an active site, which acts as a micro-reactor, generating free radicals, activated molecules and entirely different chemistry.[12] Usually not expected in the normal condition. Sonication of the water generates several free radicals (hydrogen, hydroxyl, peroxy) and activated molecular species (hydrogen peroxide, carbon dioxide, oxygen, hydrogen) as shown.[13]

$$H_2O \longrightarrow \dot{H} + \dot{O}H \quad (1)$$

$$H + H \longrightarrow H_2 \quad (2)$$

$$H\dot{O} + \dot{O}H \longrightarrow H_2O_2 \quad (3)$$

$$\dot{H} + O_2 \longrightarrow HOO \quad (4)$$

$$H\dot{O}O + HOO \longrightarrow H_2O_2 + \dot{O}_2 \quad (5)$$

$$H + \dot{H}_2O_2 \longrightarrow H_2O + OH \quad (6)$$

$$H_2 + \dot{O}H \longrightarrow H_2O + \dot{H} \quad (7)$$

$$H + \dot{O}H + HCOOH + O_2 \longrightarrow H_2O_2 + H_2O + CO \quad (8)$$

All chemical reactions, under the influence of ultrasound, therefore, proceed through free radical mechanism.[14] This makes these reactions quite different from those reactions carried under normal conditions and offer opportunity of unlimited applications of ultrasound in the sonochemical frequency range. There are three major types of sonochemical processes.[15]

Types of Sonochemical Reactions

Homogenous Reactions

Dissolved gases or bubbles in the medium act as nuclei for the formation of cavitation bubbles[16]. These micro bubbles are not enclosing a vacuum rather they contain vapour from the solvent and other volatile reagents so that on collapse, these vapours are subjected to the enormous increases in both temperature and pressure referred to above. Under such extremes, the solvent and/or reagent vapour suffers fragmentation to generate reactive species of the radical or carbine type, some of which would be high enough in energy. These high-energy species would be concentrated at the interface and thus give rise to intermolecular reactions or radical coupling.[18]

If there were volatile solutes present in homogenous reaction, these would also collect at the same interface. As a consequence these would also be subjected to extreme conditions on bubble collapse.[19] They could well react with the high-energy species generated in the vapour phase. In addition to these effects, the shock wave produced by bubble collapse could also act to disrupt solvent structure. This could then influence reactivity by altering the solvation of the reactive species in the immediate neighborhood of the disturbance.[20]

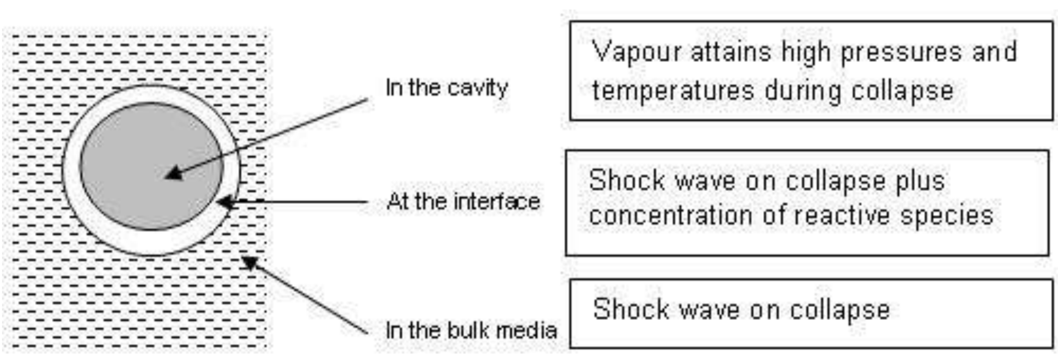

Fig 1: Chemical activity at the micro bubble in homogenous solution

Heterogeneous Reactions:

There are two types of reactions involving solids in liquids.

i. In which the solid is a reagent and is consumed in the process.
ii. In which solid functions as a catalyst.

In heterogeneous reactions, involving solids dispersed in liquids, the overall reactivity will depend upon the available reactive surface area. Ultrasonically induced enhancements in the chemical reactivity that are observed in such heterogeneous systems are because of the cleaning action of ultrasound (Fig. 2). It exposes clean or reactive surface to the reagent for further reaction. Examination of irradiated surfaces by electron microscopy reveals "pitting" of the surface of the solid, which acts both to expose new surface to the reagents and increase the effective surface area available for reaction. Two processes cause the pitting[21]

a. The implosion of cavitation bubbles formed from seed nuclei on thesurface[22]
b. Micro streaming of a jet of solvents on to the surface when a cavitation bubble collapses in the solvent close to it.

