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RESEARCH ARTICLE

Synthesis of 1,3-dihydro-4,5-(disubstituted phenyl)-2-imidazolones.

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ABSTRACT
2-imidazolones have gained immense significance in human life due to variety of their applications. Though there are several methods for the synthesis of 2-imidazolones, most of them required longer reflux time of 8 to 10 hours. Hence the proposed work was undertaken to workout simple methodology for the synthesis of 2-imidazolones and to improve the yield of the products, by employing Zeolite as a catalyst. The work presented here describes the synthesis of some substituted 2-imidazolones obtained from substituted benzoins and urea in CH_3COOH as a solvent in presence of Zeolite as a catalyst. Substituted benzoins in turn were obtained from aromatic aldehydes by their condensation in presence of aqueous NaCN. The characterisation of synthesized compounds was made on the basis of chemical properties, elemental and spectral analysis.

Keywords: Substituted benzoins, Urea, Methyl urea, Phenyl urea, Zeolite catalyst, 2-imidazolones

INTRODUCTION

Imidazolones are believed to be associated with several pharmacological activities. Many natural products are believed to contain imidazolones. The leucetta and oroidin families of alkaloids {1] have been identified which contain either 2-aminoimidazole or 2-imidazolone moiety [2-3]. Stoffel and speziale [4] described the preparation of 2-imidazolones by a novel ring closure of propynylureas with phosphorous pentachloride. 2-imidazolone was obtained via a stable isolable imidazolium chloride. AlNashef [5] reported reaction of superoxide ion with alkyl imidazolium cations of imidazolium based ionic liquids at room temperature and atmospheric pressure to give the corresponding 2-imidazolones in good yield.

Marie Pascale [6] synthesized several 2-imidazolone derivatives and screened their fungicidal and herbicidal activities. Xue et al [7] synthesized 2,3-Dihydro-*N*,3-bis(3,4,5-trimethoxyphenyl)-4-(substitutedphenyl)-2-oxo-imidazole-1-carboxamides and 1-acetyl-1,3-dihydro-3-(3,4,5trimethoxyphenyl)-4-(substitutedphenyl)-2*H*-imidazol-2-ones and reported their antitumor activities. Glass et al [8] reported 4-(4-Guanidinobenzoyl)-2-imidazolones and related compounds having phosphodiesterase inhibitors and novel cardio tonics with combined histamine H_2 receptor agonist and PDE 111 inhibitor activity. Butler and Hussain [9] carried out synthesis of 2- imidazolones by the reaction of benzoins or aliphatic acyloins with urea and methyl urea. Sang-Hyeup Lee and coworkers [10] carried out synthesis of 2-imidazolones by the reaction of substituted urea with 3-hydroxy butanone or 3-iminopentane 2,4-dione in solution or in solid phase. From the review of literature, it was observed that most of the methods of synthesis of 2-imidazolones required longer reflux time of 8-10 hours and the yield of the products was also quite low. Hence, in the context of the above observations, the proposed work was undertaken to reduce the reflux time and to improve the yield of the products by employing Zeolite as a catalyst.

EXPERIMENTAL

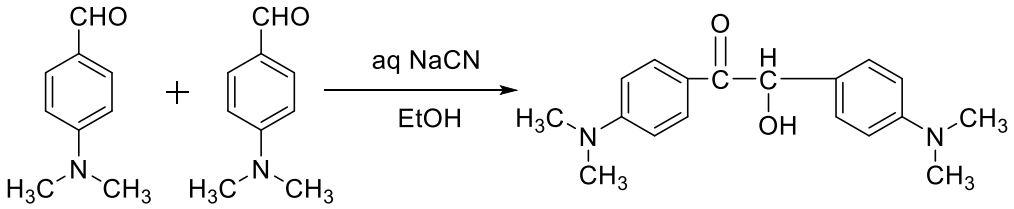
In this work, three substituted benzoins were prepared by the self-condensation of 4-dimethyl amino-benzaldehyde, 4-methoxybenzaldehyde and 2-hydroxybenzaldehyde respectively, in presence of aqueous NaCN in ethanolic medium. In the second step, each of above mentioned benzoins was reacted with urea, methyl urea and phenyl urea respectively in CH_3COOH in presence of Zeolite as a catalyst to form 1,3-dihydro-4-(4-substituted phenyl)-5-(4-substituted phenyl)-2-imidazolones and their methyl and phenyl derivatives respectively. All the synthesized compounds were characterized on the basis of chemical properties, elemental and spectral analysis.

Scheme-1 : Preparation of 4,4'-di-dimethylamino-benzoin

4-Dimethylamino benzaldehyde (0.05mol) was dissolved in ethyl alcohol in a round bottom flask. To this solution, added aqueous solution containing 2.00gms of NaCN. The solution was refluxed for one hour. It was allowed to cool and poured to ice cold water with vigorous stirring when faint violet solid product was obtained. It was isolated and washed 2-3 times with cold water-alcohol mixture and recrysatllised from ethanol.

