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Microwave-assisted acetylation of alcohols, phenols, and amines using phthalimide-*N*-sulfonic acid as an organo-catalyst under solvent-free conditions

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Abstract: An efficient, environmentally benign, and simple procedure has been developed for acetylation of various alcohols, phenols and amines. In this method, Phthalimide-*N*-sulfonic acid is used as a new and effective organo-catalyst in catalytic amount. This procedure was carried out under solvent-free conditions at room temperature as well as at microwave irradiation. Microwave-assisted reaction gives improved yields with a reduced reaction time. The present method provides noteworthy advantages of microwave irradiation and organo-catalyst such as simple work-up procedure, short reaction time with excellent yields and environmentally benign procedure. All compounds have been confirmed by ^{1}H NMR, ^{13}C NMR and IR spectroscopy.

Keywords: Microwave; phthalimide-*N*-sulfonic acid; organo-catalyst; protection; acetylation; solvent-free.

1. Introduction

In recent years, use of microwave irradiation has gained considerable attention on account of their advantages such as rapid and environmentally benign synthesis of various heterocyclic compounds[1-4] as well as in the development of various green methodologies[4-8]. It provides an enhanced reaction rate and improved product yield due to selective absorption of microwave energy by different polar functional moieties present in the molecule.

The acetylation reaction is very important reaction in synthetic organic chemistry as well as in pharmaceutical synthesis[9-11]. This reaction is used for the protection of different functional groups like alcohols, phenols, and amines during the oxidation of other functional groups, as well as in peptide coupling or glycosylation during peptide synthesis.

Recently several improved methods have been developed for acetylation reaction, which involves various catalysts such as organo-catalysts[12-17], basic catalyst[18-20], heterogeneous Brønsted acids[21-29], heterogeneous[30-38] as well as homogeneous Lewis acids[39-51], enzyme catalysts[52-53] and ionic liquids[54-57]. Many of these reported procedures involve one or more limitations such as, use of expensive catalyst, harmful solvents, harsh reaction conditions, prolonged reaction times, dreary workup procedures or poor yields. Therefore, there is scope to developing efficient and environmentally benign green chemical methods remains a major challenge for researcher in organic syntheses.

Now days, use of sulfonic acid bearing solid organo-catalysts has received significant attention due to its highly selectivity in conversion of products, metal-free conditions, non-toxicity and easy isolation from reaction media for the synthesis of various organic and heterocyclic molecules[58-62].

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Amongst these organo-catalyst, Phthalimide-*N*-sulfonic acid is non-toxic, cost-effective, easy to preparation and highly stable to air as well as thermal conditions. This solid organo-catalyst is used in development of various green reaction methodologies[63-66].

As part of our ongoing research, to develop a novel methodology using alternative procedures[67-73] herein, we report, microwave-assisted acetylation of alcohols, phenols and amines using Phthalimide-*N*-sulfonic acid as organo-catalyst under solvent-free conditions.

2. Experimental

2.1. General Methods

All chemicals were purchased from commercial sources and used without purification. ^{1}H NMR spectra were recorded on Gemini-300 spectrometer in $CDCl_3$ using TMS as an internal standard and IR spectra were recorded on Shimadzu FT-IR 8000 series spectrophotometer using neat or KBr disk. Melting points were determined using Buchi R-535 apparatus and uncorrected. All Microwave reactions were carried out using Microwave Synthesizer (TOPLAB INDIA, Model TOP-205, 700 W). Phthalimide-*N*-sulfonic acid catalyst was prepared according to the literature[63].

2.2. General Procedure

A mixture of substrate (alcohols or phenols or amines) (1 mmol), acetic anhydride (1.2 mmol) and Phthalimide-*N*-sulfonic acid (2 mol %) as organo-catalyst was subjected in 25 ml flask to microwave irradiation at power level 240 W for appropriate time as shown in Table 1. After completion of reaction monitored by TLC using ethyl acetate / hexane (2:8), Ethyl acetate (5 ml) was added to the mixture and filtered, then the solid catalyst was again washed with ethyl acetate (5 ml) and collected for reuse. Crude product was concentrated under reduced pressure then purified by column chromatography using silica gel (60-120 mesh) with eluting ethyl acetate-hexane mixture (2:8). All the pure products were confirmed by their spectroscopy data and melting points, which were consistent with literature data.

