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Usage of Sonochemistry in Medicine, Industry, Environment, and Synthesis

ABSTRACT

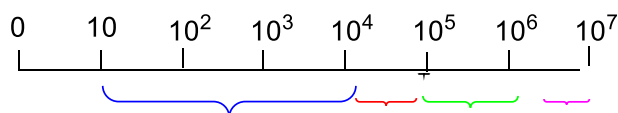
Much progress has been made regarding ultrasound in chemical science and the chemical industry in recent years. Ultrasonic waves are defined as inaudible sound waves with frequencies exceeding 20 kHz. This paper focuses on using ultrasonic technologies in some areas such as polymer degradation, polymerization reactions, removal of toxic organic contaminants in water, organic synthesis, ultra-strong transfer processes including the extraction process, adsorption process, membrane process, demulsification, crystallization process, emulsification, heterogeneous chemical reaction processes, and the electrochemical process.

Keywords: Environment, industry, medicine, organic synthesis, sonochemistry, ultrasound

Introduction

Much progress has been made regarding ultrasound in chemical science and the chemical industry in recent years. Ultrasonic waves can be defined as “inaudible sound with a high frequency for humans” the frequency of which approximately exceeds 20 kHz. This paper focuses on using ultrasonic technologies in some areas such as polymer degradation, polymerization reactions, removal of toxic organic contaminants in water, organic synthesis, ultra-strong transfer processes including the extraction process, adsorption process, membrane process, demulsification, crystallization process, emulsification, heterogeneous chemical reaction processes, and the electrochemical process.

Sound is a vibration that propagates as an acoustic wave in a transmission medium such as a gas, liquid, or solid. It represents sound waves with wavelengths between 17 meters and 1.7 centimeters in air at atmospheric pressure. Sound waves with frequencies above 20 kHz are called ultrasounds and are inaudible to humans. In contrast, sound waves below 20 Hz are referred to as infrasound. (Figure 1).



Human hearing 16 Hz-18 kHz

Conventional ultrasound 20-100 kHz

Range for sonochemistry 2 kHz-2 MHz

Diagnostic ultrasound 5-10 MHz

Figure 1. Frequency ranges of sound and its application (Mason and Bernal, 2003)

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Sonochemistry is chemistry assisted by ultrasound. Ultrasound is any sound wave with frequencies above the normal hearing range of the human ear (i.e., above 16 kHz) (Savun-Hekimoğlu, 2020). Ultrasounds have proven useful in many fields, including medical imaging, also called sonography. In addition, it is used to realize and accelerate chemical reactions. Transducers, mostly made of piezoelectric ceramics are used in sonochemistry (Mason, 1997).

As in all chemical reactions, chemical bonds are broken and re-formed in sonochemical processes (Li et al, 2021). When ultrasonic waves (if their frequency is greater than 20 kHz) propagate through a liquid, chemical bonds are broken and free radicals are formed. Ultrasound is used to accelerate organic reactions, increase yield, reduce reaction time, and provide milder reaction conditions. Ultrasound promotes effective mixing in solid-liquid and liquid-liquid mixtures (Kaur, 2019). This feature is also used in various phase transfer-catalyzed reactions (Davidson et al, 1987). The pharmaceutical and food industries (Mason, 1995; Mason et al., 1996; Fresno et al. 2018; Chen et al., 2020; Rastogi et al., 2011; Wang et al., 2018; Sharma et al., 2020), extraction processes (Mason et al., 1996; Ebringerová & Hromádková, 2010; Alves Filho et al., 2020; Juliano et al., 2017; Kumari et al., 2018; Martínez et al., 2020; Ojha et al., 2020; Paniwnyk et al., 2009; Kulkarni & Rathod, 2014; Chen, 2012; Dey & Rathod, 2013; Shoh, 1988; Mason et al., 1990) waste treatment (Shoh, 1988; Ingole & Khedkar, 2012; Savun-Hekimoğlu & Ince, 2017; Gogate et al., 2003; González-García et al., 2010; Wang & Zhu, 2005; Nejmal et al., 2014; Yousef Tizhoosh et al., 2020; Naddeo et al., 2010; Monsef et al., 2019; Yadav et al., 2020; Serna-Galvis et al., 2019; Zinatloo-Ajabshir et al., 2020; Kerboua et al., 2021; Adewuyi, 2005; Mason & Lorimer, 1988), materials chemistry (Peters, 1996; Yuan et al., 2020; Cruz-Benítez et al. 2021), analytical chemistry (Robin et al., 2017; Wieland et al., 2019; Bendicho, 2012; Seidi & Yamini, 2012), emulsification (Shoh, 1988; Mason & Lorimer, 1988), material processing (Shoh, 1988; Mason & Lorimer, 1988), food processing (Mason et al., 1990; Mason & Lorimer, 1988) and nanoparticle synthesis (Kumari et al., 2018; Monsef et al., 2019; Chatel, 2019; Hujjatul Islam et al., 2019; Dheyab et al., 2021; Ersan, et al., 2020; Asfaram et al.; 2018, Bayrami et al., 2019) are the application areas of ultrasonic waves (Machado et al., 2021).

