Asian Journal of Chemistry Vol. 18, No. 1 (2006), 757–759

NOTE

Synthesis of Some Benzimidazolyl Chalcones

M. NYATI, N.S. RAO, Y.K. SRIVASTAVA* and B.L. VERMA†
Department of Chemistry, M.P. Government College, Chittorgarh-312 001, India

Some benzimidazolyl chalcones have been synthesized by condensation of 2-acetyl benzimidazole (3) with aromatic aldehydes in presence of aqueous KOH by microwave irradiation technique.

Key Words: Synthesis, Benzimidazolyl chalcones, Microwave irradiation.

Benzimidazoles constitute an important group of heterocyclic compounds having a wide spectrum of biological activities[1–4]. 2-Substituted benzimidazole derivatives have been found to be biologically most potent. Chalcones are the natural biocides[5] and are well known intermediates in the synthesis of heterocyclic compounds exhibiting various biological activities[6]. Introduction of heterocyclic nucleus in the chalcone system has been reported to enhance the biological activities.

The use of microwave irradiation for the synthesis of heterocyclic compounds is a safe, rapid, convenient and ecofriendly method[7]. In this communication, the synthesis of some chalcones having benzimidazole nuclei using microwave irradiation technique is reported.

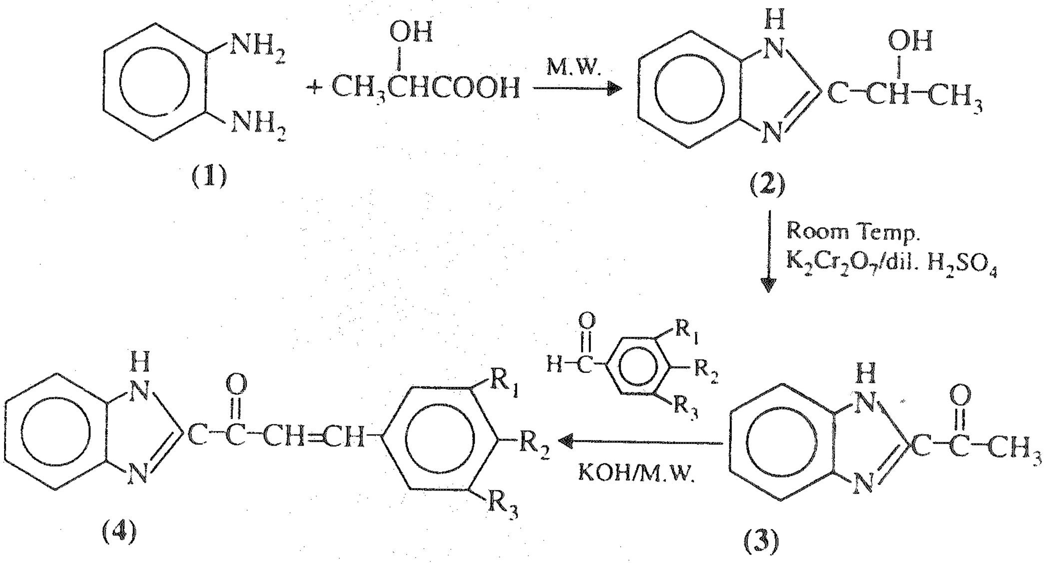
The basic nucleus 2-acetyl benzimidazole (3) was prepared by oxidation of 2-(hydroxyethyl)-benzimidazole (2). The latter compound was obtained by condensation of *o*-phenylene diamine (1) with lactic acid under Phillip's condition[8]. Oxidation of (2) with $K_2Cr_2O_7$ in dilute H_2SO_4 yielded the expected benzimidazole derivatives[9, 10]. Condensation of (3) with aromatic aldehydes in alkaline medium gave benzimidazolyl chalcones (4).

All the transformations except oxidation of (2) to (3) were carried out under microwave irradiation. The oxidation of (2) to (3) was carried out by conventional method at room temperature. The purity of all the synthesized compounds was checked out by TLC using silica gel-G. Their structure was confirmed by elemental and spectral analysis and comparison of m.p. of some compounds with authentic samples[11].

The IR spectra (KBr, cm^{-1}) of compounds (4) showed prominent absorption band at 3245 ν(NH), 3050 ν(CH═CH), 1650 ν(C═O), 1512 ν(C═N). The NMR spectra of compounds showed NMR signal at δ 3.60 (s, 3H, OCH_3–Ar), 6.89 (d, 2H, CH═CH), 7.66 (d, 2H, Ar-H), 7.66–7.85 Ar-H and 8.01 (s, lH, NH).

---

†Department of Chemistry, M.L.S. University, Udaipur-313 001, India.



4a: $R_1 = R_2 = R_3 = H$
4b: $R_1 = R_3 = H, R_2 = OCH_3$
4c: $R_1 = R_2 = OCH_3, R_3 = H$
4d: $R_1 = R_2 = R_3 = OCH_3$
4e: $R_1 = R_3 = H, R_2 = Cl$
4f: $R_1 = R_3 = H, R_2 = NMe_2$

Fig. 1

The melting points reported are uncorrected and were taken in open capillaries. The IR spectra were recorded on Perkin-Elmer spectrophotometer using KBr (cm^{-1}). PMR spectra were taken on Bruker-DRX 300 spectrometer and mass spectra were taken on JEOL-SX 100 mass spectrometer. The purity of synthesized compounds was checked by TLC using silica gel-G. The reaction was carried out in a domestic microwave oven (Samsung model M1630N, output energy 600 watts, frequency 2450 MHz).

