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Abstract: This review succinctly describes the recent progresses in sonochemistry for the preparation of nanocatalysts as well as the use of ultrasound in organic reactions involving nanocatalysts. The main recent uses of power ultrasound in these fields reported here allow to determine some interesting trends and perspectives for this research, but also highlight some needs to innovate.

Keywords: sonochemistry; power ultrasound; nanocatalysis; materials; organic reactions.

Introduction

Sonochemistry is the use of **power ultrasound** with frequencies between 20 kHz and 2 MHz (in opposition to diagnostic ultrasound particularly used in medical imaging with frequencies between 2 MHz and 200 MHz) to provide chemical, thermal and physical effects in solution [1]. The effects of ultrasound (intense local heat up to 5,000 K, very high pressure close to 1,000 bar, divergent shock waves, acoustic microcurrents and violet liquid microjets up to 100 $m.s^{-1}$) are the consequence of the **cavitation phenomenon** (formation, growth and collapse of gaseous microbubbles in a liquid phase, phenomenon represented in Figure 1) [2]. The potential and applications of sonochemistry is directly connected to the choice of the sonochemical parameters. For example, the frequency is a crucial parameter and, in water, it is usually accepted that **low frequencies** (**20-80 kHz**) lead preferentially to physical effects (microconvection, shockwaves, microjets, etc) whereas at **high frequencies** (**150-2,000 kHz**) the chemical effects and the production of hydroxyl radical species ($HO^{\bullet}$) are favored. Conditions obtained in a medium submitted to power ultrasound are accountable for a large number of physico-chemical effects such as the increase in kinetics of chemicals reactions, changes in reaction mechanisms, emulsification effects, erosion, crystallization, precipitation, etc [3,4].

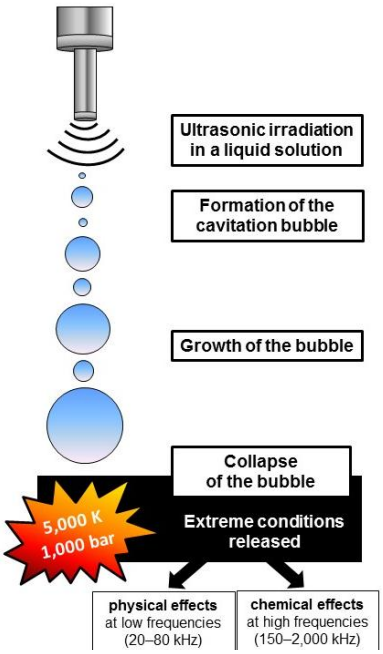

Figure 1: Cavitation phenomenon at the origin of the effects provided under power ultrasound.

Sonocatalysis, organic sonochemistry and sonochemical preparation of materials are the most investigated uses of power ultrasound these last decades, even if other applications in polymer chemistry, biomass valorization, extraction and environmental remediation also widely explored sonochemistry [5]. The scope of this review is limited to efforts of the last two years in the areas of ***sononanocatalysis***, with one part of the studies involving ultrasound for the preparation of nanocatalysts and the other part applying ultrasound in organic reactions in the presence of a nanocatalyst.

Sonochemical preparation of nanomaterials

The radical and mechanical effects of ultrasound have been widely investigated for the preparation of nanocatalysts or specific nanostructured materials. Suslick *et al.* reviewed in 2013 how ultrasound was used for the synthesis of nanostructured solids through different methods: reduction from the solvent sonolysis (Pt, Pd colloids), sonoelectrochemistry (metal colloids), spray pyrolysis and sonolysis of volatile organometals (zeolites, metal alloys, metal carbides/nitrides, sonogels, metal oxides, metal sulfides, semiconductors and exotic carbons) [6].

Recently, CO_2 reforming of CH_4 to produce hydrogen was investigated in the presence of bimetallic Ni-Co/Al_2O_3-MgO [7] and Ni-Co/Al_2O_3-ZrO_2 [8] nanocatalysts prepared under ultrasound at low frequency (20 kHz, immerged probe, Table 1, Entries 1 and 2). Authors reported that the irradiation of ultrasonic waves during impregnation process improves the morphology, specific surface area, particles size distribution, dispersion of metallic sites and consequently, the catalytic activity and anti-coking performance in the dry reforming of methane.