Applications of Sonochemistry

Based on the studies in the literature, this article will present the studies in the literature in chronological order.

Chemical effects from ultrasound in aqueous solution were first described in two separate papers in 1927 (Richards & Loomis, 1927; Wood & Loomis, 1927).

In 1953, Busnel et al. published a paper describing the relationship between wavelength and the oxidation of potassium iodide by ultrasound (Busnel et al., 1953). A similar study was published by Renaud in the same year (Renaud, 1953).

In 1955, an article on the examination of the ultrasonic energy sensor was published by d'Acoustique (d'Acoustique, 1955).

Weissler et al. sonolyzed acetonitrile under argon gas and obtained N₂, CH₄, and H₂ in 1965. When they performed the same experiment under oxygen gas, they obtained N₂, CO, CO₂, and H₂O. The ultrasound pulse-echo method has been used to obtain two-dimensional high-resolution images of the internal structures of the human heart (Åsberg, 1967). Ultrasonic flow detection was used as a useful technique in the evaluation of peripheral vascular disease (Strandness et al., 1967).

Fundamental developments in ultrasound, especially in radiological imaging, began after the 1970s. Leopold and Joel Sokoloff demonstrated that B-scan ultrasonography is useful in cases where radiographic imaging is not available or when hepatobiliary dysfunction is severe enough to preclude oral cholecystography or intravenous cholangiography (1973). Imaging of ventricular septal defects with cardiac ultrasonography has been achieved (King et al. 1973).

Styrene (1) can be cyclopropanated in 96% yield in 1 h when a combination of both sonication and mechanical stirring is used in 1982 (Figure 2) (Regen & Singh, 1982).

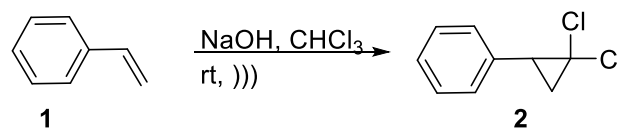


Figure 2. Cyclopropanation of styrene (39)

Davidson performed the N-alkylation of amines, synthesis of ethers and esters, and hydrolysis of esters under ultrasonic conditions in 1987. This study contains results supporting the idea that the presence of oxygen in a cavitating liquid produces singlet oxygen in addition to peroxy radicals. In another study, Henglein discussed the chemical effects of ultrasound used in medical diagnosis (. Henglein, 1987).

In 1988, Mason and Lorimer explained the theory of ultrasound and its application areas in chemistry. Suslick published articles on the chemistry, physical, and biological properties of ultrasound (Suslick, 1988). A year later, Lay and Low described the use of ultrasound in synthesis (1989).

In 1990, Mason published two studies on ultrasound chemistry. Lindley explained the usage of sonochemistry in inorganic and organometallic chemistry including catalysis (1990). Rienz et al. (1990) presented a paper on the sonochemistry of volatile and non-volatile solutes in aqueous solutions.

In 1992, the effects of ultrasonic irradiation at different frequencies, i.e. 20, 40, 60, and 80 kHz, on the activation of a model chemical reaction (iodide to iodine oxidation) were investigated (Cum et al, 1992). Mason et al. (1992) reported how ultrasonic energy input to a chemical reaction is affected by ultrasonic power used, the presence of bubbled gas, temperature, solvent composition, and reaction volume. Ultrasonic diagnostic devices can have possible harmful effects. Riesz and Kondo (1992) discussed the roles of free radicals and the mechanical effects of ultrasound in DNA degradation, inactivation of enzymes, lipid peroxidation, and cell killing.

In 1994, Olson and Barbie presented an advanced oxidation process in which ultrasound was studied in terms of its potential to oxidize natural organic matter to refractory electrolytes. Entezari and Kruus (1994) researched the effect of sonochemistry in the oxidation of iodide.

