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Ultrasonics in isocyanide-based multicomponent reactions: A new, efficient and fast method for the synthesis of fully substituted 1,3,4-oxadiazole derivatives under ultrasound irradiation

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ABSTRACT

A fast and convenient approach to the synthesis of fully substituted 1,3,4-oxadiazoles via three-component reaction of aromatic carboxylic acids, acenaphthoquinone, and (*N*-isocyanimino)triphenylphosphorane under ultrasound irradiation is described. Furthermore, a series of compounds were synthesized and characterized by melting point, IR, NMR and MS. Utilization of easy reaction conditions, very high to excellent yields, and short reaction times makes this manipulation potentially very useful.

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1. Introduction

Multicomponent reactions (MCRs) [1,2] are referred to as the one-pot processes, where multiple bonds are formed among starting materials to furnish the product with essentially all of the atoms of the reactants. Among the known multicomponent reactions, isocyanide based MCRs (IMCRs) are particularly valuable [3,4]. In addition to the added diversity of bond formation and functional group tolerance, the outstanding position of IMCRs can also be traced back to the exceptional reactivity of isocyanide. As we know, no other functional group reacts with nucleophiles and electrophiles at the same atom [5]. Consequently, MCRs involving isocyanides have been widely applied to organic synthesis, especially in drug discovery [6]. The *aza*-Wittig-type reaction has attracted much attention recently because of its high potential for the synthesis of a wide variety of nitrogen heterocycles, which can be attributed in the preparation of functionalized iminophosphoranes [7]. Through *aza*-Wittig reaction, iminophosphoranes with isocyanates, carbon dioxide, or carbon disulfide can easily be converted into functionalized hetero-cumulenes [7]. The nucleophilicity at the nitrogen is an important factor in the use of these iminophosphoranes as *aza*-Wittig reagents. Iminophosphoranes are important reagents in organic chemistry, especially in the producing of naturally occurring products, compounds with biological and pharmacological activity [7,8]. In recent years, we have confirmed a one-pot method for the preparation of organophosphorus compounds [9–15].

1,3,4-Oxadiazoles have attracted interest in medicinal chemistry as surrogates of carboxylic acids, esters, and carboxamides. They are an important class of heterocyclic compounds that have a wide range of pharmaceutical and biological activities including antimicrobial, antifungal, antiinflammatory, and antihypertensive [16–20]. Several methods have been reported in the literature for the synthesis of 1,3,4-oxadiazoles. These protocols are multistep in nature [21–26]. The most general method involves the cyclization of diacylhydrazides with a variety of reagents, such as thionyl chloride, phosphorus oxychloride, or sulfuric acid, usually under harsh reaction conditions. Few reliable and operationally simple examples have been reported for the one-step synthesis of 1,3,4-oxadiazoles, especially from readily available carboxylic acids and acid hydrazides [27–32].

Ultrasound irradiation, an efficient and innocuous technique for reagent activation in the synthesis of inorganic compounds [33–38], organic compounds and in particular heterocyclic

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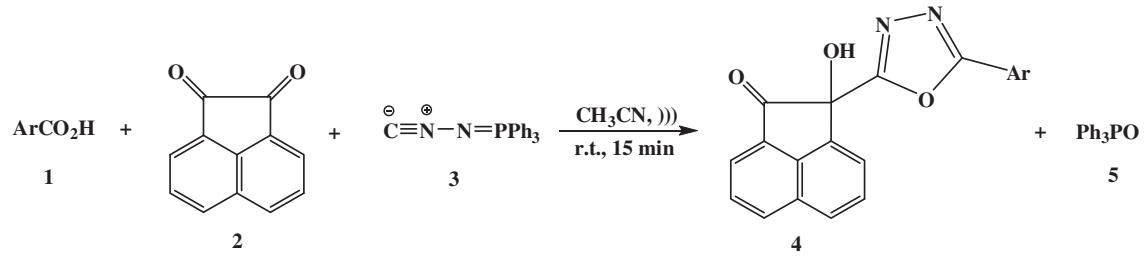

Scheme 1. Synthesis of disubstituted 1,3,4-oxadiazole derivatives under ultrasound irradiation.

compounds, has been applied with success, milder reaction condition, and higher yields in comparison to the classical methods [39–41]. Ultrasound-promoted synthesis has attracted much attention during the past few decades. One advantage of using cavitation as an energy source to promote organic reactions includes shorter reaction times [42]. During the rarefaction cycle in the cavitation process, the molecules of the liquid are separated, generating bubbles that subsequently collapse in the compression cycle. These rapid and violent implosions generate short-lived regions with temperatures of roughly 5000 °C, pressures of about 1000 atm and heating and cooling rates above 10 billion °C/s. Such localized hot spots can be thought as micro reactors in which the energy of sound is transformed into a useful chemical form [42,45,46]. This procedure has been considered as a clean and useful protocol in organic synthesis compared with traditional methods, and the procedure is in general, more convenient [42].

There are very few reports in literature about application of ultrasound irradiation in isocyanide-based multicomponent reactions. In continuation of our research program to find the effect of ultrasound irradiation on isocyanide-based multicomponent reactions [43,44], we wish to report a novel and efficient method for the synthesis of 1,3,4-oxadiazole derivatives (**4**) via an efficient condensation of aromatic carboxylic acids, acenaphthoquinone, and (*N*-isocyanimino)triphenylphosphorane under ultrasound irradiation (Scheme 1).