Fuel cell grade hydrogen was also produce *via* steam methanol reforming using CuO/ZnO/ZrO_2/Al_2O_3 [9] and CuO/ZnO/CeO_2/Al_2O_3 [10] nanocatalysts, synthesized by sonochemical coprecipitation at low frequency (Table 1, Entries 3 and 4). The FESEM (Field Emission Scanning Electron Microscope) images described that the nanocatalysts had nanometer particles which became smaller by increasing the ultrasound power from 30 W to 90 W (electric output). Low frequency ultrasound also prevented the Cr/clinoptilolite catalyst desactivation during the synthesis [11,12]. The same advantages were highlighted for the preparation of sono-sulfated zirconia nanocatalyst supported on MCM-41 (Table 1, Entry 6) [13].

Palladium oxide (PdO) nanoparticles deposited on silica nanoparticles [14], $NiFe_{2-x}Eu_xO_4$ nanostructured catalysts [15], $CoFe_2O_4$/B_2O_3-SiO_2 magnetic composite nanostructure [16], sulfuric acid-functionalized silica-based magnetic core/shell nanocomposite [17], and Cu_2O-nanocubes heterogeneous nanocatalyst [18] were synthesized and studied in fine chemistry applications, showing better catalytic activity, better yields, shorter reaction times, the possibility to easily recover the catalyst and presenting some advantages at an environmental point of view (Table 1, Entries 9-11).

Interestingly, it seems that the coated nanomaterials (with capping agents such as polyvinyl pyrrolidone, phenolics, etc) are not negatively impacted by ultrasonic irradiation. On the contrary, some examples noted that the coating is stable and cannot be removed by a simple washing procedure with water, or ethanol and/or acetone after an ultrasonic treatment [19].

Other recent applications reported from the use of ultrasonic prepared nanocatalysts are about photodegradation of dye and pollutants in aqueous effluents [20-22]. Interesting researches proposed to combine ultrasonic and solar light irradiations as degradation processes based on well define metal-supported nanoparticles [23,24]. Sonophotodeposition (ultrasound and light radiations) and sonoelectrodeposition (ultrasound and electrical fields) processes have been also recently shown synergetic actions in the syntheses of different nanomaterials for photodegradation steps [25].

Table 1: Recent examples of sonochemical preparation of nanocatalysts.

Entry	Nanocatalyst	Experimental conditions	Applications of the catalyst	Advantages described by the authors	Ref.
1	Ni-Co/Al_2O_3-MgO	20 kHz (probe), 90 W, 45 min, temp. not controlled	CO_2 reforming of CH_4 for hydrogen production	Enhancement of the dispersion of active phase and surface area in comparison with impregnation method	[7]

2	Ni-Co/Al_2O_3-ZrO_2	20 kHz (probe), 30-90 W, 20-80 min, temp. not controlled	CO_2 reforming of CH_4 for hydrogen production	Improvement of the morphology, specific surface area, particle size distribution, dispersion of metallic sites and catalytic activity	[8]
3	CuO/ZnO/ZrO_2/Al_2O_3	20 kHz (probe), 30-90 W, 45 min, temp. not controlled	Steam MeOH reforming for hydrogen production	Decrease of copper and zinc oxides crystallinity. Surface area enhancement	[9]
4	CuO/ZnO/CeO_2/Al_2O_3	20 kHz (probe), 30-90 W, 5-15 min, temp. not controlled	Steam MeOH reforming for hydrogen production	Improvement of the homogeneity and reduction of particles size	[10]
5	Cr/clinoptilolite-ZrO_2	20 kHz (probe), 90 W, 60 min, temp. not controlled	CO_2- dehydrogenation of ethane	Reduction of the formation of Cr_2O_3 and decrease to nanometer scale the chromium particles	[11]
6	S-ZrO_2/MCM-41	20 kHz (probe), 30-90 W, 30 min, temp. not controlled	Biodiesel production from sunflower oil	Improvement of the morphology, particle size, surface area and particle distribution compared to non-sonicated sample	[13]
7	PdO doped silica nanoparticle	20 kHz (probe), 80 $W.cm^{-2}$, 15 min, temp. not controlled	Selective aerobic alcohol oxidation	Uniform size and shape. Preservation of the crystalline structure without agglomeration.	[14]
8	$NiFe_{2-x}Eu_xO_4$	20 kHz (probe), 65 W, 10 min then 70 W, 30 min, temp not controlled	Synthesis of benzimidazoles, benzoxazoles, and benzothiazoles	Decrease of the particle size and increase of surface area.	[15]
9	$CoFe_2O_4$/B_2O_3-SiO_2	40 kHz (bath), 250 W (max), 1 h, 60 °C	Synthesis of dihydroquinazolinones	Increase of the catalytic activity	[16]
10	Fe_3O_4/SiO_2 Fe_3O_4/SiO_2-SO_3H	40 kHz (bath), 250 W (max), 1 h, 60 °C	Synthesis of nitroaromatic compounds	Reduction of particles size. Increase of the catalytic activity	[17]
11	Cu_2O-nanocubes	20 kHz (pulse mode 5s/5s) 15 min, temp. not controlled	Synthesis of quinazolines	Increase of the catalytic activity	[18]
12	CdO-ZnO nano-photocatalyst	20 kHz (probe), 90 W, 45 min, temp. not controlled	Removal of acid orange 7 from wastewater	Better results in both morphological characterization analyses and photocatalytic performance experiments	[20]
13	martite nanocatalyst	36 kHz (bath), 150 W, 30 min, temp. not controlled	Treatment of a textile dye	Increase of the catalytic activity	[21]
14	Ag/$AgFeO_2$ nanocatalyst	45 kHz (bath), 100 W, 0.5-20 min, temp. not controlled	Typical azo dye photodegradation	Enhancement of the photocatalytic performances	[22]