In 1995, Low explained the use of ultrasound in synthesis. Suslick et al. (1996) described the use of sonochemistry in material synthesis in 1996. In 1996, Ophir et al. presented a paper on ultrasonic imaging of tissue tension and elastic modulus under in vivo conditions in the field of breast imaging. Li et al. (1996) synthesized 5,5-disubstituted hydantoin 4 from ketone 3 ($(\text{NH}_4)_2\text{CO}_3$ and NaCN with ultrasound in 1996 (Figure 3).

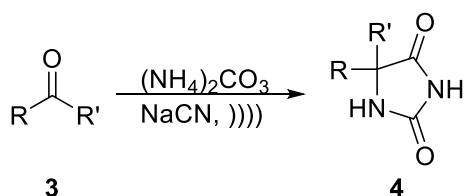


Figure 3. The synthesis of 5,5-Disubstituted Hydantoin 4

The hydrolysis of 4-nitrophenyl ester derivatives 5 ($\text{R} = \text{Me, Et, Pri, But}$) was carried out in an ultrasonic bath at 35 °C, and it was observed that the reaction rates increased in 1997 (Mason) (Figure 4).

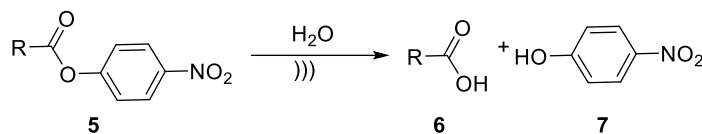


Figure 4. The hydrolysis of 4-nitrophenyl ester derivatives 5

The alkylation reaction of 5-hydroxy-4-oxo-4H-1-benzopyran-2-carboxylic acid ethyl ester (8) was carried out by Mason using several different haloalkanes under sonication in 1997.

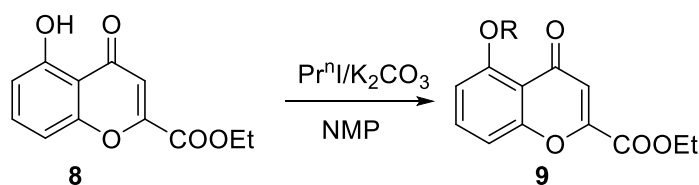


Figure 5. The alkylation reaction of 5-hydroxy-4-oxo-4H-1-benzopyran-2-carboxylic acid ethyl ester (8)

In the same study, he used ultrasound to increase the reactivity of the metal and saw that the reaction rate increased as a result (Figure 5). In another study Petrier explained the degradation rate of phenol and carbon tetrachloride at different frequencies, 20 kHz, 200 kHz, 500 kHz, and 800 kHz (1997).

Fillion and Luche (1998) described the effect of sonochemistry in cycloaddition reactions in their books. Crum et al. (1999) published a chapter on sonochemistry. Lubinski et al. (1999) described the use of ultrasound in medical ultrasound elasticity imaging.

There are examples of sonochemical [4+2] and [3+2] cycloaddition reactions in the literature. One of these reactions is the addition of o-benzoquinone (10) with furan. The effect of sonochemistry has been investigated in studies (Figure 6) (Cabello et al, 2003; Avalos et al., 2003).

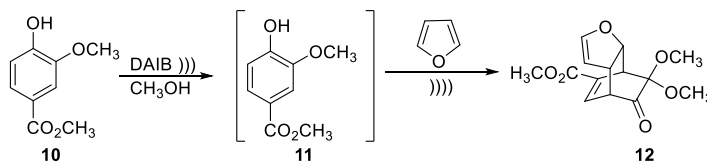


Figure 6. Sonochemical [4+2] and [3+2] cycloaddition

reactions

Li et al. (2003) succeeded in the synthesis a,a-bis(substituted benzylidene) cycloalkanones **14** catalyzed by $\text{KF}/\text{Al}_2\text{O}_3$ under ultrasound irradiation (Figure 7).

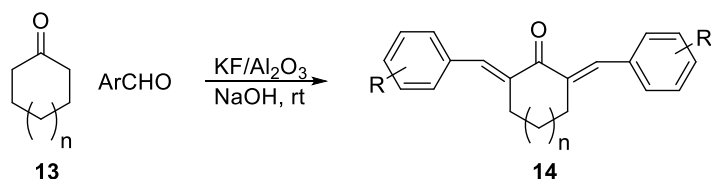


Figure 7. The synthesis of substituted benzylidene cycloalkanones

Leveque and Cravotto (2006) explained a method using ultrasound in green chemistry. For example, Hydrazinolysis of methyl salicylate **15** occurred in ultrasound with a yield of 79%. Another example is the Mannich reaction of **17** under ultrasound irradiation (Figure 8).

