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Research Article

# ZINC SULPHATE CATALYZED SYNTHESIS OF 2-ARYL BENZIMIDAZOLES UNDER SONICATION

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### ABSTRACT

An expeditious synthesis of 2-aryl-benzimidazoles by the condensation of *o*-phenylenediamine with various arylaldehydes is described. This greener protocol is catalyzed Zinc sulphate and proceeds efficiently in the absence of any organic solvent under sonication.

**Keyword:** *o*-phenylenediamine, Imidazoles, Aldehydes, Zinc Sulphate, Sonication.

### INTRODUCTION

The benzimidazole contains a phenyl ring fused to a imidazoles ring, as indicated in the structure of benzimidazole. The important group of substances has found practical application in a number of fields. Recently in benzimidazole chemistry has been revived somewhat by the discovery that the 5,6-dimethyl benzimidazole moiety is a part of the chemical structure of vitamin B12. Historically, the first benzimidazole was prepared in 1872 by Hoebrecker, who obtained 2, 5 or 2, 6-dimethyl benzimidazole (II) by the reduction of 2-nitro-4-methyl acetanilide. The benzimidazoles are also known as benziminazolones or benzoglyoxalines. They have been named also as derivatives of *o*-phenylene diamines<sup>1</sup>. Benzimidazoles are very useful intermediates for the development of molecules of pharmaceutical and biological interest. Substituted benzimidazole derivatives have found applications in diverse therapeutic areas including antiulcers, antihypertensives, antivirals, antifungals, anticancers, and antihistaminics<sup>2</sup>.

Methods of benzimidazole synthesis include the condensation of *o*-aryldiamines and carboxylic acids<sup>5</sup> or its derivatives<sup>6</sup> in the presence of strong acids and even sometimes combined with very high temperature<sup>7</sup>. The other method is that benzimidazoles have been prepared by classical cyclocondensation of *o*-phenylenediamine and aldehydes under oxidative conditions employing sulfamic acids<sup>8</sup>, IL<sup>9</sup>, In(OTf)<sub>3</sub><sup>10</sup>, Sc(OTf)<sub>3</sub><sup>11</sup>, activated carbon<sup>12</sup>, FeCl<sub>3</sub><sup>13</sup>, 6H<sub>2</sub>O<sup>13</sup>, H<sub>2</sub>O<sub>2</sub>/HCl<sup>14</sup>, sulfonic acid functionalized silica<sup>15</sup>, DDQ<sup>16</sup>, NH<sub>4</sub>OAc<sup>17</sup>, IL<sup>18</sup> (bromodimethyl) sulfonium bromide<sup>19</sup>, iodo

benzene diacetate<sup>20</sup>, air<sup>21</sup>, AIKIT<sup>22</sup> mono and bifunctional solid Catalyst<sup>23</sup>, scolecit<sup>24</sup>, Copper(I) Chloride<sup>25</sup>, manganese (III) acetate<sup>26</sup>, silica-supported thionyl chloride<sup>27</sup> and SDS<sup>28</sup> as catalysts. Unfortunately, some of these methods have one or more drawbacks such as expensive reagents, drastic reaction conditions, low yields, tedious work up procedures and co-occurrence of several side reactions. Therefore, the discovery of mild and practical routes for synthesis of 2-substituted benzimidazoles continues to attract the attention of researchers. Catalysts and reagents supported on inorganic substrates have received increasing attention in recent years as a means to develop more convenient or selective catalysts or reagents.

We develop a simple system catalyzed by Zinc sulphate and sonication gives good yield and purity.

### MATERIALS AND METHODS

All chemicals were obtained from commercial suppliers and used without further purification. Melting points were uncorrected. <sup>1</sup>H NMR spectra were recorded on a Bruker AVANCE 600 (600 MHz) spectrometer using TMS as internal standard and DMSO.

**General procedure for the Synthesis of 2-substituted benzimidazoles** *o*-Phenylenediamine (1, 1 mmol) and aromatic aldehyde (1, 1 mmol) were dissolved in DMF (2 mL) in a 25 mL Round bottom flask, add 1 mmol of Zinc sulphate was then added and the mixture was stirred at 25 °C on sonicator for the specified time as indicated in The reaction was followed by TLC. After the completion of the reaction, the mixture was dissolved in ethyl

acetate, and the catalyst was removed by filtration and washed with ethyl acetate. The solvent was evaporated under pressure to give the crude product, which was purified by column chromatography on silica gel eluted with petroleum ether or the mixture of EtOAc and petroleum ether. All of the compounds were identified by comparing their melting points with that reported in literatures 8,24,27 and characterized by <sup>1</sup>H NMR .

**Table: Physicochemical data of synthesized compounds**

Sr. No.	Aromatic group	Time (min)	Yield (%)	MP (°C)
1	C <sub>6</sub> H <sub>5</sub>	20	86	291
2	2-ClC <sub>6</sub> H <sub>4</sub>	22	87	131
3	3- ClC <sub>6</sub> H <sub>4</sub>	23	87	233
4	4- MeOC <sub>6</sub> H <sub>4</sub>	22	88	224
5	2-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	24	89	256
6	3- NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	25	87	307
7	4- NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	23	88	314
8	4-MeC <sub>6</sub> H <sub>5</sub>	22	89	266
9	4-Me <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	23	90	238
10	2-Furanyl	24	91	283
11	3,4-OCH <sub>2</sub> OC <sub>6</sub> H <sub>3</sub>	25	92	247

## CONCLUSION

In summary, we have found a practical procedure for the preparation of 2-substituted benzimidazoles catalyzed by ferrous sulphate stirred at ambient temperature. Our procedure is characterized by milder conditions, shorter reaction time, higher yield and involvement of non toxic and expensive catalyst

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