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Heterocycles via Cycloaddition underUltrasound Irradiation: A Greener feature

Pravin Chavan,[1] Amol Ghoti,[1] Shivaji Jadhav[2*], Rashmi Pathrikar[3], and Megha Rai[4]

[1]Department of Chemistry, Doshi Vakil College, Goregaon-Raigad, MS, India
[2]Department of Chemistry, Tarai College, Paithan-Aurangabad, MS, India
[3]Department of Chemistry, Rajshri Shahu College Pathri, Aurangabad, MS,India
[4]Department of Chemistry, Dr. Rafiq Zakaria College for Women, Aurangabad, MS,India
**Corresponding author: Email:dr.shivaji1985@gmail.com*
[1]Email: gmmalik2010@gmail.com*
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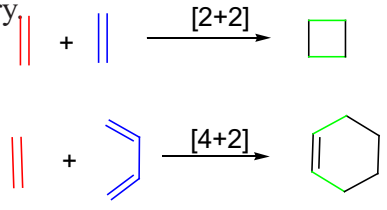
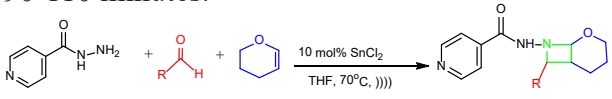
The big question facing researchers and manufacturers is how to practice synthetic organic chemistry with minimum impact on the natural environment.Major challenges include handling waste, designing eco-friendly procedures, and the preservation and efficient reuse of limited resources. It is necessary, with considerable urgency, to move to greener synthesis,[1-3,] and cycloaddition reactions[4-5] such as those sketched in Scheme 1, offer many advantages in the greening of synthetic organic chemistry.



Scheme 1.2π+2π and 4π+2π cycloadditions

These advantages include the simplicity of a single-step reaction, the specificity of the process which leads to few or no by-products, and the amenability of cycloaddition to the use of green solvents and catalysts. The purpose of this brief review is to discuss recent advances in a synthesis that have used cycloaddition reactions promoted by ultrasonic irradiation, with an effort to demonstrate the broad diversity of structures, many of them heterocycles, that can be achieved using this approach.

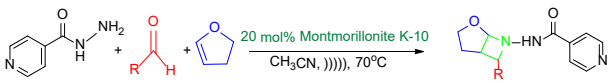
Chavan and co-workers[6]have reported the synthesis of substituted azetidines through a (2π+2π) cycloaddition route (Scheme 2). In the example shown, the intermediate imine formed by condensation of isoniazid (isonicotinic acid hydrazide)andanaromaticaldehydeimmediately cyclizes with dihydropyran stannous chloride as a catalyst under ultrasonic irradiation at 70°C. These reactions have been carried out within 90-110 minutes.



Scheme 2.Synthesis of azetidines

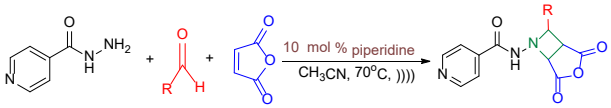
The azetidines showed antibacterial, antituberculosis, and anti-inflammatory activities.

Chavan and colleagues[7] have also reported the green one-pot multicomponent reaction for the synthesis of azetidines shown in Scheme 3. The initial formation of the dienophile imine is followed by a(2π+2π) cyclization with dihydrofuran using a clay catalyst and ultrasonication.



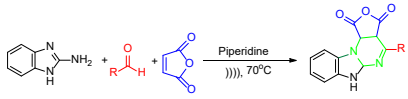
Scheme 3.Synthesis of azetidines from multicomponent cyclization of dihydrofuran with isoniazid imines

The derivatives prepared in this way showed both antibacterial and anti-inflammatory activity.Chavan and co-workers[8]further used this multicomponent approach with isoniazid, aromatic aldehydes, and maleic anhydride in the presence of a piperidine catalyst and ultrasound at 70°C (Scheme 4). The process was notable for high yields and pure products. The latter demonstrated anti-inflammatory and antibacterial properties.



Scheme 4.Synthesis of azetidine derivatives from maleic anhydride

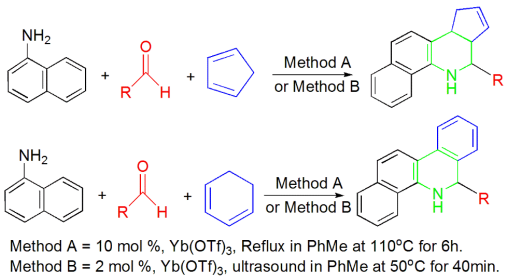
Chavan and collaborators[9]further investigated this method in the preparation of the extended heterocyclic anhydrides shown in Scheme 5, again with useful biological potential.