Sonochemistry in the presence of nanocatalysts

Several reviews have covered the large scale of applications of ultrasound in sonocatalysis [5,26,27, 28]. Generally, it is difficult to clearly identify the real effects brought by ultrasound irradiation and to exactly understand and prove how it can affect the reactivity. However, the main consequences observed are interesting such as an improvement of yield, selectivity or rate, reduction of reaction time, and sometimes a change in the mechanistic pathway [5].

Recently, a study compared an ultrasonic-assisted catalytic transesterification using ZnO nanocatalyst with the conventional stirring method (Table 2, Entry 1). In the sonochemical method, 96% of yield was achieved at constant temperature and reaction time was reduced from 1 h to 15 min. The properties of the obtained biodiesel from both methods showed many similarities [29].

Table 2 reports the studies of the studies from the literature using ultrasound as activation methods for catalytic organic reactions, in the presence of a nanocatalyst [30-39]. In all these cases, only low ultrasonic frequencies (19.5 to 40 kHz) were used *via* an immerged probe or a cleaning bath. The consequences of the use of ultrasound were the same in all the studies: (i) the reaction time was drastically reduced (4-45 min) compared to silent conditions; (ii) the conversion and the yield were improved; (iii) the reaction was performed at low temperature and pressure (generally ambient). These results have been explained by the effects of mechanochemical energy or micromixing and emulsification effects, both provided by cavitation [40].

Table 2: Recent examples of sonochemical reaction performed in the presence of nanocatalysts.

Entry	Nanocatalyst	Experimental conditions	Reaction/applications	Advantages reported by authors	Ref.
1	ZnO	32 kHz (bath), 15 min, 60 °C	Transesterification for bidiesel production	Better yields and higher conversion efficiency. Reaction time significantly reduced	[29]
2	$CuFe_2O_4$	20 kHz (probe), 30-60 W, 4-10 min	Synthesis of pyrido[2,3-d:6,5-d]dipyrimidines	Excellent yields, high purities and short reaction times	[30]
3	Fe_3O_4/Ph-SO_3H	Low frequency (bath), 10-50 min,	Synthesis of tetrahydrobenzo[b]pyrans	Excellent results were obtained at room temperature and short time	[31]

		room temperature			
4	Fe_3O_4/ 4,5-imidazoledicarboxylic acid	37 kHz (probe), 150 W, 5-12 min, room temperature	Synthesis of benzimidazoles	High yields, low reaction times, and easy catalyst workup by an external magnetic field	[32]
5	Fe_3O_4	20 kHz (probe), 180 W, 45 min, 60°C	Synthesis of secondary amides *via* Beckmann rearrangement	Simplicity, excellent yields, mildness and eco-friendly aspects of the synthetic protocol	[33]
6	Fe_3O_4/PEG/succinic anhydride	No frequency indicated, 60 W, 15-35 min	Synthesis of benzoxanthenes.	Easy work-up, short reaction times, recycling of the catalyst, little catalyst loading	[34]
7	Fe_3O_4/SiO_2-CO-C_6H_4-NH_2	40 kHz, 250 W, 3x30 min,	Multicomponent synthesis of pyridoimidazoisoquinolines	Easy workup procedure, recyclability of the heterogeneous nanocatalyst by an external magnet	[35]
8	Fe_3O_4/clay core/shell nanocomposite	40 kHz (bath), 250 W, 15 min, room temperature	Synthesis of imidazo(thiazolo) pyrimidines	Avoiding hazardous organic solvents and toxic catalysts, easy work-up procedure, short reaction times, excellent yields and mild reaction conditions	[36]
9	Multiwall carbon nanotubes/TiO_2	40 kHz (bath), 250 W, 15-20 min, room temperature	Selective conversions of alcohols and alkenes to aldehydes, ketones and epoxides.	Easy workup procedure, excellent conversion and yields, short reaction times and mild reaction conditions	[30]
10	$CoFe_2O_4$	40 kHz (bath), 770 W, 40 °C, 20-30 min	Synthesis of 2,4,5-trisubstituted imidazoles	Very fast, green and low-cost procedure	[35]
11	SBA-SO_3H	19.5 kHz (probe), 600 W, 8-25 min, 90-140 °C	Synthesis of imidazoles	Enhancement of the mass transfer with no need to anchor, confine or embed an additional hydrophobic group or ionic liquid	[36]
12	Cu/TiO_2	24 kHz (probe), 400 W, 8-12 min	Multicomponent synthesis of 1,4-disubstituted 1,2,3-triazoles in water	Reduction of the reaction time and temperature	[37]
13	Fe_2O_3@SiO_2-$(CH_2)_3NHC(O)(CH_2)_2PPh_2$ organic–inorganic core–shell nanomagnetic catalyst	40 kHz (bath) 250 W, 11 min	Synthesis of tetrazolo[1,5-a]pyrimidine derivatives	Easy workup procedure, excellent yields and mild reaction conditions.	[38]
14	$MgFe_2O_4$	20 kHz (probe), 35 W, 10-30 min, 45 °C	Knoevenagel condensation	Mild reaction conditions, excellent yields and short reaction times	[39]

