

Al₂O₃/MeSO₃H (AMA) as a novel heterogeneous system for the nitration of aromatic compounds by magnesium nitrate hexahydrate

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Mg(NO₃)₂·6H₂O was an efficient nitrating agent in the presence of a mixture of Al₂O₃/MeSO₃H (AMA) as a novel heterogeneous system for the nitration of aromatic compounds without use of any organic solvents and with high selectivity. The reaction proceeds at room temperature for the nitration of highly reactive aromatic compounds such as phenols and anilines.

Keywords: nitration, Al₂O₃, MeSO₃H, aromatic compounds, Mg(NO₃)₂·6H₂O, solvent-free conditions

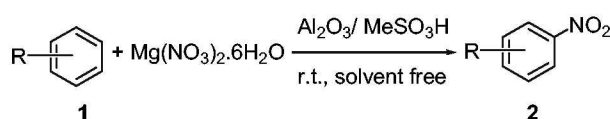
Nitroaromatic compounds are important chemicals that have application as or in the formation of solvents, dyes, pharmaceuticals, agrochemicals, explosives, and plastics in industry. They are also useful intermediates for the preparation of other compounds, particularly amines, by reduction of nitro groups.^{1–4} Electrophilic aromatic nitration is one of the most important reactions in organic chemistry, and the central role of the nitronium ion, NO₂⁺, in these processes has been well established.^{5–7} However, the majority of the reported methods for nitration of aromatic compounds suffer from one or more disadvantages such as low regioselectivity, over nitration, strongly acidic media, tedious work-up, oxidation of the reagents, not suitable for highly reactive and easily oxidised substrates such as aniline and phenols, and safety problems (storage, handling, using toxic reagents/catalysts or solvents).^{8–20} These disadvantages have encouraged extensive efforts to develop alternative procedures such as using solid acids^{2,9,15,17,21–25}, other sources of NO₂⁺,^{8,9,26–28} organic nitrating agents (acetyl nitrate, benzoyl nitrate, and trimethylsilyl nitrate),^{29,30} etc.^{31–33} However, these reported nitration procedures required special solvents or co-reagents.

Some of our previous work on the use of a mixture of Al₂O₃/MeSO₃H (AMA) as a heterogeneous system for various organic reactions,^{34–41} encouraged us to measure the efficiency of AMA in the nitration of aromatic compounds. Therefore, we report herein that AMA could conveniently be used to generate the nitronium ion by using magnesium nitrate (Scheme 1).

A typical experimental procedure is as follows: Mg(NO₃)₂·6H₂O (1 mmol) was added to a mixture of MeSO₃H (1 ml, 15 mmol) and Al₂O₃ (acidic type 506 C, 0.27 g, 3 mmol) and after a few minutes the aromatic compound **1** (1 mmol) was added. The mixture was stirred for the appropriate time and then the reaction mixture was extracted with ethyl acetate and after evaporation of the organic layer nitrated products **2** were obtained in a good to excellent yields. We also carried out the same reaction with a number of nitrate salts such as Cd(NO₃)₂·6H₂O, Ni(NO₃)₂·9H₂O, Cu(NO₃)₂·3H₂O and Zn(NO₃)₂·6H₂O. Although Zn(NO₃)₂·6H₂O showed reactivity with **1a** in the presence of AMA to give the nitrated product in a total isolated yield of 62%, the other nitrate salts gave a mixture of nitrated and oxidised products and the reactions were not complete even after 24 h.

Under the best reaction conditions described above, various aromatic compounds (Table 1, Entries 1–14) were nitrated. The products were identified by ¹H and ¹³C NMR, MS and IR analysis. The results are listed in Table 1.

