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ORIGINAL ARTICLE

Three component, one-pot synthesis of amidoalkyl naphthols using polyphosphate ester under solvent-free conditions

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KEYWORDS

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Abstract Amidoalkyl naphthols are synthesized via a simple, one-pot, three-component reaction between aldehydes, 2-naphthol and amides or ureas using polyphosphate ester (PPE) as a reaction mediator under solvent-free conditions in good to excellent yields. High yields, short reaction time, easy work-up, elimination of solvents and toxic catalysts are the advantages of this procedure.

1. Introduction

Multi-component reactions (MCRs), are one-pot processes in which three or four easily accessible components react to form a single product, which incorporates essentially all the carbon atoms of the starting materials (Tietze, 1996; Ramon and Yus, 2005; Zhu, 2003). MCRs are a promising and vital field of chemistry because the synthesis of complicated molecules can be achieved in a very fast, efficient and time saving manner without the isolation of any intermediate (Menendez, 2006; Devi and Bhuyan, 2004). There has been tremendous development in three or four component reactions specially the Bigenilli (Prajapati and Sandhu, 2004; Shimokawa et al., 2004), Ugi (Domling and Ugi, 2000; Cristau et al., 2001), Passerini (Bossio et al., 1996) and Mannich (Akiyama et al., 2004; Zhao et al., 2004) reactions which have further led to renaissance of MCRs. Nevertheless, development and discovery of new MCRs is still in demand.

1-amidoalkyl 2-naphthols can be converted to useful and important biological building blocks and to 1-aminoalkyl 2-naphthols by an amide hydrolysis reaction, since compounds exhibit depressor and bradycardia effects in humans (Szatmari and Füloop, 2004; Shen et al., 1999). Moreover this 1-aminoalkyl alcohol-type ligand has been used for asymmetric synthesis and also as a catalyst (Hulst et al., 1996; Li et al., 1999).

A literature search revealed that many procedures have been developed to the preparation of biologically important amidoalkyl naphthols in the presence of various catalysts such as *p*-toluenesulfonic acid (Khodaei et al., 2006), montmorillonite K10 clay (Kantevari et al., 2007), Iodine (Das et al., 2007),

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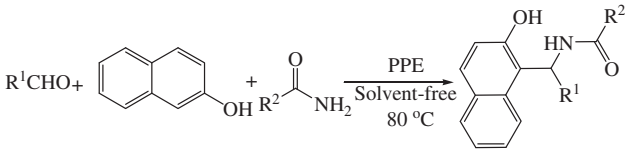

Scheme 1

$K_5CoW_{12}O_{40}{\cdot}3H_2O$ (Nagarapu et al., 2007), Sulfamic acid (Patil et al., 2007), $Yb(OTf)_3$ in ionic liquid (Kumar et al., 2006), TMSCl/NaI (Sabitha et al., 2010), $InCl_3$ (Chavan et al., 2010), P_2O_5 (Nandi et al., 2009), $H_3MoO_{40}P$ (Gawand et al., 2009). However, the combination of solvents, toxic reagents and long reaction times makes some of these methods environmentally hazardous.

Furthermore, the yields of the corresponding amidoalkyl naphthols are not always satisfactory. Because of the importance of these compounds, a simple, general and efficient procedure for the synthesis of these compounds is required.

In continuation of our work (Khosropour et al., 2005; Khodaei et al., 2006; Foroughifar et al., 2008, 2009, 2010) under the framework of "Green chemistry" we have developed an environmentally benign solvent-free approach for the synthesis of amidoalkyl naphthols from aldehydes, 2-naphthols and amides or ureas using polyphosphate ester (PPE) under solvent-free conditions (Scheme 1).

Polyphosphate ester (PPE) is one of the phosphate-based reagents that are related to polyphosphoric acid (PPA). It differs from PPA in that it is aprotic and soluble in organic media. It is often compared to the related polyphosphoric acid trimethylsilyl ester (PPSE). In general, the advantages of PPE include its solubility in organic solvents, the mild conditions under which it is used, and its relatively nonhazardous, nonnoxious nature. Although the reagent is not commercially available it is readily prepared by treatment of phosphorous oxide with anhydrous ether (Dixon, 1995).