Scheme 5.Synthesis of substituted heterocyclic anhydrides

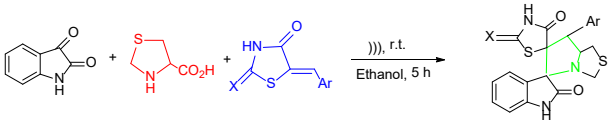
Pelit and co-workers[10] studied the preparation of heterocyclic derivatives using lanthanide triflate catalysis (Scheme 6). In this work, they reported the one-pot three-component ytterbium (III) triflate catalyzed aza-Diels-Alder (ADA, Povarov) reaction under ultrasonic conditions. There had been only limited examples of tetrahydroquinoline and hexahydro-phenanthridine derivatives synthesized by ADA reactions of cyclopentadiene and 1,3-cyclohexadiene.

In Method A, the reaction was explored by refluxing a mixture of 1-naphthylamine, various aromatic aldehydes,and cyclopentadiene/ cyclohexadiene with 10 mol% of ytterbium (III) triflate at 110°C in CH_3CN for 6 hours.In Method B,the reaction was explored by ultrasonic method, a mixture of 1-naphthylamine, various aromatic aldehydes,and cyclopentadiene/ cyclohexadiene with 2 mol% of ytterbium (III) triflate at 50°C in CH_3CN for 40 min. Under both methods, significant products were formed.



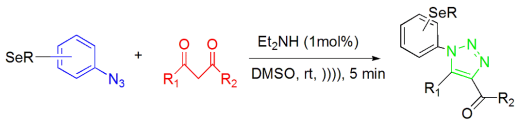
Scheme 6.Synthesis of tetrahydroquinoline and hexahydroquinoline derivatives from cyclopentadiene and 1,3 cyclohexadiene respectively

Hu and colleagues[11] have successfully explored the 1,3-dipolar cycloaddition of azomethine ylides under ultrasonic irradiation (Scheme 7). The unusual dispirooxindolecyclo[pyrrolo[1,2-c]thiazole-6,7-thiazolidine] derivatives were prepared from isatin and thiazolidine-4-carboxylic acid and 5-benzylidene-2-thioxothiazolidin-4-ones in ethanol.



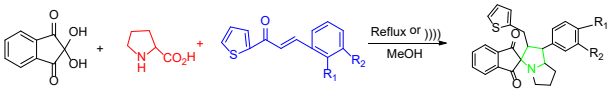
Scheme 7.Synthesis of unique dispirooxindolecyclo[pyrrolo[1,2-c]thiazole-6,7-thiazolidine] derivatives

Costaet al.[12]have described the use of sonochemistry for the organo catalytic enamine-azide (3+2) cycloaddition between 1,3-diketones and aryl azidophenyl selenides (Scheme 8). Good to excellent yields of the selenium-containing 1,2,3-triazole compounds were obtained in less time than conventional methods, due to the sonochemical effect.



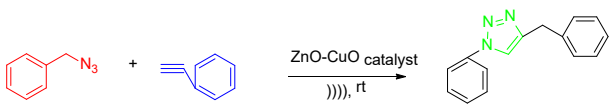
Scheme 8.Organocatalytic enamine-azide (3+2) cycloaddition between 1,3-diketones and aryl azidophenyl selenides

Chandralekha and co-workers[13]have prepared a new series of thiophene-based heterocycles via (3+2) cycloaddition of azomethine ylides with unusual thiophene dipolarophiles, trying both ultrasonic and irradiation and conventional heating (Scheme 9). The product yields from the ultrasonic method were higher than those from the conventional method.



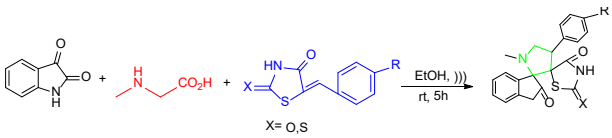
Scheme 9.Synthesis ofthiophene-based in heterocycles

Park and research partners[14]have described ZnO-CuO-nano catalyst and monitored its activity on the (3+2) cycloaddition reaction of azides and alkynes under ultrasound irradiations (Scheme 10). The catalyst is beneficial because of its reusability.



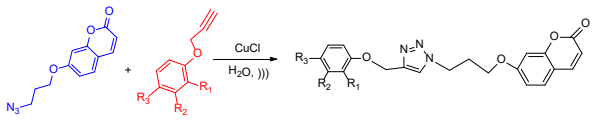
Scheme 10.Synthesis of(3+2) cycloaddition reaction ofazides and alkynes using ZnO-CuO nano-catalyst

Liu and co-authors[15] have developed three-component reactions of isatin, sarcosine, and 5-arylidene-1,3-thiazolidine-2,4-diones or 5-arylidene-4-thioxo-1,3-thiazolidine-2-ones for the synthesis of dispiropyrrolidine derivatives in ethanol under ultrasonic irradiation (Scheme 11). The study suggested that the ultrasonic irradiation technique is superior to the previous conventional technique.