In many cases, where the cleaning effect alone is insufficient to explain the extent of sonochemically enhanced activity, it is thought that sonication also serves to sweep reactive intermediates or products, clear off the metal surface and thus present renewed clean surface for reaction.[23]

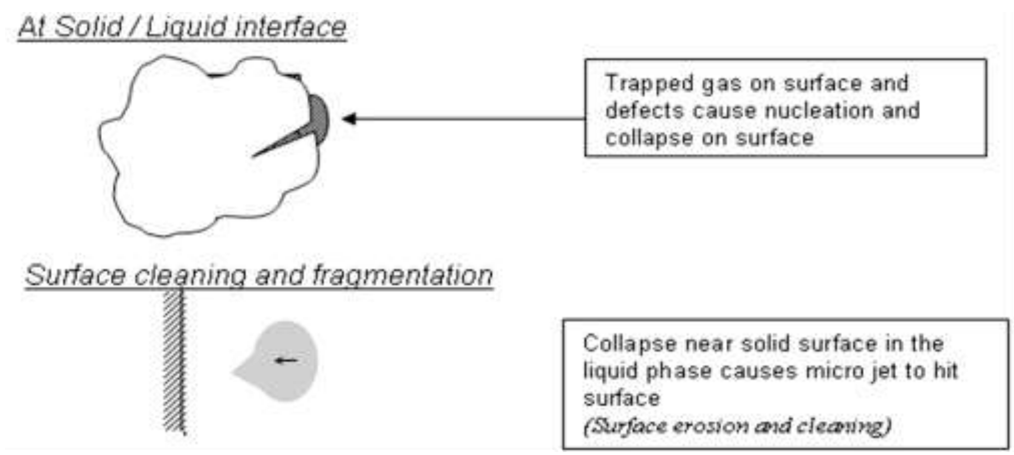

Fig 2: Cleaning of solid surface due to pitting action of ultrasound

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Reactions at a Liquid – Liquid interface: Ultrasound generates extremely fine emulsions from mixtures of immiscible liquids (Fig. 3). One of the main consequences of these emulsions is increase in the interfacial contact area between the liquids and increase in the region over which any reaction between species dissolved in the liquids can take place. This emulsification also leads to the use of ultrasound in place of phase transfer catalyst.[24]

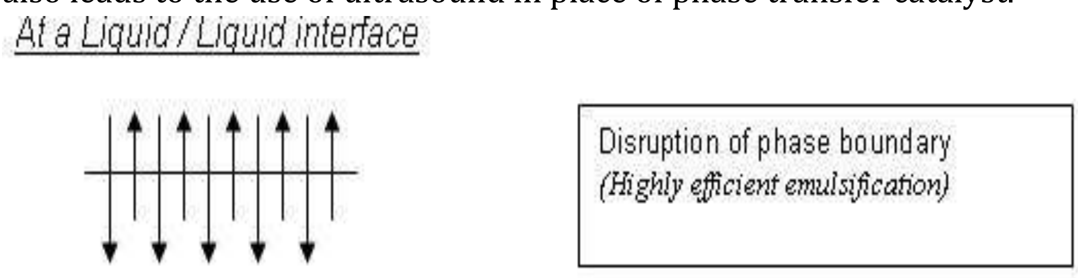

Fig 3: Inter-boundary movement of two immiscible

Applications of Sonochemistry

The real impetus to the research activity in the area related to sonochemistry began from the second half of the twentieth century and now a huge list of applications involving ultrasound can be made.[25] It is almost impossible to describe even in brief all of these applications, therefore, only a summarized description is being given in Table 1 to highlight the importance of this branch of chemistry[26], a brief discussion of some of the major applications, to economize the space in this article, is provided at the end.

Table 2: Summary of major areas of application of sonochemistry

PROCESS IMPROVEMENT	SOLIDS APPLICATIONS
i. Facilitating flow of viscous products by removing fluid friction (e.g. food-stuffs)[25]	i. Street relieving[30] ii. Facilitating powder transport in pipe[31]conduits extruders
ii. Extractions (e.g. mining industry, precious metals, perfumes[26] iii. Deforming (including large surfaces) incineration of liquids[27] iv. Petrochemical cracking[28] v. Superficial liquid CO_2 reactors fuel cells[29] vi. Nano-particles production[35]	iii. Wire and tube drawing[32] iv. Cutting (e.g. food, fabrics, industrial products)[33] **CLEANING** i. Continuous cleaning inside pipes[34] Nuclear industry heat exchanger, Micro-encapsulation, coating[36] and surface impregnation

Fuel injection Industrial fluids atomizers and gas mixing (air conditioning, semiconductor technologies) Large volume humidifies and dust removal Liquid alloy atomizers and solder atomizers Incineration of waste and dangerous liquids by atomizing	Ultra-filtration Fast meat defrosting and preparation For fuming and drying Sterilization Ice and Snow making Dust removal Relaxation and message therapies