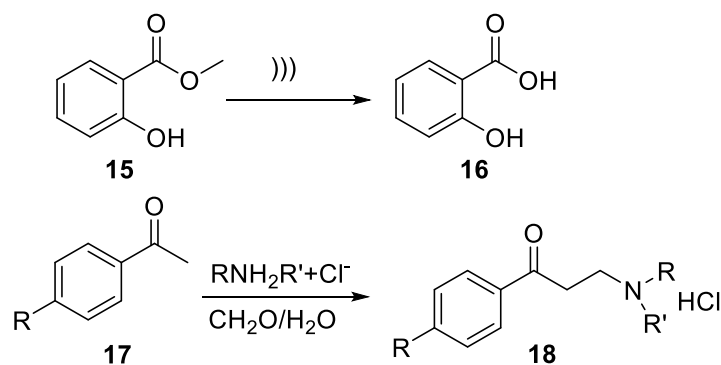


Figure 8. Reaction examples in ultrasound irradiation

Guzen et al. (2006) explained that the condensation reactions of o-phenylenediamines with a diketone or a series of ketones occurred by ultrasound irradiation in the presence of APTS with good to excellent yields without catalyst (Figure 9).

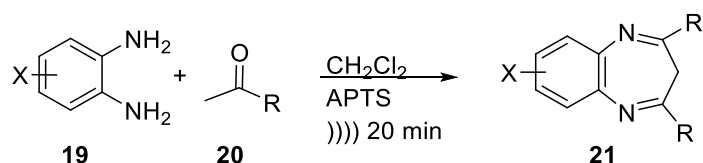


Figure 9. The synthesis of benzodiazepines **21**

Hickenboth et al. succeeded in the synthesis of molecule **24** by sonication (Figure 10) (2007).

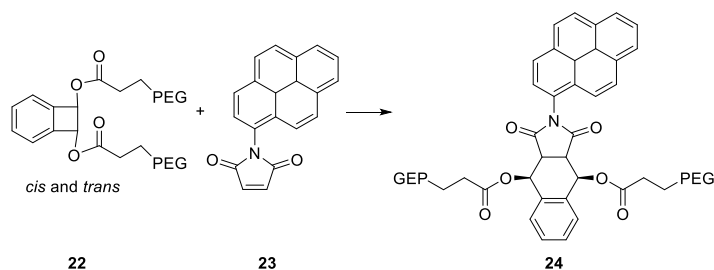


Figure 10. Ring-opening reaction of benzocyclobutene mechanophore (**22**)

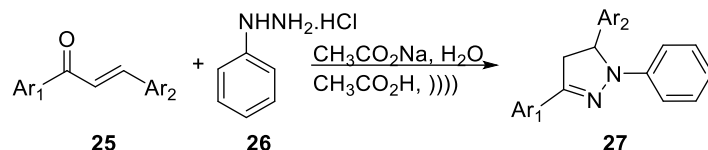


Figure 11. Synthesis of Pyrazoline Derivatives

1,3,5-triaryl-2-pyrazolines **27** were synthesized with chalcones **25** and phenylhydrazine hydrochloride **26** under ultrasound irradiation in 83-96% yields (Figure 11) (Li et al., 2007).

Anna et al. (2009) synthesized 2-substituted-2-imidazolines **30** from the substituted aldehydes and ethylenediamine by ultrasound irradiation with NBS in an aqueous medium (Figure 12).

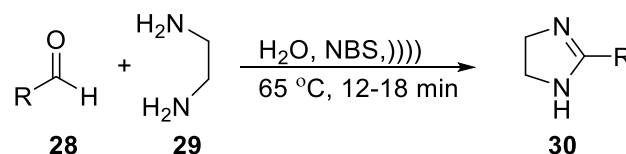


Figure 12. Synthesis of 2-Imidazolines (**30**)

2,4,5-Triarylimidazoles **32** were synthesized by the one-pot three-component condensation of benzil/benzoin **31**, aldehydes, and ammonium acetate in the presence of the catalytic amount of ceric (IV) ammonium nitrate under ultrasound (Figure 13) (Shelke, et al., 2009).