2.3. Spectral Data of Reported Compounds

Benzyl acetate ***(3a)***: colorless oil (97 %); IR (KBr): ν_{max} = 3035, 2926, 2855, 1725, 1590, 1462, 1383, 1129 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.40-7.30 (m, 5H, Ar-H), 5.12 (s, 2H, Ar-CH_2), 2.10 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 171.0, 136.6, 128.7, 128.1, 127.5, 66.4, 21.0 ppm.

4-Methoxybenzyl acetate ***(3b)*****:** Colorless oil (97 %); IR (KBr): ν_{max} = 2928, 2856, 1742, 1621, 1505, 1253, 1120 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.38-7.30 (m, 2H, Ar-H), 6.88-6.84 (m, 2H, Ar-H), 5.05 (s, 2H, Ar-CH_2), 3.81 (s, 3H, -$COCH_3$), 2.02 (s, 3H, -CH_3) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 170.9, 159.3, 130.8, 128.1, 114.0, 66.1, 55.9, 21.1 ppm.

4-Chlorobenzyl acetate ***(3c)***: Colorless oil (95 %); IR (KBr): ν_{max} = 2927, 2855, 1739, 1608, 1502, 1384, 1130 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.40-7.36 (m, 2H, Ar-H), 7.33-7.27 (m, 2H, Ar-H), 5.06 (s, 2H, Ar-CH_2), 2.10 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 171.1, 134.7, 134.2, 129.7, 128.3, 65.9, 20.9 ppm.

Benzhydryl acetate ***(3d)***: Colorless oil (92 %); IR (KBr): ν_{max} = 3034, 2942, 2856, 1732, 1582, 1482, 1384, 1232, 1140 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.48-7.44 (m, 5H, Ar-H), 7.40-7.28 (m, 5H, Ar-H), 6.81 (s, 1H, $(Ar)_2$-CH), 2.14 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 170.9, 140.1, 129.1, 128.3, 127.2, 77.7, 21.5 ppm.

2-Oxo-1,2-diphenylethyl acetate ***(3e)*****:** Colorless oil (90 %); IR (KBr): ν_{max} = 3052, 2926, 2855, 2816, 1734, 1693, 1560, 1462, 1383, 1234 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.82 (d, 2H, *J* = 7.4 Hz, Ar-H), 7.66-7.60 (m, 2H, Ar-H), 7.50-7.38 (m, 4H, Ar-H), 7.34-7.26 (m, 2H, Ar-H), 6.84 (s, 1H, Ar-CH),

2.20 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 193.7, 170.8, 134.7, 133.8, 133.5, 129.3, 129.2, 128.8, 128.5, 128.3, 128.1 77.8, 46.8 36.5, 20.8 ppm.

Octyl acetate ***(3f)*****:** Colorless oil (98 %); IR (KBr): ν_{max} = 2926, 2855, 1740, 1436, 1384, 1130 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 4.04 (t, 2H, *J* = 7.6 Hz, -OCH_2), 2.08 (s, 3H, -$COCH_3$), 1.66-1.64 (m, 2H), 1.44-1.32 (m, 10H), 0.90 (t, 3H, J = 6.4 Hz) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 171.1, 64.4, 32.0, 31.7, 29.5, 29.4, 29.1, 28.5, 25.7, 22.5, 20.9, 13.9 ppm.

Cyclohexyl acetate ***(3g)*****:** Colorless oil (97 %); IR (KBr): ν_{max} = 2981, 1740, 1433, 1384, 1127 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 4.06-4.02 (m, 1H), 2.02 (s, 3H, -$COCH_3$), 1.82-1.76 (m, 4H), 1.62-1.52 (m, 6H) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 170.0, 72.2, 35.6, 31.7, 29.6, 25.0, 23.9, 22.5, 21.2 ppm.

