Phosphomolybdic Acid (PMA) – SiO_2 as a Heterogeneous Solid Acid Catalyst for the One-Pot Synthesis of 2*H*-Indazolo[1,2-*b*]phthalazine-triones

by **Gowravaram Sabitha***, **Chitti Srinivas**, **Avula Raghavendar**, and **Jhillu Singh Yadav**

Organic Division-I, Indian Institute of Chemical Technology, Hyderabad – 500007, India (fax: +91-40-27160512; e-mail: gowravaramsr@yahoo.com)

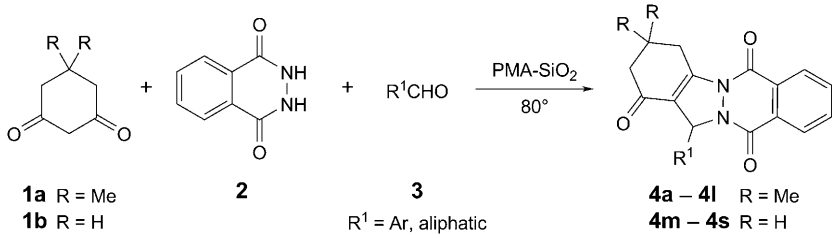
Phosphomolybdic acid (PMA) – SiO_2 was found to be an efficient catalyst for the three-component condensation reaction of phthalhydrazide, 1,3-diketone, and aldehydes to produce 2*H*-indazolo[1,2-*b*]phthalazine-triones in excellent yields. The catalyst can be recovered and reused without significant loss of activity.

Introduction. – Nitrogen-containing heterocyclic compounds are widespread in nature, and their applications to biologically active pharmaceuticals, agrochemicals, and functional materials are becoming increasingly important [1]. Among a large variety of N-containing heterocyclic compounds, heterocycles containing hydrazine moieties as 'fusion site' have received considerable attention because of their pharmacological properties and clinical applications [2]. Moreover, fused phthalazines have been found effective for the inhibition of p38 MAP kinase [3], for selective binding of GABA receptor [4], and as anti-anxiety drug [5], antitumor agent [6], and high-affinity ligand to the a2d-1 subunit of calcium channel [7]. Fused phthalazine derivatives also possess some biological activities such as anticonvulsant [8], cardiotonic [9], and vasorelaxant [10]. Thus, the development of an efficient and versatile method for the preparation of fused phthalazine derivatives is an active ongoing research area, and there is a potential for further improvement toward green chemistry and improved yields. Even though many reports have been published on the solution-phase synthesis of fused phthalazine derivatives [3–7][11], reports on the synthesis of phthalazines fused with indazole are limited [12]. While our work on the synthesis of title compounds **4** and **5** (*cf. Schemes 1* and *2*) was in review process, two reports on the synthesis of **4** have appeared [13].

Heterogeneous catalysts have gained much importance in recent years due to economic and environmental benefits [14][15]. Recently, the use of heteropolyacids (HPAs), as environmentally friendly and economically viable solid acids, is increasing continuously owing to their ease of handling and high catalytic activities. Among them phosphomolybdic acid (PMA, $H_3PMO_{12}O_{40}$) is one of the less expensive and commercially available catalysts [16].

Phthalhydrazide (=2,3-dihydrophthalazine-1,4-dione) is a very interesting fused heterobicyclic compound, which has two rather NH acidic protons [17]. In the present study, we report an efficient, one-pot, three-component method for the preparation of 2*H*-indazolo[1,2-*b*]phthalazine-trione derivatives by condensation of 1,3-diones **1a**–**1c**, phthalhydrazide **2**, and aldehydes **3** under solvent-free conditions (*Scheme 1*).

Scheme 1



Results and Discussion. – Initially, a mixture of dimedone **1a** (= 5,5-dimethylcyclohexane-1,3-dione), phthalhydrazide (**2**), and benzaldehyde **3a** was heated at 80° in the presence of a catalytic amount of PMA – SiO_2 under solvent-free conditions. The reaction was complete in 30 min, and the product, 2,3,4,13-tetrahydro-3,3-dimethyl-13-phenyl-1*H*-indazolo[1,2-*b*]phthalazine-1,6,11-trione (**4a**) was isolated in 85% yield (*Scheme 1*). Encouraged by this result, to demonstrate the scope of the procedure, a variety of aromatic aldehydes **3b** – **3i**, as well as aliphatic aldehydes **3j** – **3l**, were treated with **1a** and **2**. In all cases, the reactions were clean and proceeded efficiently at 80° under solvent-free conditions affording the corresponding products **4b** – **4l** in excellent yields, and the results are listed in *Table 1*. No side-products or decomposition of the products are observed. All the products were characterized by ^{1}H- and ^{13}C-NMR, and IR spectroscopy, and mass spectrometry.