Some recent examples also reported the use of heterogeneous nanocatalysts for environmental remediation applications, testing their sonocatalytic [41-45] and sonophotocatalytic activities [46] for the degradation of pollutants in liquid effluents. In these recent examples, low ultrasonic frequencies have been used allowing the reduction of treatment times. The difference with other examples of catalytic organic reaction is the difference of initial concentration in substrate, very low in the case of degradation processes (only traces of pollutants to degrade until total mineralization).

Conclusions, perspectives and recommendations:

Interestingly, the majority of the studies reported during these two last years has been published in the Elsevier journal *Ultrasonics Sonochemistry*. It is logical that studies involving power ultrasound in chemistry processes are published in this thematic and specialize journal, especially dedicated to the community of sonochemists. However, it could be also interesting to communicate in journals with more general audience of chemists to demonstrate the potential of ultrasound for all the areas of chemistry. Indeed, it is important to remember that ultrasound is not just a *simple mixing tool*, but **a real innovative technology** that can lead to results never obtained in classical conditions.

In conclusion, Table 1 shows that ultrasonic probes are mainly used (direct irradiation) compared to ultrasonic baths (indirect irradiation), probably to benefit from the sufficient power of irradiation and improve the physical effects provided by low frequency ultrasound. Generally, it was clearly reported that, under ultrasound, treatment times are reduced and performance of the nanocatalysts are improved. In addition, the main physical effects of ultrasound are from the high-speed jets and intense shock waves induced by cavitation. Enhanced mass transfer as a consequence of acoustic streaming and bulk thermal heating is another physical effect of ultrasound used in the preparation

of nanomaterials [47]. Physical effects of ultrasound can also be used to promote the diffusion of dopant ions into spherical nanoparticles [48].

However, the control of the temperature during the process is not often performed. The rigorous control of temperature conditions during the ultrasonic irradiation is crucial to clearly identify the effects of ultrasound in the preparation of a catalyst, but also in terms of reproducibility of its syntheses.

Table 2 shows the advantages of ultrasound for applications in fine chemistry and production of fuels, in addition of the studies about the degradation of pollutants. In all these cases, the reaction times are very short (10-30 min in average) and the energy consumption is low, compared to silent conditions, showing some interesting advantages in term of green chemistry. The contribution of sonochemistry to green chemistry have been recently discussed, based on the advantageous consequences obtained under ultrasound [49]: better yields and selectivities, reduced reaction times, new reactivity, use of water as solvent, increasing or optimization of the reactivity in the presence of catalysts, dispersion and reduction of particles, etc.

However, in the recent examples, no data on the real acoustic power dissipated in the sonochemical reactor has been reported. Only electric powers indicated by the generator (provided by the constructor) have been specified. It is clearly recommended to systematically report an estimation of the dissipated ultrasonic power and energy costs, to energetically compare the systems with each other. As an industrial point of view, the overall energy consumption of a sonochemical process is also essential.

A last, from a theoretical point of view, investigations on mechanisms are also required to better understand and use of sononanocatalysis in order to really benefit from the entire potential of ultrasound. The research only based on "*is better than*" is not sufficient at this stage: new clues and new proofs are clearly necessary for advances in the area.

Competing interests statement:

The author declares having no competing interests.

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