Phenyl acetate ***(3h)*****:** Colorless oil (90 %); IR (KBr): ν_{max} = 3055, 2942, 2855, 1742, 1598, 1485, 1384, 1140 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.35-7.30 (m, 2H, Ar-H), δ 7.24-7.14 (m, 3H, Ar-H), δ 2.31 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.8, 150.5, 129.8, 126.1, 121.3, 21.2 ppm.

4-Methylphenyl acetate ***(3i)*** : Colorless oil (92 %); IR (KBr): ν_{max} = 2988, 2845, 1745, 1598, 1485, 1390, 1145 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.12-7.06 (m, 4H, Ar-H), 2.32 (s, 3H, -CH_3), 2.18 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.7, 148.7, 136.1, 129.5, 121.6, 22.6, 20.3 ppm

4-Methoxyphenyl acetate ***(3j)***: Yellow oil (92%); IR (KBr): ν_{max} = 2954, 2922, 2850, 1754, 1610, 1459, 1375, 1129 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ = 7.01-6.87 (m, 4H, Ar-H), 2.68 (s, 3H, -OCH_3), 2.2 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ = 169.7, 157.4, 143.2, 122.5, 113.8, 55.2, 20.2 ppm.

4-Nitrophenyl acetate ***(3k)*** : Faint Yellow solid (88 %); m.p. 78-80˚C (Lit[28] 76-80˚C); IR (KBr): ν_{max} = 3056, 2927, 2855, 1738, 1612, 1488, 1384, 1132 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 8.18 (d, 2H, *J* = 7.6 Hz, Ar-H), 7.43 (d, 2H, *J* = 7.6 Hz, Ar-H), 2.10 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 170.5, 156.3, 145.1, 122.6, 121.7, 20.4 ppm.

4-Formylphenyl acetate ***(3l)***: Colorless liquid (89 %); IR (KBr): ν_{max} = 2928, 2855, 2765, 1755, 1627, 1384, 1133 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 9.2 (s, 1H, Ar-CHO), 7.96 (d, 2H, *J* = 7.4 Hz, Ar-H), 7.16 (d, 2H, *J* = 7.4 Hz, Ar-H); 2.38 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 191.6, 169.0, 157.1, 133.2, 131.9, 131.0, 122.8, 122.1, 20.6 ppm.

Naphthalen-1-yl acetate ***(3m)***: White solid (92 %); m.p. 66-68 ˚C (Lit[21] 69-70˚C); IR (KBr): ν_{max} = 2935, 2865, 1742, 1598, 1488, 1372, 1194 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.84-7.74 (m, 2H, Ar-H), 7.58-7.46 (m, 3H, Ar-H), 7.35-7.27 (m, 2H, Ar-H), 2.45 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.2, 147.8, 133.4, 131.0, 129.8, 127.9, 127.3, 126.5, 125.1, 121.7, 118.8, 20.7 ppm.

Naphthalen-2-yl acetate ***(3n)***: White solid (90 %); m.p. 74-76˚C (Lit[28] 68-72˚C); IR (KBr): ν_{max} = 2937, 2862, 1748, 1672, 1437, 1384, 1152 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.88-7.84 (m, 3H, Ar-H), 7.62 (s, 1H, Ar-H), 7.48-7.46 (m, 1H, Ar-H), 7.36-7.32 (m, 1H, Ar-H), 7.16 (d, 1H, *J* = 7.8 Hz, Ar-H), 2.30 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.9, 148.7, 133.6, 131.0, 129.1, 127.9, 127.2, 126.2, 125.4, 120.4, 118.4, 21.0 ppm.

N-Benzyl-acetamide ***(3o)***: White solid (95 %); m.p. 60-62 ˚C, IR (KBr): ν_{max} = 3298, 2923, 2853, 1665, 1612, 1543, 1508, 1136 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.52-7.34 (m, 5H, Ar-H), 6.18 (brs, 1H, N-H), 4.00 (d, 2H, *J* = 2.2 Hz, Ar-CH_2), 2.04 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR in $CDCl_3$: δ 170.0, 137.1, 133.7, 128.6, 1127.6, 126.1, 43.8, 23.5ppm

N-(4-Methylbenzyl)-acetamide ***(3p)***: White solid (95 %); m.p. 150-152˚C; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.22-7.08 (m, 4H, Ar-H), 5.60 (s, 1H, N-H), 4.38 (d, 2H, *J* = 2.3 Hz, Ar-CH_2), 2.38 (s, 3H, -CH_3), 2.10 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR in $CDCl_3$: δ 169.4, 137.8134.9, 129.5, 127.7, 23.2, 21.3 ppm.