Table 1. *PMA – SiO_2-Catalyzed Synthesis of Indazolo[1,2-*b*]phthalazine-triones* **4**[a])

Entry	Aldehyde **3**	Product **4**	Time [min]	Yield [%][b])	M.p. [°][c])
1	Benzaldehyde	**4a**	30	85	198 – 200 (204 – 206)
2	4-Chlorobenzaldehyde	**4b**	30	90	256 – 258 (262 – 264)
3	4-Fluorobenzaldehyde	**4c**	30	85	218 – 220 (217 – 219)
4	2-Bromobenzaldehyde	**4d**	40	85	242 – 244
5	3-Chlorobenzaldehyde	**4e**	35	86	195 – 197
6	4-Methoxybenzaldehyde	**4f**	40	85	207 – 209
7	4-Nitrobenzaldehyde	**4g**	30	90	225 – 227 (223 – 225)
8	4-Isopropylbenzaldehyde	**4h**	40	87	204 – 206
9	4-(*tert*-Butyl)benzaldehyde	**4i**	40	87	214 – 216
10	Butanal	**4j**	20	92	133 – 135
11	Hexanal	**4k**	20	91	140 – 142
12	2-Methylpropanal	**4l**	25	90	168 – 170
13	Benzaldehyde	**4m**	30	84	223 – 225
14	4-Isopropylbenzaldehyde	**4n**	35	82	195 – 197
15	4-Methoxybenzaldehyde	**4o**	40	81	242 – 244
16	4-Nitrobenzaldehyde	**4p**	40	80	252 – 254
17	Butanal	**4q**	25	86	172 – 174
18	Pentanal	**4r**	25	84	semisolid
19	2-Methylpropanal	**4s**	30	84	semisolid

[a]) All products were characterized by spectral data and known compounds were compared with authentic samples. [b]) Isolated pure products. [c]) Known melting points are given in parantheses.

Similarly, the protocol was extended to synthesize indazolophthalazines-triones **4m** – **4s** by reacting cyclohexane-1,3-dione (**1b**) with **2**, and aromatic as well as aliphatic aldehydes (*Scheme 1* and *Table 1*) in the presence of a catalytic amount of PMA – SiO_2 at 80° in good yields within short reaction times. Under similar conditions, acetylacetone (= pentane-2,4-dione; **1c**) also reacts efficiently with **2** and aromatic aldehydes as well as aliphatic aldehydes to give the corresponding products **5a** – **5c** in 80 – 84% yield within 40 – 45 min (*Scheme 2* and *Table 2*). These results clearly shows the generality of the reaction using PMA – SiO_2 under solvent-free conditions. However, heteroaromatic aldehydes such as furan- and thiophene-carbaldehydes did not undergo this condensation reaction; instead, the starting materials were recovered.

Scheme 2

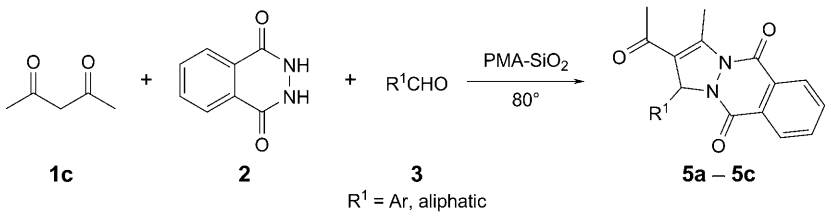


Table 2. *PMA – SiO_2-Catalyzed Synthesis of Pyrazolo[1,2-*b*]phthalazin-triones*[a])

Entry	Aldehyde **3**	Product **5**	Time [min]	Yield [%][b])	M.p. [°]
20	Benzaldehyde	**5a**	40	82	semisolid
21	4-Methoxybenzaldehyde	**5b**	45	80	semisolid
22	Butanal	**5c**	40	84	semisolid

[a]) All products were characterized by spectral data, and known compounds were compared with authentic samples. [b]) Isolated pure products.