2. Experimental

2.1. General

All of the products are known compounds and were identified by their physical and spectroscopic data with those reported in the literature. Melting points were measured by using capillary tubes on an electro thermal digital apparatus. IR spectra were recorded as KBr disc on a galaxy series FT-IR 5000 spectrometer. NMR spectra were recorded on a brucker spectrometer in DMSO-d_6 with TMS as an internal standard.

2.2. General procedure

To a mixture of aldehyde (1 mmol), 2-naphthol (1 mmol) and amide/urea (1.1 mmol), PPE (100 mg) were added. The reaction mixture stirred magnetically at 80 °C for appropriate time as shown in Table 1. The progress of the reaction was monitored by thin layer chromatography. After completion of the reaction, the reaction mixture was cooled to room temperature, washed with water, and the residue was recrystallized from ethanol.

2.3. Representative spectral data

Table 1 entry 4; IR (KBr) (ν_{max}): 3405, 3260–2420, 1639, 1584, 1525, 1419, 1345, 1015, 946, 745 cm^{-1}. NMR (DMSO-d_6) 2.02 (s, 3H), 7.70–7.15(m, 6H), 8.10–7.80 (m, 5H), 8.65 (d, J = 8.0 Hz, 1H), 10.22 (s, 1H).

Table 1 entry 5; IR (KBr) (ν_{max}): 3400, 3250–2400, 1625, 1567, 1528, 1426, 1345, 1022, 940, 820, 744 cm^{-1}. NMR

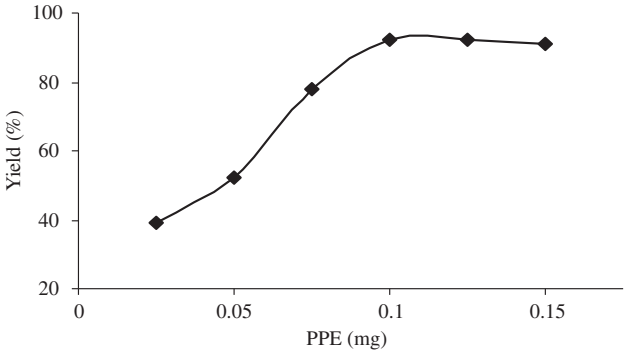

Figure 1 Effect of PPE on the reaction of benzaldehyde, 2-naphthol and acetamide (reactions carried out at 80 °C for 15 min).

Table 1 Synthesis of amidoalkyl naphthols in the presence of polyphosphate ester (PPE).

Entry	Aldehyde ($R^{1)}$	Amide (R^2)	Time/min	Yield (%)[a]	M.P. (lit. m.p.) (°C)
1	C_6H_5	CH_3	15	91	240–242 (241–243, Nagarapu et al., 2007; Shaterian et al., 2008)
2	2-ClC_6H_5	CH_3	20	88	205–206 (206–207, Nandi et al., 2009; Shaterian et al., 2008)
3	4-$CH_3C_6H_5$	CH_3	15	90	221–223 (222–223, Shaterian et al., 2008)
4	3-$NO_2C_6H_5$	CH_3	10	92	254–255 (255–256, Su et al., 2008; Shaterian et al., 2008)
5	C_6H_5	C_6H_5	10	90	232–234 (234–236, Patil et al., 2007; Kantevari et al., 2007)
6	2-$NO_2C_6H_5$	C_6H_5	15	85	263–255 (266–267, Nandi et al., 2009)
7	4-$CH_3OC_6H_5$	C_6H_5	20	87	206–207 (206–208, Nandi et al., 2009)
8	C_6H_5	NH_2	15	89	173–175 (172–174, Patil et al., 2007; Nagawade and Shinde, 2007)
9	4-BrC_6H_5	NH_2	20	88	171–173 (170–172, Patil et al., 2007)
10	4-ClC_6H_5	NH_2	15	91	168–170 (168–169, Patil et al., 2007; Nagawade and Shinde, 2007)
11	C_6H_5	CH_3NH	10	90	192–193 (190–191, Patil et al., 2007)
12	3-$NO_2C_6H_5$	CH_3NH	10	93	192–193 (191–192, Patil et al., 2007; Khodaei et al., 2006)
13	4-ClC_6H_5	$CH_3CH{=}CH$	10	89	214–216 (213–215, Patil et al., 2007; Khodaei et al., 2006)
14	3-$NO_2C_6H_5$	$CH_3CH{=}CH$	10	91	197–199 (195–196, Patil et al., 2007; Zhang and Zhang, 2009)

[a] Isolated yields.