N-Phenyl-acetamide ***(3q)***: White solid (94 %); m.p. 112-114˚C; IR (KBr): ν_{max} = 3295, 3028, 1663, 1607, 1542, 1468, 1370, 1130 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.40 -7.30 (m, 5H, Ar-H), 7.05 (brs, 1H, N-H), δ 2.09 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.1, 137.8, 128.3, 124.4, 120.4, 24.4 ppm.

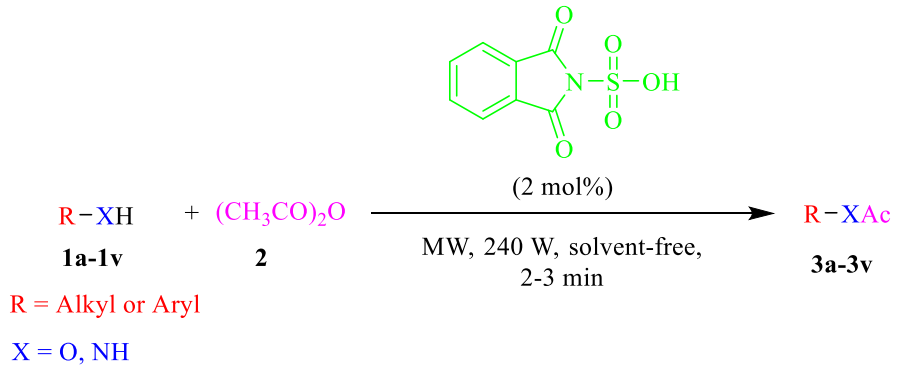
N-(4-Chlorophenyl)-acetamide ***(3r)***: White solid (91 %); m.p. 170-172˚C (Lit[28] 172-175˚C); IR (KBr): ν_{max} = 3303, 2919, 2851, 1778, 1601, 1537, 1247, 1126 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 8.06 (brs, 1H, N-H), 7.62 (d, 2H, *J* = 7.8 Hz, Ar-H), 7.24 (d, 2H, *J* = 7.8 Hz, Ar-H), 2.10 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.5, 137.9, 133.7, 128.9, 119.9, 23.6 ppm.

N-(4-acetylphenyl)-acetamide ***(3s)*****:** White solid (90 %); m.p. 168-170 ˚C; IR (KBr): ν_{max} = 3149, 2859, 1680, 1594, 1479, 1436, 1384, 1312, 1128 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.90 (d, 2H, *J* = 5.6 Hz, Ar-H), 7.74 (d, 2H, *J* = 2.4 Hz, Ar-H), 7.53 (brs, 1H, N-H), 2.59 (s, 3H, -$COCH_3$), 2.20 (s, 3H, Ar-$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.4, 137.8, 134.9, 129.5, 127.7, 121.6, 43.5, 23.2, 21.3 ppm

N-(4-Nitrophenyl)-acetamide ***(3t)***: White solid (89 %); m.p. 218-220˚C (Lit[28] 213-217˚C); ^{1}H-NMR (300 MHz, $CDCl_3$): δ 9.10 (s, 1H, N-H), 7.98 (d, 2H, *J* = 7.7 Hz, Ar-H), 7.80 (d, 2H, *J* = 7.7Hz, Ar-H), 2.12 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR in $CDCl_3$: δ 169.1, 145.4, 141.1, 124.6, 118.4, 24.4 ppm.

N-(Naphthalen-1-yl)-acetamide ***(3u)***: White solid (92 %); m.p. 134-136˚C (Lit[28] 127-129˚C); IR (KBr): ν_{max} = 3305, 2926, 2854, 1641, 1384, 1089 cm^{-1}; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 8.16 (brs, 1H, N-H), 7.94-7.76 (m, 2H, Ar-H), 7.60-7.46 (m, 3H, Ar-H), 7.38 (t, 1H, *J* = 7.2 Hz, Ar-H), 6.96 (d, 1H, *J* = 7.1 Hz, Ar-H), 2.06 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 169.3, 133.2, 129.0, 128.7, 127.5, 127.0, 126.0, 125.2, 122.8, 122.1, 118.8, 22.5 ppm.