According to the Table 1, various aromatic compounds with electron-donating and electron-withdrawing groups are nitrated by AMA. Because nitrophenols are very important organic synthesis intermediates, various phenolic compounds



Scheme 1

were nitrated using AMA (entries 1–6, 9 and 10). In the case of phenol, the nitration reaction afforded 4-nitrophenol and 2-nitrophenol in 47 and 45% isolated yields, respectively (Entry 1). However, other phenolic compounds containing an electron-withdrawing group at the *para* position gave regioselectively nitration at the position *ortho* to the OH group (entries 2–6). It is interesting that *p*-aminophenol gave only 4-amino-2-nitrophenol (**2d**) in high yield (entry 4). It should be noted that amino groups were protonated in the presence of AMA (as an acidic media) to give the –NH₃⁺ group, which is electron-withdrawing. Therefore, nitration of *p*-aminophenol (entry 4) and *p*-bromoaniline (Entry 11) occurred regioselectively at the *ortho* position relative to the OH and Br groups, respectively. The nitration of substituted benzaldehydes such as 4-methoxy- and 4-chlorobenzaldehydes afforded the nitrated products with the NO₂-*ortho* to the –OMe and –Cl groups, respectively (entries 7 and 8). In the case of 2-hydroxybenzaldehyde, the reaction was regioselective and only 2-hydroxy-4-nitrobenzaldehyde (**2i**) was obtained (entry 9). It is interesting to note that less reactive aromatic compounds such as chlorobenzene, cyanobenzene, and nitrobenzene were also nitrated in the presence of AMA in moderate to good yields (entries 12–14). Finally, although the corresponding 4-nitro-1,3-benzenediol was obtained in 85% yield by the nitration of 1,3-benzenediol (entry 10), under the same reaction conditions, the reaction of 1,4-benzenediol and 1,2-benzenediol (entries 15 and 16) resulted in complicated products containing nitrated and oxidised compounds.

In conclusion, we have discovered a very simple and efficient nitration process in the presence of AMA. This new nitration process employs AMA as heterogeneous system without using a halogenated organic solvent and the nitration of highly activated aromatic compounds such as phenols and anilines by magnesium nitrate hexahydrate were carried out at room temperature in air in good to excellent yields and also the reaction was easily worked-up.

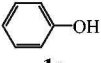
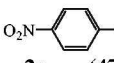

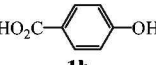
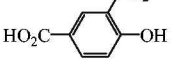
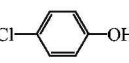
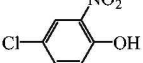
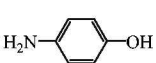
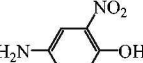
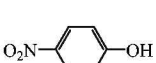
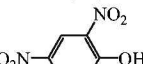
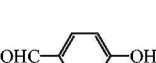
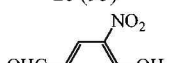

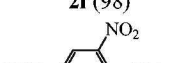
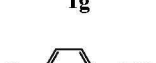
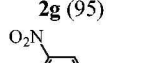
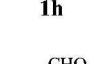
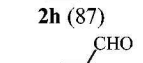
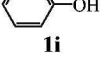
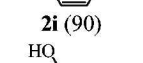
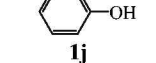
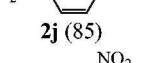
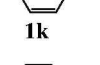
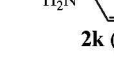


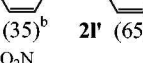
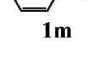
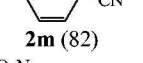
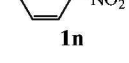
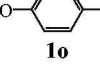
Experimental

General

¹H NMR and ¹³C NMR spectra were measured on a Bruker Advance DPX FT 250 and 62.9 MHz spectrometer with TMS as an internal standard. IR spectra were obtained on Perkin–Elmer or FTIR-800 instruments. Mass spectra were obtained on a Shimadzu GCMS0QP 1000EX at 20 and/or 70 eV. Elemental analyses were performed on PerkinElmer 240-B microanalyser. Al₂O₃ (acidic type 506 C) was purchased from the Merck company.