Some Specific Application

Ultrasound can destroy all micro-organisms such as bacteria, phytoplankon and zooplanktons present in water and this water remains sterilized for over a week. Further, sonication also precipitates the dissolved bicarbonates of calcium and magnesium by converting those to respective carbonates, thus reducing the level of hardness of water up to about 14 percent and changing it to portable water[18]

A simple purification system for underground water, contaminated with arsenic, can be made using ultrasound, wherein ultrasound is propagated in a chamber partially filled with iron sulphide and overflowing with contaminated water. Arsenic, even in ppb range, precipitates as sulphide and can be filtered out.[19]

Application in Modifying Materials

Crystallization of a number of inorganic salts and new composites doped with rare earths may be synthesized in ultrasonic field for their possible application as novel and efficient photocatalysts besides the materials used as semiconductors in electronic industries.[37] Due to the impaction of ultrasound waves different atoms may be forced to new locations and sites in the lattice, modifying the electrical properties of materials.

Application in Acceleration of Reaction

Rate of reaction may be accelerated in a number of reactions, from hydrolysis to photochemical through decomposition, as a result of the enhancement from 2 to 64 fold due to the perturbation and turbulence created by the propagating ultrasound waves apart from the enhanced activity of the catalysts as a result of the particle size reduction[38] in some other reactions. Forcing conditions of many reactions may be avoided in sonicated conditions and processes may also be simplified.

Application in Extraction of Natural Products

Extraction of natural products becomes easier and yield of active ingredients also increases many fold due to the cavitational effect which is the effect of the implosive collapse of the cavity followed by attainment of very high temperature and pressure which are short-lived. When the imploding bubbles collapse, the cell wall bursts and releases the cell contents into the extracting solvent at much lower temperature in contrasts to that required for such extractions under refluxing conditions.[39] The requirement of the solvent and time is also much less than required otherwise. A change in chemical pathway may also occur and the products are sometimes entirely different from those normally obtained under unsonicated condition.[40]

Application in Crude Oil Refining

Sonication of crude oil before the separation of different fractions of petroleum products increases the distillable fraction as much as 25% than from those samples which are fractionally distilled without sonication.[41]Many refineries have already commissioned sonicating chambers in their refineries. This is a major breakthrough for increasing the bulk of distillate without increasing the production of crude oil.

Application in Textile and Drying

The use of a phase transfer catalysts is not at all required in those reactions which require solutes of different solubilities in solvents entirely different miscibilities.[42] A member of synthetic and textile dyes may be degraded without using any oxidizing reagent or chemical processing. This is a green way of removing organic moieties from effluents being discharged from small scale[43]cloth dying units. This will be useful as the government is trying to revive the textile industries in different parts of the country.

Application in geology

Applications of sonochemistry in the extraction of marine sediments and detection of metallic beads in the sea-beds make this technology useful to geologists as well.[44]

Application in Synthesis of Nano-Particles

Recent work have also demonstrated the application of ultrasound in the synthesis of nano-particles of a number of metals and their compounds[45] such as Cu, Se, Au, Cr, Mn and Co.

Application in Biomaterial Preparation

Another important application has been the sonochemical preparation of biomaterials,[46] most notably protein microspheres. Using high intensity ultrasound and simple protein solutions, a remarkably easy method to make both air-filled microbubbles and liquid-filled microcapsules was developed. This sonochemical method of micro encapsulation uses high intensity ultrasound to induce emulsification and to create oxidative cross linking of protein cysteines. These microspheres are stable for months, and being slightly smaller than erythrocytes, can be intravenously injected. These protein micro spheres have a wide range of biomedical applications, including uses for in vivo thermotherapy, in vivo oximetry, MRI contrast agents, and drug delivery, as shown in Fig. 4.

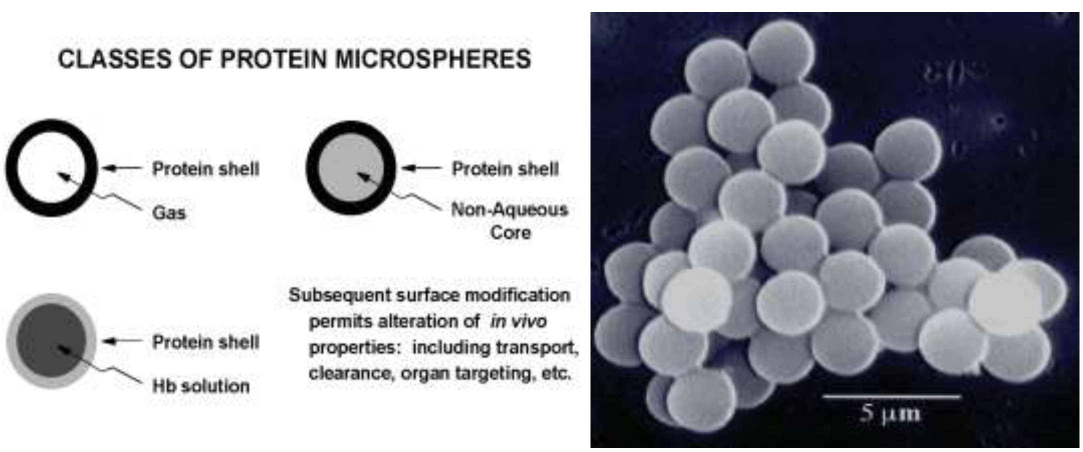

Fig 4: Sonochemical Preparation of protein Nanoparticles Micropheres

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Applications of Sonochemistry in Analytical Chemistry