Scheme 11. Synthesis of dispiropyrrolidine derivatives

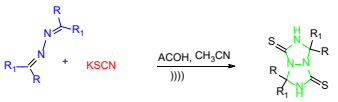
Li and colleagues[16] have described a new method (1,3-dipolar cycloaddition reaction) for the synthesis of 1,4-disubstituted 1,2,3-triazoles from different azides and various terminal alkynes under ultrasonic irradiation in water (Scheme 12). The modern CuAAC reaction features an enormous reaction rate acceleration in water compared with the same reaction in an organic solvent at room temperature.



Scheme 12. Synthesis of 1,4-disubstituted 1,2,3-triazoles

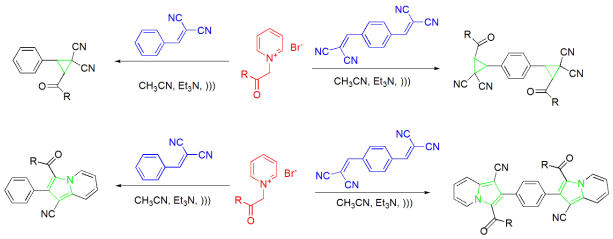
The Safari group[17] prepared perhydrotriazolotriazoledithiones from two successive 1,3-dipolar cycloadditions of azine derivatives and potassium isothiocyanate under ultrasonic irradiation (Scheme 13). This protocol provides a simple, very fast, and

low-cost procedure for the synthesis of these products.



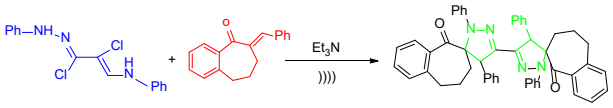
Scheme 13.Synthesis of perhydrotriazolotriazoledithiones

Abaszadeh and co-workers[18] have synthesized indolizine and bis-indolizine derivatives from 1,3-dipolar cycloaddition reactions of 2-chloropyridinium ylides with 2—benzylidene malononitrile or 2,2'-(1,4-phenylene bis(methanylylidene))dimalononitrile in acetonitrile under ultrasonic irradiation (Scheme 14).The developed protocols offer several advantages, such as high yields, simplicity, mild reaction conditions, and a very straightforward product separation process.



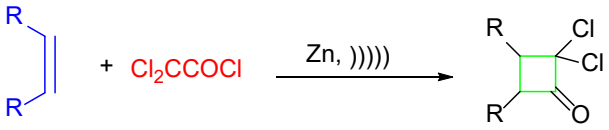
Scheme 14.a.Synthesis of cyclopropane and bis-cyclopropane derivatives
b. Formation of indolizine and bis-indolizine derivatives

Behbehani and co-workers[19] reported that the one-pot 1,3-dipolar cycloaddition reaction of bis-hydrazonoyl chlorides with 2-arylidene-1-benzosuberone derivatives afforded the corresponding bis-[1,4-diaryl-1-oxo-spiro-benzosuberane-2,5-pyrazoline] compounds under ultrasonic irradiation techniques (Scheme 15). Ultrasonic irradiation proved to be superior for the promotion of these reactions using ethanol and triethylamine as a base.



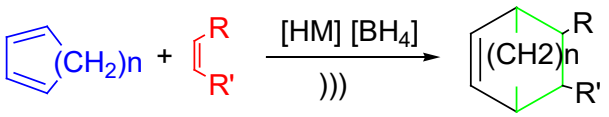
Scheme 15.Synthesis of bis-[1,4-diphenyl-1-oxo-spiro-benzosuberane-2,5-pyrazolines]

Mehta and colleagues[20]developed the ultrasound promoted 2+2 cycloaddition reaction of olefins with dichloroketene to yield substituted dichlorocyclobutanone (Scheme 16). This method had such significant aspects as short reaction times and good yields.



Scheme 16.Synthesis of substituted cyclobutanones

Bravo and research associates[21]reported a series of cycloadditions involving cyclopentadiene or 1,3-cyclohexadiene with carbonyl dienophiles in an imidazolium-based ionic liquid as a reaction medium (Scheme 17) to prepare the bicyclic systems. Compared to conventional methods, shorter reaction times and higher yields were generally observed; however, stereoselectivities were essentially the same.



Scheme 17.Sonochemical cycloadditions in ionic liquid

This brief review article summarizes recent studies on the use of cycloaddition reactions under ultrasonic irradiation in the synthesis of heterocyclic compounds. Advantages of this technique include clean reaction profiles, cost-effectiveness, one hundred percent atom economy, and mild reaction conditions. Given

these benefits, it seems likely that ultrasonic irradiation will continue to be explored for the preparation of heterocyclic compounds via cycloaddition reactions.

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Conflict of Interest: The authors have no conflicts of interest regarding this investigation.

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