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Table 1 Nitration of aromatic compounds using AMA and $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$

Entry	Aromatic compounds	Reaction time	Nitrated compound/% yield ^a
1	 1a	30 min	 2a (47)  2a' (45)
2	 1b	90 min	 2b (98)
3	 1c	60 min	 2c (95)
4	 1d	6 h	 2d (98)
5	 1e	24 h	 2e (95)
6	 1f	35 min	 2f (98)
7	 1g	30 min	 2g (95)
8	 1h	4 h	 2h (87)
9	 1i	40 min	 2i (90)
10	 1j	30 min	 2j (85)
11	 1k	24 h	 2k (95)
12	 1l	20 h	 2l (35) ^b  2l' (65) ^b
13	 1m	24 h	 2m (82)
14	 1n	48 h	 2n (31)
15	 1o	6 h	decomposed
16	 1p	6 h	decomposed

^aYields are of the isolated compounds. ^bProportions were determined by ¹H NMR.

General procedure for nitration of aromatic compounds

CAUTION: The isolation of nitrated products must be carried out carefully with due precautions as some such compounds are known human carcinogens.

(1) Aromatic compounds without amino groups

$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (1 mmol) was added to a mixture of MeSO_3H (1.0 ml) and Al_2O_3 (0.27 g, 3.0 mmol). After a few minutes the aromatic compound (1 mmol) was added to this mixture. The mixture was stirred at room temperature for the appropriate time (Table 1). Then the mixture was poured into water (20 ml) and extracted four times with ethyl acetate (40 ml). The organic layer was washed with a saturated solution of sodium bicarbonate (20 ml), dried over Na_2SO_4 and evaporated under reduced pressure. The resulting product was purified by preparative TLC (silica) using a solvent mixture of EtOAc-hexane, 1:2 as eluent to afford the pure nitrated derivative.

(2) Aromatic compounds with amino groups

The reaction conditions were as described above but the work-up was a little different. After the reaction was completed the mixture was poured into a saturated solution of sodium bicarbonate (20 ml) and extracted five times with ethyl acetate (40 ml). Then the organic layer was dried over Na_2SO_4 and evaporated under reduced pressure. The resulting product was purified as described above.

4-Nitrophenol (2a): Yellow powder; m.p. 112–114°C (lit.,⁴² 114°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3331, 1614, 1592, 1500, 1346; ¹H NMR (250 MHz, CDCl_3): δ 8.18 (d, 2H, $J = 5.50$ Hz, ArH), 7.00 (d, 2H, $J = 5.49$ Hz, ArH), 6.35 (s, 1H, –OH); GC-MS/EI: m/z (%) = 139 (M^+ , 14), 109 (50), 81 (33), 65 (84), 53 (31), 46 (22), 39 (100), 30 (77), 28 (41).

2-Nitrophenol (2a'): Yellow powder; m.p. 42–44°C (lit.,⁴⁰ 45°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3241, 1623, 1606, 1593, 1538, 1479; ¹H NMR (250 MHz, CDCl_3): δ 10.5 (s, 1H, –OH), 8.10 (d, 1H, $J = 8.50$ Hz, ArH), 7.59 (m, 1H, ArH), 7.16 (d, 1H, $J = 8.4$ Hz, ArH), 7.00 (m, 1H, ArH); GC-MS/EI: m/z (%) = 139 (M^+ , 40), 109 (35), 81 (55), 65 (60), 53 (38), 39 (100), 30 (66).

4-Hydroxy-3-nitrobenzoic acid (2b): Yellow powder; m.p. 179–182°C (lit.,⁴³ 184°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3309, 3089, 2842, 1689, 1627, 1539, 1434, 1342; ¹H NMR (250 MHz, DMSO): δ 11.73 (brs, 1H, –CO₂H), 10.23 (brs, 1H, –OH), 8.33 (s, 1H, ArH), 7.98 (d, 1H, $J = 7.9$ Hz), 7.16 (d, 1H, $J = 7.9$ Hz, ArH); GC-MS/EI: m/z (%) = 183 (M^+ , 7), 149 (22), 129 (17), 97 (38), 57 (100).