It is assumed that the reaction proceeded by *Knovenagel* condensation of 1,3-dione and aldehyde, followed by *Michael* addition of the phthalhydrazide (**2**), and subsequent cyclization to afford the product **4** or **5**. It is important to note that the catalyst was recovered by simple filtration and reused in subsequent runs with no decrease in activity. The possibility of recycling the catalyst, *i.e.*, PMA – SiO_2, is one of the key advantages of this procedure, which was demonstrated using benzaldehyde as a model reaction. The recovered catalyst was recycled upto three times with no decrease in the conversion.

In conclusion, we have developed a simple and general method for the synthesis of novel 2*H*-indazolo[1,2-*b*]phthalazine-triones *via* a three-component reaction of 1,3-diones, phthalhydrazide (**2**), and aldehydes catalyzed by PMA – SiO_2 as a green recyclable and heterogeneous catalyst under solvent-free conditions. Advantages of this method are its generality, short reaction times, easy workup, and ease of recovery and reuse of the catalyst, which make this procedure quite simple, more convenient, and environmentally benign.

Experimental Part

General. Commercial reagents were used without further purification. All solvents were purified by standard techniques. Column chromatography (CC): silica gel (SiO_2; 60–120 mesh). M.p.: *Fisher Johns* apparatus; uncorrected. IR Spectra: *Perkin-Elmer 683* spectrometer. NMR Spectra: in $CDCl_3$; *Varian Gemini 200*, *Bruker 300*, or *Varian Unity 400* NMR spectrometers; chemical shifts (δ) are given in ppm and are referenced to tetramethylsilane (TMS) as internal standard; coupling constants (J) are given in Hz. MS: *Finnigan MAT 1020B* or *micro mass VG 70-70 H* spectrometers operating at 70 eV using a direct inlet system.

Preparation of PMA–SiO_2 Catalyst. SiO_2 (100–200 mesh; 450 mg) was added slowly to a soln. of $H_3PMo_{12}O_{40}\cdot 24\,H_2O$ (50 mg) in MeOH (5 ml). The mixture was stirred at r.t. for 6 h. MeOH was evaporated under reduced pressure to afford PMA–SiO_2 as a yellowish powder.

General Procedure for the Synthesis of **4**. To a mixture of the cyclohexane-1,3-dione **1a** or **1b** (1 mmol), aldehyde (1 mmol), and **2** (1 mmol) in 1.0 g of PMA–SiO_2 (0.05 mmol) was heated at 80° for the appropriate time (see *Table 1*). After completion of the reaction as indicated by TLC, the mixture was diluted with AcOEt, and the catalyst was recovered by simple filtration, and the crude product was purified by CC to afford pure product.

*13-(2-Bromophenyl)-2,3,4,13-tetrahydro-3,3-dimethyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4d**). Yellow powder (85%). M.p. 242–244°. IR (KBr): 2924, 2854, 1711, 1659, 1463, 1361, 1310, 1268, 1148, 1029, 794, 759, 698. ^{1}H-NMR (300 MHz): 1.23 (*s*, 3 H); 1.25 (*s*, 3 H); 2.29 (*s*, 2 H); 3.23, 3.38 (*AB*, J = 19.6, 2 H); 6.61 (*s*, 1 H); 7.11–8.36 (*m*, 8 H). ^{13}C-NMR (50 MHz): 28.6; 28.7; 34.6; 38.1; 50.8; 65.7; 117.3; 127.5; 127.6; 127.9; 128.5; 128.6; 128.9; 129.9; 133.4; 133.7; 134.6; 151.7; 154.3; 156.3; 191.6. HR-ESI-MS: 473.0472 ([M + Na]$^+$; calc. 473.0476).