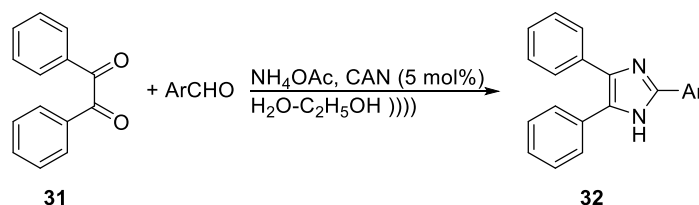


Figure 13. Synthesis of 2,4,5-triarylimidazole derivatives (**32**)

Jin et al. (2009) studied the epoxidation of chalcones **33**

with urea-hydrogen peroxide (UHP) in an ultrasonic bath and obtained epoxide 34 with high yields (Figure 14).

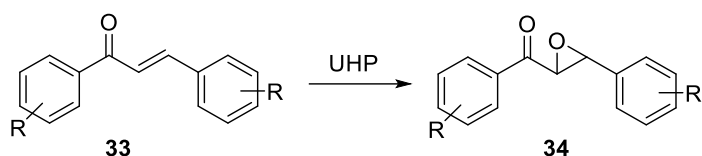


Figure 14. The epoxidation of chalcones

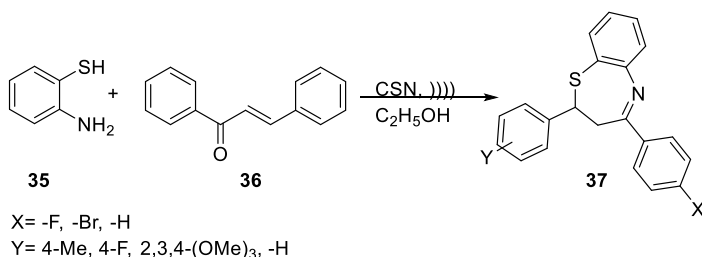


Figure 15. Synthesis of 1,5-benzothiazepines 37 from chalcones 36 and o-aminothiophenol 35

Chate et al. (2011) found an easy method for the preparation of 1,5-benzothiazepine derivatives in the presence of 10 mol% catalysts of CAN under ultrasonic irradiation (Figure 15).

Ultrasound is used to destroy toxic organic molecules using advanced oxidation processes (AOPs). Thus, it can control pollution and environmental protection. Bremner et al. (2011) used ultrasound at different frequencies for the oxidation of polycyclic aromatic hydrocarbons and phenol and examined the effect of ultrasound on degradation.

Zou et al. (2011) developed a method for preparing dihydropyrano[2,3-c]pyrazoles via a four-component reaction of aromatic aldehydes, hydrazine, ethyl acetoacetate, and malononitrile under ultrasound irradiation in water.

The Michael addition reaction of some active methylene compounds 39 to cyclohexenone 38 was achieved in high yields under ultrasonic irradiation, without using solvents (Figure 16) (Oge et al., 2012).

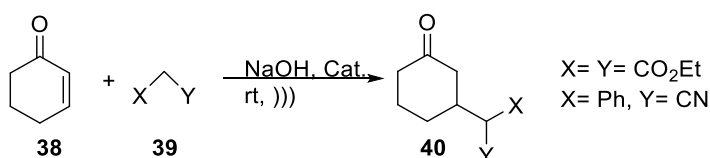


Figure 16. The Michael addition reaction to cyclohexenone 38

IKK-b inhibitors, A2A adenosine receptor antagonists and potent HIV-1 integrase inhibitors (Safari et al., 2012) and 2-amino-pyridine derivatives 44 identified as anti-tuberculosis agents (Chunxia 2013), are potential derivatives for the treatment of prion disease (Figure 17) (Guo et al., 2008).

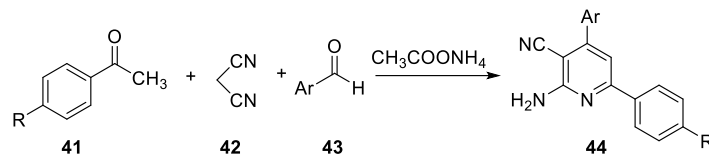


Figure 17. The synthesis of 2-amino-pyridine derivative 44

Eftekhari-Sis and Vahdati-Khajeh (2013) published a three-component, catalyst-free swift, and efficient procedure for the synthesis of pyridazine (26) and pyrrole (29) derivatives using water as a solvent and under ultrasonic irradiation (Figure 18).