N-(Naphthalen-2-yl)-acetamide ***(3v)***: Light yellow solid (92 %); m.p. 132-134˚C; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 8.14 (brs, 1H, N-H), 7.88-7.80 (m, 2H, Ar-H), 7.62 (s, 1H, Ar-H), 7.48-7.38 (m, 4H, Ar-H), 2.04 (s, 3H, -$COCH_3$) ppm; ^{13}C-NMR (75 MHz, $CDCl_3$): δ 168.8, 135.2, 133.8, 129.1, 127.2, 126.4, 121.2, 118.1, 116.5, 22.6 ppm.



Scheme 1. Microwave-assisted acetylation of alcohols, phenols and amines under solvent-free conditions.

3. Results and Discussion

In continuation our research interest in the development of green synthetic methodologies, we decided to explore the use of recyclable organo-catalyst under microwave irradiation at solvent-free conditions for the acetylation of alcohols, phenols and amines.

In a typical experimental procedure, acetylation of alcohol carried out using benzyl alcohol (1.0 mmol.), acetic anhydride (1.2 mmol.) and Phthalimide-*N*-Sulfonic acid (2 mol%) as catalyst under

microwave irradiation at solvent-free condition. The reaction was completed within 2 minutes, as shown in Scheme 1.

To optimize the reaction conditions and catalyst, initially, the reaction was carried out using catalyst-free and solvent-free conditions at room temperature for a prolonged time (180 minutes) but the poor yield (10 mol%) of product was obtained. After that, we chose mild, non-toxic and highly stable Phthalimide-*N*-Sulfonic acid as catalyst. The role of catalyst has been screened using different mole ratios (from 1-10 mol%) for 40-120 minutes under solvent-free condition. The observation shows that 10 mol% equivalent of catalyst is enough for the completion of the reaction (Table 1).

Table 1. Optimization of catalyst under different conditions

Entry	Catalyst (mol%)	Room Temperature		Microwave Irradiation	
		Time (min)	Yield (%)	Time (min)	Yield (%)
1	No Catalyst	180	10	20	30
2	1	120	30	10	53
3	1.5	90	46	5	78
4	2.0	60	64	**2**	**97**
5	2.5	60	73	2	97
6	5	60	79	2	97
7	10	**40**	**90**	2	98

Reaction conditions: 1.0 mmol benzyl alcohol, 1.2 equiv Ac_2O.

After that, we decided to investigate the influence of microwave irradiation on a reaction mixture in presence as well as in absence of the catalyst under solvent-free condition. Initially reaction was carried out in the absence of catalyst under microwave irradiation with 280 W power at solvent-free condition to obtained poor yield (30 %) for extended time (20 minutes). After that, activity of catalyst was screened by using different mole ratios (from 1-10 mol%) for 2-10 minutes. The observation shows that 2 mol% equivalent catalyst is sufficient for the completion of the reaction in high yield. It is observed that the use of microwave irradiation resulted in shorter reaction time with excellent yields than room temperature. All are results summarized in Table 1.

After the optimization of the catalysts and conditions, we moved towards the optimization of the different power levels of microwave irradiation. Initially, reaction was carried out at 140 W to obtain corresponding ester product with 70% yield in 10 minutes (Table 2).