*2,3,4,13-Tetrahydro-13-(4-methoxyphenyl)-3,3-dimethyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4f**). Yellow powder (85%). M.p. 207–209°. IR (KBr): 2958, 1663, 1628, 1604, 1513, 1465, 1425, 1361, 1312, 1265, 1242, 1172, 1099, 1027, 842, 797, 699. ^{1}H-NMR (300 MHz): 1.23 (*s*, 3 H); 1.24 (*s*, 3 H); 2.30 (*s*, 2 H); 3.21, 3.42 (*AB*, J = 18.9, 2 H); 3.75 (*s*, 3 H); 6.38 (*s*, 1 H); 6.80–8.35 (*m*, 8 H). ^{13}C-NMR (75 MHz): 28.4; 28.7; 34.6; 38.1; 50.9; 55.1; 64.5; 114.1; 118.5; 127.6; 127.9; 128.3; 128.5; 128.9; 129.1; 133.4; 134.9; 1507; 154.2; 156.0; 159.6; 192.2. HR-ESI-MS: 425.1464 ([M + Na]$^+$; calc. 425.1477).

*2,3,4,13-Tetrahydro-13-(4-isopropylphenyl)-3,3-dimethyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4h**). Yellow powder (87%). M.p. 204–206°. IR (KBr): 2956, 1664, 1618, 1509, 1465, 1421, 1362, 1306, 1267, 1144, 1104, 1053, 1019, 972, 842. ^{1}H-NMR (300 MHz): 1.20 (*s*, 3 H); 1.23 (*s*, 6 H); 1.24 (*s*, 3 H); 2.31 (*s*, 2 H); 2.85 (*m*, 1 H); 3.22, 3.42 (*AB*, J = 18.9, 2 H); 6.39 (*s*, 1 H); 7.13–8.34 (*m*, 8 H). ^{13}C-NMR (75 MHz): 23.6; 23.8; 28.5; 28.6; 34.6; 38.0; 40.8; 50.9; 64.7; 118.6; 126.7; 127.0; 127.6; 127.9; 128.9; 129.1; 130.3; 133.4; 134.4; 149.1; 150.8; 154.2; 156.0; 192.2. HR-ESI-MS: 437.1847 ([M + Na]$^+$; calc. 437.1841).

*2,3,4,13-Tetrahydro-3,3-dimethyl-13-propyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4j**). Yellow powder (92%). M.p. 133–135°. IR (KBr): 2953, 2929, 2867, 1651, 1467, 1434, 1365, 1299, 1274, 1149, 1078, 1029, 781, 693, 624, 575. ^{1}H-NMR (200 MHz): 0.89 (*t*, J = 6.6, 3 H); 1.03–1.16 (*m*, 2 H); 1.20 (*s*, 3 H); 1.23 (*s*, 3 H); 2.34 (*s*, 2 H); 1.97–2.48 (*m*, 2 H); 3.24, 3.34 (*AB*, J = 19.8, 2 H); 5.63 (*s*, 1 H); 7.77–7.93 (*m*, 2 H); 8.26–8.37 (*m*, 2 H). ^{13}C-NMR (75 MHz): 13.7; 16.7; 28.4; 28.7; 31.5; 34.5; 38.1; 50.9; 62.8; 117.3; 127.4; 127.8; 128.8; 128.9; 133.3; 134.4; 151.6; 154.7; 156.1; 192.9. HR-ESI-MS: 361.1526 ([M + Na]$^+$; calc. 361.1528).

*2,3,4,13-Tetrahydro-13-phenyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4m**). Yellow powder (84%). M.p. 223–225°. IR (KBr): 2925, 1659, 1495, 1463, 1418, 1367, 1304, 1278, 1177, 1093, 992, 896, 833, 804, 765, 701, 561. ^{1}H-NMR (200 MHz): 2.17–2.51 (*m*, 4 H); 3.17 (*dt*, J = 19.8, 5.9, 1 H); 3.56 (*dt*, J = 19.8, 5.9, 1 H); 6.41 (*s*, 1 H); 7.22–8.40 (*m*, 9 H). ^{13}C-NMR (75 MHz): 22.2; 24.4; 36.8; 64.8; 119.5; 127.1; 127.6; 127.8; 128.5; 128.9; 129.9; 133.4; 134.4; 136.2; 152.2; 154.1; 155.9; 192.4. HR-ESI-MS: 367.1061 ([M + Na]$^+$; calc. 367.1058).