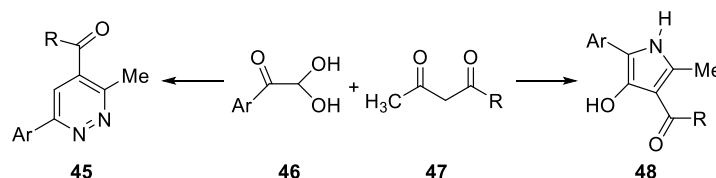


Figure 18. The synthesis of pyridazine 45 and pyrrole 48 derivatives

Mady et al. (2013) carried out the synthesis of homoallylic alcohols (50, 51) with ultrasonic support in high yields (Figure 19).

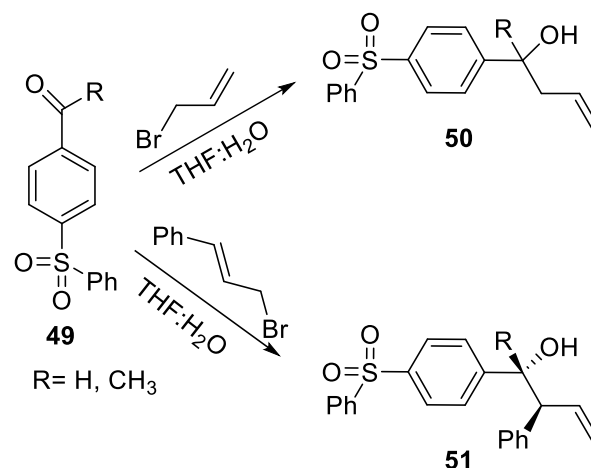


Figure 19. The synthesis of homoallylic alcohols (50, 51)

Cravotto et al. (2015) tried a series of reactions such as oxidation, bromination, aza-Michael, C-C couplings, MCR,

and aldol reactions in water under sonochemical conditions, thus showing that organic synthesis can be successful under sonochemical conditions in green chemistry.

Silva et al. (2016) explained the preparation of isatin derivatives 53 using 5-azido-spiro[1,3-dioxolane-2,3'-indol]-2'(1'H)-one (52) in the presence of various alkynes under acidic conditions and ultrasound irradiation (Figure 20).

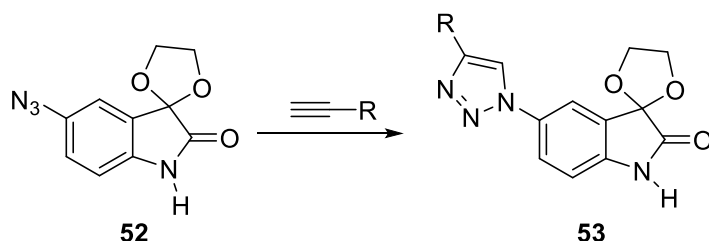


Figure 20. The synthesis of isatin derivatives 53

Zhang (2016) explained that sonochemistry can be used in the chemical and chemical industries in 2016. Draye et al presented a review of the recent improvements in sonochemical and combined sonochemical oxidation processes.

Crawford succeeded in the synthesis of molecules 54 and 55 in an ultrasound bath with high yields in 2017 (Figure 21).

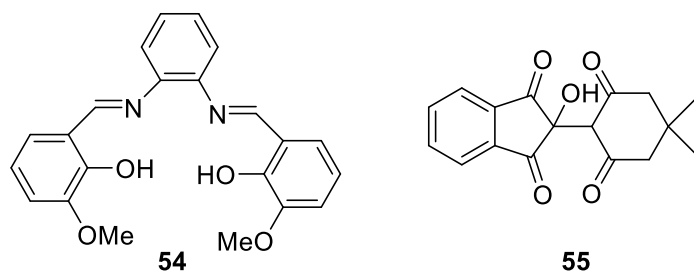


Figure 21. The synthesis of diamine 54 and aldol product 55

Sharda et al. (2018) also studied the condensation reactions of o-phenylenediamines 56 and synthesized 2,3-dihydro-1H-1,5-benzodiazepines 58 using a catalyst in 2018 (Figure 22).

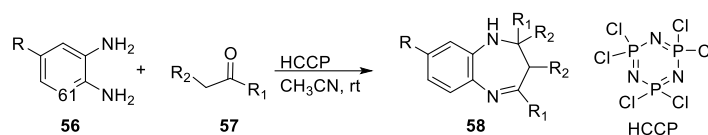


Figure 22. The condensation reactions of o-phenylenediamines 56

Navjeet explained the synthesis of nitrogen containing five-membered heterocycles under ultrasonic irradiation in 2019.

In 2020, Mallakpour and Azadi presented a sonochemical method for organo-synthesis, organo-modifications, and preparation of hybrids of titanium dioxide (TiO₂), which can be used in a wide variety of fields such as photocatalysis, antibacterial agents, present, self-cleaning.