Ultrasonic assistance is gradually becoming quite common place in analytical chemistry. The different steps of analytical process can be expedited and/or improved by use of ultrasound energy[47]. One of the most common applications of ultrasound is sample preparation[48]. Preparation of solid sample is unavoidable in chemistry ultrasound has been found useful in acceleration of sample digestion the use of ultrasound in sample preparation has provided more effective than the normal orthodox digestion method as the heat in this method tempers with the matriz of the sample[49].

Application of Sonochemistry in Food and Pharmaceutical Industries

The application of sonochemistry in the pharmaceutical industry will be discussed separately because of the sensitivity of the area. Sonochemistry has been applied in several aspects of pharmaceuticals from extraction of active ingredient used in preparation of drugs to the delivery of the drug to the patient's blood stream[50] we shall discuss some of these applications briefly;

Ultrasound chemistry:

A rapid sensitive and accurate ultrasound extractive and rapid gravimetric method has been developed for the determination and extraction of nicotine in pharmaceutical formulation. The result obtained in that study show that ultrasound is a fast reliable too for the extraction of nicotine in pharmaceutical formulations[51].

Ultrasound driven powder transport system:

The transport and dosage of granular materials are important component of process engineering, very accurate mixing process in chemical, pharmaceutical and food industries demand for an exact control of powder feeding a novel powder-feeding device developed at the Heinz[52].

Nixdorf institute and is based on piezoelectrically stimulated travelling waves. An acrylic pipe, which is stimulated to oscillations in the form of travelling waves piezoelectric impulses is used to convey the powder[53]. An ultrasound driven powder transport system was developed which distinguished itself from conventional dosage system through the following points: simple, nearly no wear and tear in generation, cost effective and easy to integrate into existing production plant[54].

Application of ultrasound in transdermal drug delivery (TDD)

Transdermal drug delivery offer several advantages over traditional drug delivery systems such as oral delivery and injections the attractive attributes of Tdd includes, avoidance of first pass metabolisms, elimination of pain associated it injection[55] and opportunity for the sustained release of drugs. TDD has been enhanced by the action of ultrasound referred to as sonophoresis. Sonophoresis is an active form of a Tdd which enhances the transport of permeants such as drugs through cell membranes[56] as a result of ultrasound energy. Ultrasound sound waves cause acoustic cavitation, the resultant effect of which microscopically disrupt the lipid bilayers of the stratum corneum and thereby influencing the influx of permeants[57] sonophoresis increases the penetration of various low molecular weight drugs as well as high molecular weight proteins[58]. Sonocrystallization in formation of aerosols.

Sonochemistry in cell therapy

Nacl aerosols have been widely used as part of bronchial provocation test to identify people with active asthma, exercise induced asthma. But this procedure is energy intensive, time consuming and impurity including production of smaller sized crystals and cost effectiveness of apparatus, the process can be run at ambient conditions and reaction vessel involved is of simple geometry making the cleaning process simple for pharmaceutical requirements[31].

Use of ultrasound in chemotherapy

Ultrasound has been used in medical imaging and diagnostics cald ultra sound, scan, it has also been used in biological cell disruption and fermentation processes. The application of power ultra sound (20-100KHz or even up to 2MHz) to biological cells destroys the cells.

Ultrasound irradiation of toxic effluents

Considerable interest has been shown on the application of ultrasound for hazardous chemical destruction, including among others. The degradation of chlorinated hydrocarbon, aromatic compounds and pesticides.

Sonochemistry in cell therapy

By combining focused ultrasound technology with the properties of magnetic resonance imagine a system has been developed (Exablate, 2000) which enables precise targeting within tissues. Studies have been carried out in many different areas including breast, brain and liver tumors and uterine fibroids.[41]

Conclusion

This review has tried to highlight the field of sonochemistry as a promising branch of chemistry that can be further harnessed for the good of man because of the numerous are shown of applications of sonochemistry. It has shown that further research on this area could open up major breakthrough in science and technology.

Recommendations

I recommend that more efforts of research should be directed toward sonochemistry. Adequate funding from government and educational institutions should be provided for research on this branch of chemistry. This will be good for national development.

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