4-Chloro-2-nitrophenol (2c): Yellow powder; m.p. 84–86°C (lit.,⁴² 85–89°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3490, 1631, 1531, 1469, 1384, 1346, 1238; ¹H NMR (250 MHz, DMSO): δ 11.33 (brs, 1H, –OH), 7.90 (s, 1H, ArH), 7.54 (d, 1H, $J = 8.9$ Hz), 7.14 (d, 1H, $J = 8.9$ Hz, ArH); GC-MS/EI: m/z (%) = 173 (M^+ , 16), 156 (7), 143 (26), 128 (10), 115 (41), 99 (39), 73 (29), 63 (100), 51 (20), 37 (13), 30 (26).

4-Amino-2-nitrophenol (2d): Yellow powder; m.p. 123–125°C (lit.,⁴² 125–127°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3460, 3255, 2958, 1627, 1546, 1469, 1373, 1234; ¹H NMR (250 MHz, DMSO): δ 9.64 (brs, 2H, –NH₂), 8.92 (brs, 1H, –OH), 7.30 (s, 1H, ArH), 7.23 (d, 1H, $J = 8.4$ Hz), 7.09 (d, 1H, $J = 8.4$ Hz, ArH); GC-MS/EI: m/z (%) = 154 (M^+ , 5), 137 (14), 124 (6), 108 (12), 79 (47), 53 (100), 28 (10).

2,4-Dinitrophenol (2e): Yellow powder; m.p. 107–110°C (lit.,⁴² 108–112°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3587, 3529, 1608, 1569, 1531, 1523, 1438, 1357, 1280; ¹H NMR (250 MHz, DMSO): δ 11.02 (brs, 1H, –OH), 8.65 (s, 1H, ArH), 8.26 (d, 1H, $J = 9.3$ Hz), 7.22 (d, 1H, $J = 9.2$ Hz, ArH); GC-MS/EI: m/z (%) = 184 (M^+ , 4), 167 (10), 129 (10), 111 (12), 85 (22), 69 (100).

4-Hydroxy-3-nitrobenzaldehyde (2f): Yellow powder; m.p. 138–140°C (lit.,⁴³ 140–141°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3236, 2816, 1693, 1620, 1562, 1423, 1334, 1269, 1126; ¹H NMR (250 MHz, CDCl_3): δ 11.01 (brs, 1H, –OH), 9.94 (s, 1H, –CHO), 8.63 (s, 1H, ArH), 8.14 (d, 1H, $J = 3.5$ Hz), 7.33 (d, 1H, $J = 3.5$ Hz, ArH); GC-MS/EI: m/z (%) = 167 (M^+ , 10), 149 (25), 129 (11), 111 (11), 85 (23), 69 (100).

4-Methoxy-3-nitrobenzaldehyde (2g): Yellow powder; m.p. 99–101°C (lit.,⁴² 97–100°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3487, 3001, 2623, 1697, 1620, 1539, 1431, 1357, 1288; ¹H NMR (250 MHz, DMSO): δ 9.84 (s, 1H, –CHO), 8.10 (s, 1H, ArH), 8.07 (d, 1H, $J = 8.7$ Hz, ArH), 7.45 (d, 1H, $J = 8.7$ Hz, ArH), 3.96 (s, 3H, –OMe); GC-MS/EI: m/z (%) = 181 (M^+ , 6), 167 (11), 149 (11), 129 (8), 111 (8), 85 (13), 69 (100).

4-Chloro-3-nitrobenzaldehyde (2h): Yellow powder; m.p. 140–147°C (lit.,⁴² 148–150°C); IR(KBr) ($\nu_{\text{max}}/\text{cm}^{-1}$): 3101, 2873, 2715, 2596, 1701, 1593, 1431, 1330; ¹H NMR (250 MHz, DMSO): δ 10.1 (s, 1H, –CHO), 8.22 (s, 1H, ArH), 7.90 (d, 1H, $J = 3.1$ Hz, ArH), 7.55 (d, 1H, $J = 3.1$ Hz, ArH); GC-MS/EI: m/z (%) = 185 (M^+ , 9), 167 (10), 149 (22), 129 (15), 97 (19), 73 (45), 57 (100).