Gharat et al. (2020) studied the application of sonochemistry in commercial industries such as food, pharmaceuticals, cosmetics, and chemicals and designed the reactor accordingly and stated that the ultrasonic system is more suitable in terms of cost-effectiveness, high efficiency, low waste, and low energy (2020). Sonochemistry is also used successfully in polymer synthesis. Hatami et al. are interested in ultrasound-assisted emulsion polymerization, mass and suspension polymerization, ring-opening polymerization, preparation of hydrogel polymers, synthesis of copolymers, phase transfer catalysis polymerization, reversible addition-fragmentation chain transfer (RAFT) procedure, polymerization (ATRP) and preparation of polymer nanocomposites were accomplished under ultrasonic conditions (2020). Metal-supported work can be done with ultrasonication. Since ultrasonic irradiation affects the metal surface, it also changes the mechanism, speed, and duration of the reaction. Bhuyan et al. tried ultrasonic conditions for various coupling reactions and were successful (Figure 23).

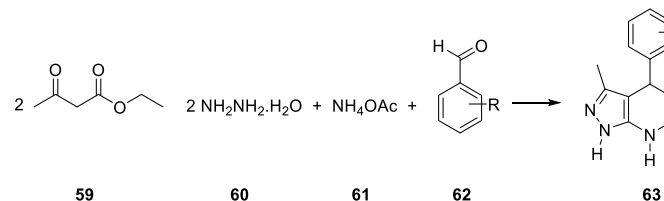


Figure 23. The synthesis of pyrazole 63

Tran and Nguyen have worked on alkylation reactions and used sonochemistry reactions, an environmentally friendly method, to improve reaction rates, product yield,

and selectivity and to find new synthetic routes in 2020. Blanco et al. (2020) synthesized 1,3-dibenzyl-1H-4,5,6,7-tetrahydro-1,3-diazepinium and 1,4,5,6,7,8-hexahydro-1,3-diazepinium salts under ultrasonic conditions with high yields (Figure 24).

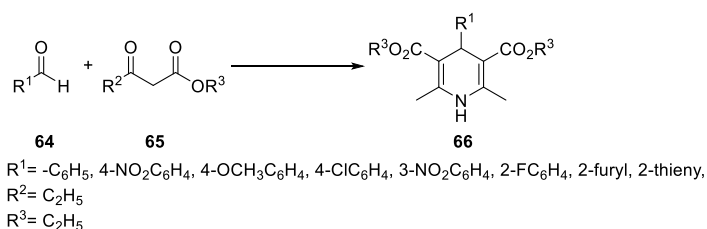


Figure 24. The synthesis of diazepinium 66

Draye et al. (2020) presented a review on the application of ultrasound in green chemistry in 2020. In 2021, the semisynthetic reaction of enantiopure Milbemycin- β (68) is successful with sonication in the first step but fails without sonication (Figure 25).

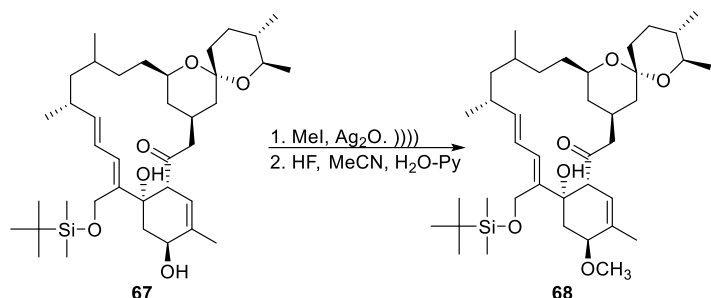


Figure 25. The synthesis Milbemycin- β (68)

Machado explained the synthesis of N-heterocyclic compounds 69 and 70 that are pharmacologically important using green solvents and catalysts in the ultrasonic bath in 2021 (Figure 26).

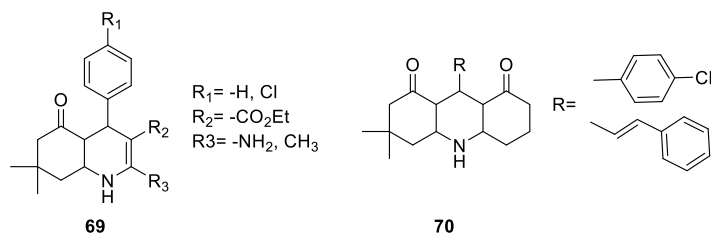


Figure 26. The synthesis of N-heterocyclic compounds

Casey et al. (2022) have written a review on the use of ultrasound to measure skeletal muscle. Shabir et al. (2022) presented the synthesis of 5-7 membered heterocyclic rings in sonochemistry with high yields.