Synthesis of 2-(hydroxy ethyl)-benzimidazoles (2): In a flask with a loose stopper *o*-phenylene diamine (0.1 mol) and lactic acid (0.1 mol) were placed. The reaction mixture was irradiated at 300 watt power of microwave for 3 min. It was then cooled and 10% KOH solution was added to it with stirring till the mixture became alkaline to litmus. The separated solid was filtered, washed with ice-cold water and crystallized with ethanol to get **2**.

Synthesis of 2-acetyl benzimidazol (3): It was prepared by literature method[10, 11].

Synthesis of benzimidazolyl chalcones (4): To a solution of compound (3) (0.01 mol) in a conical flask added 30% KOH solution (10 mL) and aromatic aldehyde (0.012 mol) with stirring. The mixture was irradiated in microwave oven for 5 min with occasional disruption. It was then cooled and poured in ice-cold water. The separated solid was filtered, washed well with water and neutralized with dilute CH_3COOH. It was then filtered and crystallized with ethanol to get compounds **4a–f** in 70–80% yield. **4a:** m.p. 205°C; IR (KBr, cm^{-1}) 3249 (NH), 3064 ν(—CH *str.*), 1658–1590ν (combined vibration C=O and C=N); mass BP m/z 248 for $C_{16}H_{12}N_2O$ (248). **4b:** m.p. 225°C; IR (KBr, cm^{-1}) 3262ν (NH), 3050 ν(—CH *str.*), 1635–1580 ν(combined vibration C═O and C═N); NMR δ 3.60 (S, 3H, OCH_3), 6.89 (d, 2H, CH═CH), 7.66–7.85 (Ar-H) and 8.01 (s, lH, NH); mass BP m/z 278 for $C_{17}H_{14}N_2O_2$ (279). **4c:** m.p. 212°C, IR (KBr, cm^{-1}) 3261 ν(NH), 2819 ν(—CH *str.*), 1580–1513ν (combined vibration C═O and C═N); mass BP m/z 307 for $C_{18}H_{16}N_2O_3$ (308). **4d:** m.p. 217°C, IR (KBr, cm^{-1}) 3246 ν(NH), 2937 ν(—CH *str.*), 1651–1578 ν(combined vibration C═O and C═N); mass BP m/z 339 for $C_{19}H_{18}N_2O_4$ (338). **4e:** m.p. 224°C, IR (KBr, cm^{-1}) 3276 ν(NH), 3060 (—CH *str.*), 1659–1596 ν(combined vibration C═O and C═N); mass BP m/z 283 for $C_{16}H_{11}N_2OCl$ (282.5). **4f:** m.p. 260°C, IRν ν(KBr, cm^{-1}) 3258 ν(NH), 2925 ν(—CH *str.*), 1650–15ν65 ν(combined vibration C═O and C═N); mass BP m/z 291 for $C_{18}H_{17}N_3O$ (291).

ACKNOWLEDGEMENTS

The authors are thankful to the Director CDRI Lucknow for analytical and spectral analysis and U.G.C. New Delhi for financial assistance to YKS.

REFERENCES

1. H. Norbrt, U. Ries, H. Priepke and G. Mehn, *Chem. Abstr.*, **132**, 137386 (2000).
2. N.M. Gaur, S.V. Patil, V.K. Maurya and S.B. Wagh, *Chem. Abstr.*, **133**, 150502 (2001).
3. B.V. Kumar and V.M. Reddy, *Indian J. Chem.*, **24B**, 928 (1985).
4. O.N. Preston, *Chem. Rev.*, **74**, 279 (1974).
5. W.B. Ciegar and J.E. Conn, *J. Am. Chem. Soc.*, **67**, 112 (1945); D.H. Mariane, P.B. Russel and A.R. Todd, *J. Chem. Soc.*, 1419 (1947); D.N. Dhar, Chemistry of Chalcones, Wiley, New York (1981).
6. S.S. Misra and B. Nath, *Indian J. Appl. Chem.*, **34**, 260 (1971); R. Kuhn and H.R. Hansal, *Chem. Ber.*, **86**, 1333 (1953); S.S. Misra, *J. Indian Chem. Soc.*, **50**, 355 (1973); R. Aries, *Chem. Abstr.*, **80**, 146152 (1974).
7. S. Caddick, *Tetrahedron*, **51**, 10403 (1995); M. Kidwai and P. Kumar, *J. Chem. Res. (S)*, 254 (1994); M. Kidwai, Y. Goel, P. Kumar and K. Kumar, *Indian J. Chem.*, **36B**, 135, 782 (1997); K. Mogilaiah and Ch. Srinivas Reddy, *Indian J. Chem.*, **43B**, 2010 (2004).
8. M.A. Phillip, *J. Chem. Soc.*, 2393 (1928); R.I. Augustine (Ed.), Oxidation Technique and Application in Organic Synthesis, Marcel-Dekker, New York, Vol. 1, p. 215 (1960).
9. H. Zellnev, G. Zellnev, F. Kopple and J. Dirubergor, *Monatsch Chem.*, **98**, 643 (1967).
10. Fr. Pat. 81811937; *Chem. Abstr.*, **32**, 2251 (1938).
11. K. Ramaiah, J.S. Grosert, D.L. Hooper, P.K. Dubey and J. Ramnathan, *J. Indian Chem. Soc.*, **76**, 140 (1999).

(*Received*: 26 April 2005; *Accepted*: 7 November 2005) AJC-4482