2-Hydroxy-5-nitrobenzaldehyde (2i): Yellow powder; m.p. 127–129°C (lit.⁴² 129–130°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3460, 3074, 2935, 1666, 1620, 1477, 1334, 1323; ¹H NMR (250 MHz, DMSO): δ 11.5 (brs, 1H, –OH), 9.91 (s, 1H, –CHO), 8.49 (s, 1H, ArH), 8.32 (d, 1H, $J = 9.1$ Hz, ArH), 7.05 (d, 1H, $J = 9.1$ Hz, ArH); GC-MS/EI: m/z (%) = 167 (M⁺, 11), 149 (13), 129 (7), 111 (12), 85 (24), 69 (100).

4-Nitro-1,3-benzenediol (2j): Yellow powder; m.p. 120–123°C (lit.⁴² 122°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3388, 3300, 1628, 1592, 1525, 1511, 1444, 1327, 1284; ¹H NMR (250 MHz, CDCl₃): δ 10.2 (brs, 1H, –OH), 8.13 (d, 1H, $J = 9.2$ Hz, ArH), 7.25 (s, 1H, ArH), 6.62 (d, 1H, $J = 9.2$ Hz, ArH), 6.44 (brs, 1H, –OH); GC-MS/EI: m/z (%) = 155 (M⁺, 7), 139 (31), 121 (10), 109 (74), 77 (12), 65 (47), 51 (19), 39 (48), 30 (25), 28 (100).

4-Bromo-3-nitroaniline (2k): Yellow powder; m.p. 125–128°C (lit.⁴² 128–132°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3475, 3363, 1639, 1531, 1346; ¹H NMR (250 MHz, DMSO): δ 9.00 (s, 1H, ArH), 8.75 (d, 1H, $J = 9.0$ Hz, ArH), 8.50 (d, 1H, $J = 9.0$ Hz, ArH), 6.41 (brs, 2H, –NH₂); GC-MS/EI: m/z (%) = 217 (M⁺, 3), 213 (6), 185 (7), 149 (22), 129 (14), 97 (23), 73 (49), 57 (100).

4-Nitrochlorobenzene (2l): Yellow powder; m.p. 82–84°C (lit.⁴² 84–86°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3106, 1604, 1578, 1519, 1477, 1344; ¹H NMR (250 MHz, DMSO): δ 8.24 (d, 2H, $J = 8.5$ Hz, ArH), 7.52 (d, 2H, $J = 8.5$ Hz, ArH); GC-MS/EI: m/z (%) = 157 (M⁺, 15), 129 (21), 127 (19), 111 (84), 99 (33), 75 (100).

2-Nitrochlorobenzene (2l'): Yellow powder; m.p. 30–32°C (lit.⁴² 32°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3096, 1688, 1550, 1467; ¹H NMR (250 MHz, DMSO): δ 7.32–7.87 (m, 4H, ArH); GC-MS/EI: m/z (%) = 157 (M⁺, 15), 129 (21), 127 (20), 111 (83), 99 (33), 75 (100).

3-Nitrobenzonitrile (2m): Yellow powder; m.p. 110–113°C (lit.⁴² 114–117°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3111, 2955, 1926, 2238, 1751, 1619, 1582, 1533, 1463; ¹H NMR (250 MHz, DMSO): δ 8.76 (s, 1H, ArH), 7.53–8.74 (m, 3H); GC-MS/EI: m/z (%) = 148 (M⁺, 21), 102 (75), 90 (34), 75 (22), 57 (100).

1,3-Dinitrobenzene (2n): Yellow powder; m.p. 84–85°C (lit.⁴² 86–89°C); IR(KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3112, 1616, 1544, 1471, 1346; ¹H NMR (250 MHz, DMSO): δ 8.84 (s, 1H, ArH), 7.62–8.61 (m, 3H); GC-MS/EI: m/z (%) = 168 (M⁺, 13), 122 (24), 76 (100), 75 (76), 57 (23).

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