Table 2. Optimization of different power levels of microwave for the acetylation of alcohols, phenols and amines

Entry	Power level in Watt	Time (min)	Yield[a] (%)
1	140	10	70
2	210	5	82
3	**240**	**2**	**97**
4	280	2	96
5	350	2	90
6	420	2	76

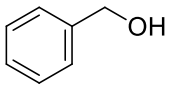
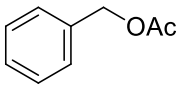
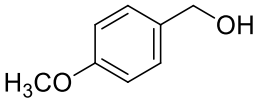
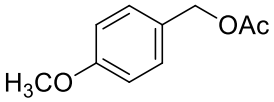
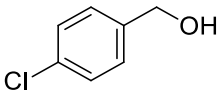
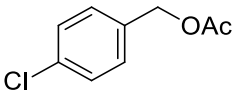
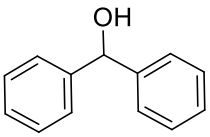
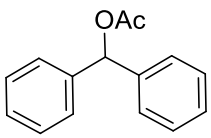
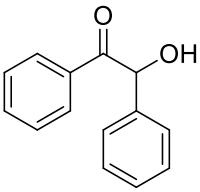
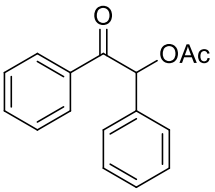
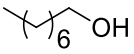
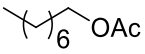
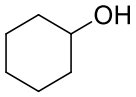
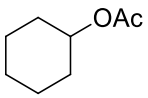
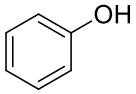
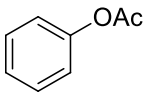
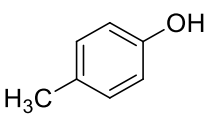
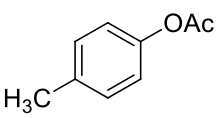
After that, we moved to improve the product yield by using different power levels (from 210 to 420 W) at microwave to obtain corresponding benzyl acetate with 82, 97, 96, 90, 82 % yields

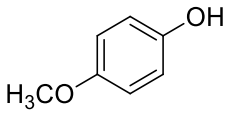
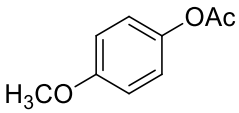
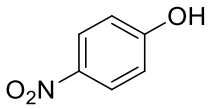
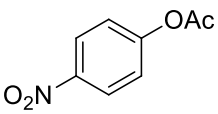
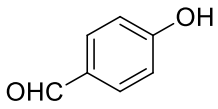
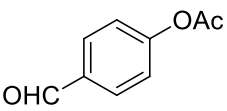
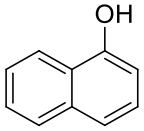
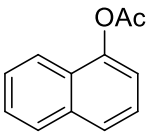
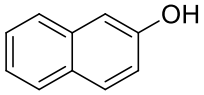
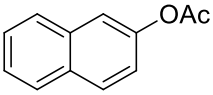
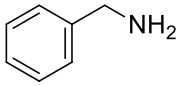
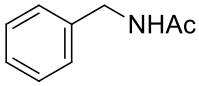
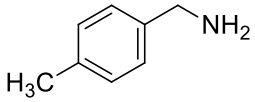
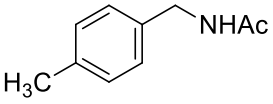
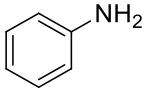
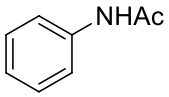
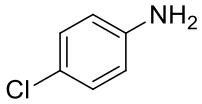
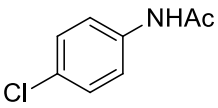
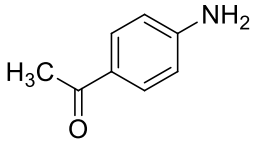
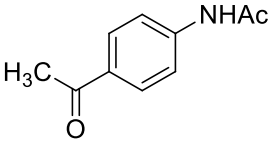
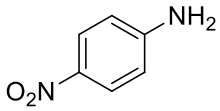
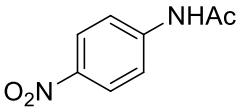
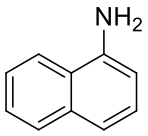
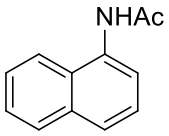
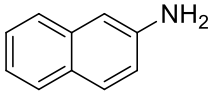
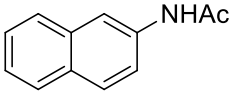
respectively. Finally, we observed that the reaction was very clean and efficient at 240 W power level and corresponding product was formed within 2 minutes with excellent yield. Results of all compounds are summarized in Table 2.