2. Experimental

2.1. Apparatus, materials and measurements

(*N*-Isocyanimino)triphenylphosphorane (**3**) was prepared based on reported procedures [8,47]. Other starting materials and solvents were purchased from Merck (Germany) and Fluka (Switzerland) and were used without further purification. The methods used to monitor the reactions are TLC and NMR. Melting points were determined on an Electrothermal 9100 apparatus. IR spectra (ν_{max}, cm^{-1}) were recorded on a Jasco 6300 FTIR spectrophotometer using KBr technique. ^1H and ^{13}C NMR spectra ($CDCl_3$) were recorded on a Bruker DRX-250 Avance spectrometer at 250.0 and 62.5 MHz, respectively. Mass analyses were carried out using a Finnigan MAT-8430 mass spectrometer operating at an ionization potential of 70 eV. Preparative layer chromatography (PLC) plates were prepared from Merck silica gel (F_{254}) powder. Sonication was performed in a Bandelin SONOPULS ultrasonic homogenizers (made in Germany) with 20 kHz processing frequency, a nominal power 250 W, uniform sonic waves.

2.2. General procedure for the synthesis of the title compounds in CH_3CN

A mixture of (*N*-isocyanimino)triphenylphosphorane (302 mg, 1.0 mmol), acenaphthoquinone (182 mg, 1.0 mmol), and aromatic carboxylic acid (1.0 mmol) in CH_3CN (10 mL) was stirred at for 24 h in room temperature. The solvent was removed under reduced pressure, and the viscous residue was purified by PLC [silica gel (F_{254}) powder; petroleum ether/ethyl acetate 4:1].

2.3. Ultrasound-promoted typical procedure for synthesis of title compounds

The carboxylic acid derivatives (1.0 mmol), acenaphthoquinone (182 mg, 1.0 mmol), (*N* isocyanimino)triphenylphosphorane (302 mg, 1.0 mmol) and CH_3CN (10 mL) were added into a 25 mL round bottomed flask. During the ultrasound irradiation, the temperature of the mixture was controlled with ice bath (temperature was maintained in room temperature). The reaction mixture was sonicated under 100 W for the period of time (The reaction was monitored by TLC). The solvent was removed under reduced pressure, and the viscous residue was purified by PLC [silica gel (F_{254}) powder; petroleum ether/ethyl acetate 4:1]. The authenticity of the samples (**4a**–**4p**) was established by their ^1H NMR, ^{13}C NMR and MS.

2.4. Data spectra of products

2.4.1. Compound **4a**

*2.4.1.1. 2-Hydroxy-2-(5-phenyl-1,3,4-oxadiazol-2-yl)-1(2H)-acenaphthylenone (**4a**, $C_{20}H_{12}N_2O_3$).* Yellow powder; yield 80%; R_f = 0.36 (petroleum ether/ethyl acetate 4:1); m.p.: 162–164 °C; ^1H NMR (250.13 MHz, $CDCl_3$): *d* = 4.59 (br s, OH), 7.40–8.30 (m, 11CHarom) ppm; ^{13}C NMR (62.53 MHz, $CDCl_3$): *d* = 87.74 (C–OH), 122.56, 123.88, 126.97, 127.13, 128.79, 128.96, 129.07, 132.02, 132.85 (11CH of arom), 123.22, 127.80, 129.60, 135.12, 142.96 (5C of arom), 161.05, 166.13 (2C=N of oxadiazole), 198.51 (C=O) ppm; IR (KBr): ν_{max} = 3366, 3073, 1718, 1603, 1448, 1012 cm^{-1}; MS (EI, 20 eV): m/z (%) = 328 (M^+), 198 (16), 182 (34), 154 (83), 126 (100), 98 (28), 85 (28), 76 (39), 62 (38), 43 (45).

2.4.2. Compound **4b**

*2.4.2.1. 2-[5-(4-Bromophenyl)-1,3,4-oxadiazol-2-yl]-2-hydroxy-1(2H)-acenaphthylenone (**4b**, $C_{20}H_{11}BrN_2O_3$).* Yellow powder; yield 78%; R_f = 0.33 (petroleum ether/ethyl acetate 4:1); m.p.: 151–153 °C; ^1H NMR (250.13 MHz, $CDCl_3$): *d* = 4.57 (br s, OH), 7.57 (*d*, ^3JHH = 8.3 Hz, 2CHarom), 8.03 (*d*, ^3JHH = 8.3 Hz, 2CHarom), 7.69–7.86, 8.09–8.30 (m, 6CHarom) ppm; ^{13}C NMR (62.53 MHz, $CDCl_3$): *d* = 86.50 (C–OH), 122.59, 123.96, 127.03, 128.51, 128.83, 129.08, 132.33, 132.94 (10CH of arom), 122.12, 126.80, 129.50, 130.98, 134.22, 140.25 (6C of arom), 164.02, 165.03 (2C=N of oxadiazole), 202.02 (C=O) ppm; IR (KBr): ν_{max} = 3204, 3077, 1733, 1600, 1481, 1007 cm^{-1}; MS (EI, 20 eV): m/z (%) = 407 (M^+), 198 (8), 182 (37), 154 (78), 126 (100), 98 (22), 85 (22), 75 (55), 62 (45), 50 (18).

2.4.3. Compound **4c**

*2.4.3.1. 4-[5-(1,2-Dihydro-1-hydroxy-2-oxo-1-acenaphthylenyl)-1,3,4-oxadiazol-2-yl]benzonitrile (**4c**, $C_{21}H_{11}N_3O_3$).* Yellow powder; yield 73%; R_f = 0.30 (petroleum ether/ethyl acetate 4:1); m.p.: 161–163 °C;

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