*2,3,4,13-Tetrahydro-13-(4-methoxyphenyl)-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4o**). Yellow powder (81%). M.p. 242–244°. IR (KBr): 2927, 1658, 1509, 1462, 1364, 1220, 1173, 1099, 1028, 832, 698, 531. ^{1}H-NMR (300 MHz): 2.20–2.50 (*m*, 4 H); 3.28 (*dt*, J = 19.6, 5.3, 1 H); 3.55 (*dt*, J = 19.6, 5.3, 1 H); 3.75 (*s*, 3 H); 6.36 (*s*, 1 H); 6.75–8.35 (*m*, 8 H). ^{13}C-NMR (75 MHz): 22.5; 24.7; 37.1; 55.4; 64.8;

114.3; 119.8; 127.9; 128.1; 128.5; 128.7; 129.1; 129.4; 133.6; 134.6; 152.4; 154.4; 156.2; 159.9; 192.8. HR-ESI-MS: 397.1149 ($[M + Na]^+$; calc. 397.1164).

*2,3,4,13-Tetrahydro-13-propyl-1*H-*indazolo[1,2-*b*]phthalazine-1,6,11-trione* (**4q**). Yellow powder (86%). M.p. 172–174°. IR (KBr): 2956, 2930, 2858, 1656, 1466, 1427, 1368, 1319, 1279, 1176, 1132, 1083, 1010, 968, 905, 843, 795, 758, 695, 590, 525. ^{1}H-NMR (300 MHz): 0.89 (*t*, *J* = 7.2, 3 H); 1.10–1.28 (*m*, 2 H); 1.96–2.56 (*m*, 6 H); 3.21 (*dt*, *J* = 19.3, 5.5, 1 H); 3.44 (*dt*, *J* = 19.3, 5.5, 1 H); 5.65 (*s*, 1 H); 7.80–7.90 (*m*, 2 H); 8.28–8.37 (*m*, 2 H). ^{13}C-NMR (75 MHz): 13.9; 16.8; 22.5; 24.6; 31.7; 37.1; 63.1; 118.9; 127.7; 128.1; 129.1; 1292; 133.5; 134.6; 153.1; 154.8; 156.3; 193.5. HR-ESI-MS: 333.1211 ($[M + Na]^+$; calc. 333.1215).

*2-Acetyl-3-methyl-1-phenyl-1*H-*pyrazolo[1,2-*b*]phthalazine-5,10-dione* (**5a**). Brown semisolid (82%). IR (KBr): 2925, 2854, 1689, 1655, 1601, 1490, 1464, 1415, 1353, 13119, 1279, 1204, 1134, 1107, 1075, 1028, 960, 929, 832, 755, 698, 637, 589, 539. ^{1}H-NMR (300 MHz,): 2.06 (*s*, 3 H); 3.09 (*s*, 3 H); 6.47 (*s*, 1 H); 7.29–8.35 (*m*, 9 H). ^{13}C-NMR (75 MHz): 14.4; 30.5; 66.1; 118.9; 127.2; 127.9; 128.2; 128.5; 128.7; 128.8; 129.1; 133.4; 134.1; 136.3; 146.1; 154.1; 156.2; 192.8. HR-ESI-MS: 355.1050 ($[M + Na]^+$; calc. 355.1058).

*2-Acetyl-3-methyl-1-propyl-1*H-*pyrazolo[1,2-*b*]phthalazine-5,10-dione* (**5c**). Brown semisolid (84%). IR (KBr): 2963, 2931, 2873, 1851, 1774, 1687, 1652, 1465, 1421, 1340, 1292, 1204, 1199, 1125, 1087, 1023, 968, 898, 791, 756, 699, 663, 631, 525. ^{1}H-NMR (300 MHz): 0.88 (*t*, *J* = 7.6, 3 H); 1.09–1.31 (*m*, 2 H); 1.71–1.88 (*m*, 1 H); 2.15–2.36 (*m*, 1 H); 2.39 (*s*, 3 H); 2.90 (*s*, 3 H); 5.72 (*s*, 1 H); 7.77–7.92 (*m*, 2 H); 8.26–8.37 (*m*, 2 H). ^{13}C-NMR (75 MHz): 13.6; 14.6; 16.3; 30.3; 32.6; 63.1; 119.6; 127.1; 127.9; 128.6; 129.4; 133.3; 134.2; 145.3; 154.3; 156.3; 193.3. HR-ESI-MS: 321.1220 ($[M + Na]^+$; calc. 321.1215).

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