Kamble et al. (2022) developed an environmentally friendly synthesis method for bioactive molecules (71-74) containing nitrogen and sulfur using ultrasonic energy in a short time (Figure 27).

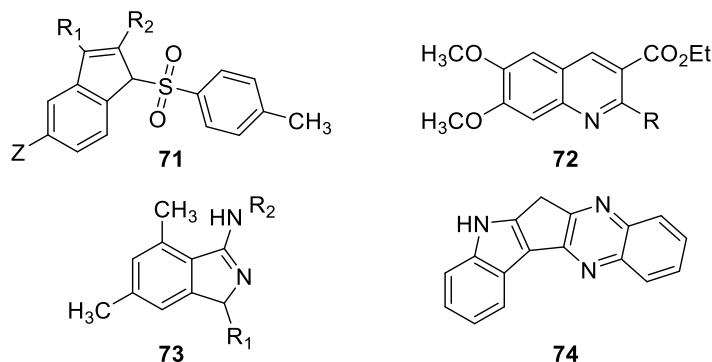


Figure 27. Synthesized bioactive molecules 71-74 using ultrasonic energy

In 2023 Sezen et al. studied the addition reaction of dimethyl cyclohexa-1,4-diene-1,2-dicarboxylate (46) and dichloroketene under ultrasonic and non-ultrasonic conditions. They showed that the ultrasonic reaction is faster than the non-ultrasonic reaction (Figure 28).

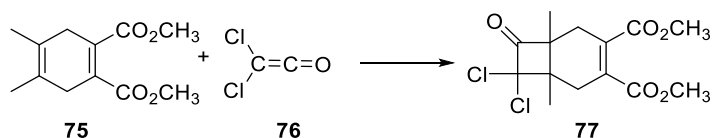


Figure 28. The ketene addition reaction of dimethyl cyclohexa-1,4-diene-1,2-dicarboxylate 77

Barmin et al. (2023) explained new perspectives on ultrasound-responsive polymeric designs, envisaging their current and future applications in ultrasound imaging and therapy. Abdelmonsef et al. (2023) found that ultrasound-assisted green synthesis of triazole analogs is a promising class of molecular entities for the development of new anticancer therapies, through targeting of some Rab proteins (Figure 29).

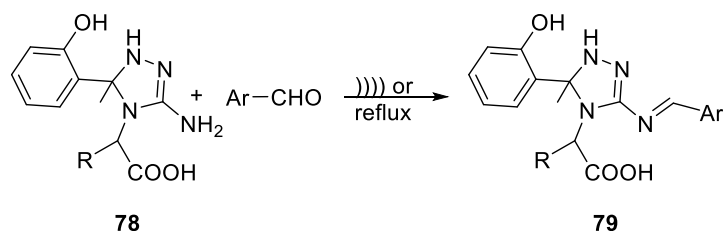


Figure 29. The synthesis of triazole analogs

In 2024, Jiao et al. (2024) wrote a review on ultrasound-

assisted sonochemical synthesis and biological applications, to promote the next generation of ultrasound technology-assisted applications.

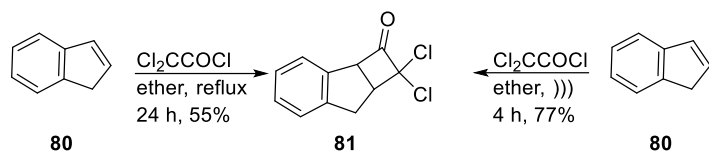


Figure 29. The synthesis of dichloro ketone 81.

Gökçay Bilici et al. (2024) synthesized dichloro ketone 81 with the addition reaction dichloroketene to indene with high yields under ultrasonic conditions.

Conclusion

The article gives short but detailed information about sonochemistry with various applications in the medicinal-pharmaceutical-drug-chemistry industries, and imaging techniques. While information about sonochemistry is given in the article, developments, and syntheses year by year since 1927 are mentioned. Much progress has been made regarding ultrasound in chemical science and the chemical industry in recent years. This paper focuses on using ultrasonic technologies in some areas such as polymer degradation, polymerization reactions, removal of toxic organic contaminants in water, organic synthesis, ultra-strong transfer processes including the extraction process, adsorption process, membrane process, demulsification, crystallization process, emulsification, heterogeneous chemical reaction process, and electrochemical process.

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