Based on these optimal reaction conditions, 2 mol% of catalyst was used to convert a variety of alcohols, phenols and amines with acetic anhydride into the corresponding esters and amides under microwave irradiation (240 W) at solvent free conditions (Table 3).

In this procedure, Benzylic alcohols and its derivatives smoothly reacted with acetic anhydride to obtain corresponding benzylic esters (entry **3a-3c**) with 97, 97 and 95% yields respectively. Next, sterically hindered 1-phenylethanol and diphenylmethanol were also converted into its corresponding esters (entry **3d-3e**) in 92 and 90 % yields respectively in appropriate reaction time. Later, Aliphatic and cyclic alcohols like 1-octanol and cyclohexanol quantitatively converted into their desired esters (entry **3f-3g**) with 98 and 97 % yields, respectively.

Table 3. Acetylation of alcohols, phenols and amines

Sr. No.	Alcohol/Phenol/ Amine	Product	Time (min)	Yield[a] (%)	M.P. °C [Lit. M.P.][Ref]
a.			2	97	Colorless Oil
b.			2	97	Colorless Oil
c.			2	95	Colorless Oil
d.			2	92	Colorless Oil
e.			2	90	Colorless Oil
f.			2	98	Colorless Oil
g.			2	97	Colorless Oil
h.			3	90	Colorless Oil
i.			3	92	Colorless Oil

j.			4	92	Colorless Oil
k.			4	88	78-80 [76-80] [28]
l.			4	89	Colorless Oil
m.			3	92	66-68 [69-70][21]
n.			3	90	74-76 [68-72] [28]
o.			2	95	60-62
p.			2	95	150-152
q.			3	94	112-114
r.			4	91	170-172 [172-175] [28]
s.			4	90	168-170
t.			4	89	218-220 [213-217] [28]
u.			3	92	156-158 [127-129] [28]
v.			3	92	132-134

[a]Isolated yield

Similarly, phenol and its derivatives were examined with same reaction condition. They were reacted slowly than alcohols due to presence of resonance as well as inductive effects on aromatic ring which decreases the nucleophilic character of phenol and obtained their corresponding esters **(entry 3h-3n**) with little extended reaction time (3-4 minutes).

Furthermore, this methodology was extended to acylation of benzyl amines and aromatic amines including different substituents present on aromatic ring. Interestingly, benzyl amines and its derivatives were converted to its respective amides in higher yields (entry **3o-3p**) within stipulated reaction time. But aromatic amines and its derivatives reacted slowly than benzyl amines as like phenols (same reason mentioned above) to obtained corresponding aromatic amides (entry **3q-3v**) in good yields increased reaction time (3-4 minutes). All details were clearly mentioned in Table 3.

For the investigation of reusability of the catalyst, reaction was carried out using model reaction (Scheme 1). After the completion of the reaction, ethyl acetate was added to the reaction mixture and solid catalyst was filtered off. Separated catalyst washed with ethyl acetate and reused up to four times for further reactions without loss of its a significant **efficiency (Table 4).**

Table 4. Recyclability study of the Phthalimide-N-sulfonic acid catalyst

Sr. No.	Cycle	Isolated Yields (%)
1	1	97
2	2	95
3	3	92
4	4	90

In general, all the reactions were very neat in terms of conversion as well as isolation of products. All the products were characterized by their spectroscopy analysis such as ^{1}H, ^{13}C NMR, and IR spectroscopy.

4. Conclusion

In summary, we have demonstrated a simple, efficient and environmentally benign novel methodology for the acetylation of various alcohols, phenols and amines using Phthalimide N-sulfonic acid as organo-catalyst under microwave irradiation at solvent-free conditions. This organo-catalyst is recycled and reused several times without loss of its efficiency. The present method offers remarkable advantages of microwave irradiation and organo-catalyst such as simple work-up procedure, short reaction time with excellent yields and environmentally benign procedure. All compounds have been confirmed by ^{1}H NMR, ^{13}C NMR, and IR spectroscopy.

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Supporting Information

Supporting information accompanies this paper on http://www.acgpubs.org/journal/organic-communications

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