Yield : 68%

Melting point : 138^{0}C



4, 4'-didimethylaminobenzoin (1a)

IR (KBr,cm-1) : 3480 (O-H str); 3047 (Ar,C-H str); 2908 (Aliph,C-H); 2816 (C-H str in methyl group); 1678 (C=O); 1543 (Ar-H,C=C); 1234 (C-O str);

^{1}H-NMR (DMSO) (δ) : 7.66 (d,4H,Ar-H); 6.70 (d,4H,Ar-H); 3.64 (s,1H,CH-OH); 3.04 (S,12H,N(CH_3)$_2$); 2.55(s,1H,Aliph.C-H)

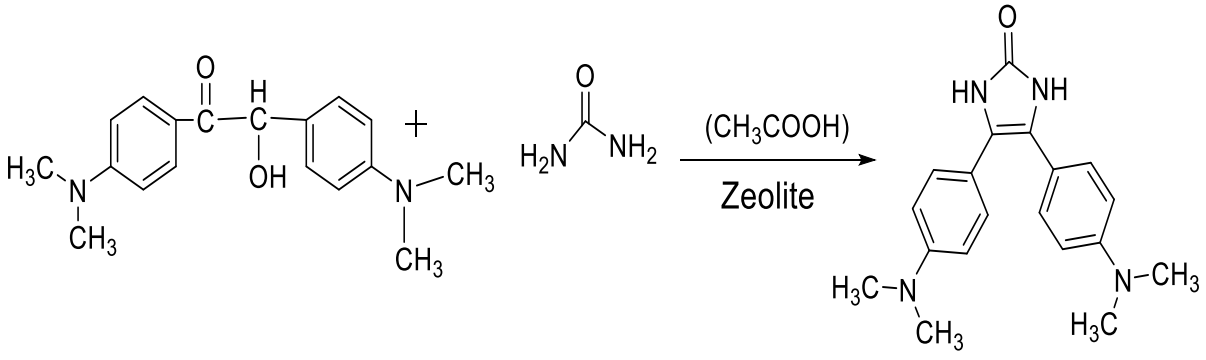
Elemental Analysis for $C_{18}H_{22}N_2O_2$ (298.39)

Calculated C, 72.46; H, 7.43; N, 9.39;

Found C, 72.42; H, 7.40; N, 9.35;

Scheme 2 : Synthesis of 1,3-dihydro-4-(4-dimethylaminophenyl)-4-(4-dimethylaminophenyl) -2-imidazolone.
Dissolved 4,4'-dimethylaminobenzoin (0.01mol) in glacial acetic acid (20ml). To this solution, added urea (0.1mol) followed by Zeolite (1.00gm) as a catalyst. The reaction mixture was reflux for two and half hours. It was allowed to cool and poured to ice cold water and recrystalllised from ethanol.

Yield: 65% Melting point: 165^{0}C



(2a)

IR (KBr,cm-^{1}) : 3317 (N-H str); 3182 (Ar,C-H str); 2912 (C-H str in methyl group); 1674 (C=O); 1450 (Ar-H,C=C); 1377 (C-N str);
^{1}H-NMR (DMSO) (δ) : 9.66(s,2H,-NH); 7.66 (d,4H,Ar-H); 6.74 (d,4H,Ar-H); 3.06 (S,12H,N(CH_3)$_2$);

Elemental Analysis for $C_{19}H_{22}N_4O$ (322.41)
Calculated C, 70.78; H, 6.88; N, 17.38;
Found C, 70.76; H, 6.85; N, 17.35;

RESULTS AND DISCUSSION

We synthesized nine variedly substituted -2-imidzolones by the condensation of each of three substituted benzoins with urea, methyl urea and phenyl urea respectively. The target compounds gave positive tests for Nitrogen as well as for C=O linkage (red coloration with 1% solution of m-dinitrobenzene in ethanol) The IR spectrum showed sharp bands at 3317cm^{-1} (N-H starching) and 1674 cm^{-1} (C=O starching) and 1450 cm^{-1} (Ar, C=C starching) similarly, in ^{1}H-NMR spectrum chemical shifts at 9.66ppm (s,2H,-NH); 7.66ppm (d,4H,Ar-H); 6.74ppm (d,4H,Ar-H); 3.06 (S,12H,N(CH_3)$_2$) with elemental analysis further confirmed the formation 2-imidazolones. The synthesized compound along with their percent yield and melting point are given in the following table.

Table: List of synthesized compounds along with their % yield and melting point

Sr. No	Compound	**Percent Yield (%)**	**Melting point (°C)**
1	1, 3-dihydro-4-(4-dimethylaminophenyl)-5-(4-dimethylaminophenyl)-2-imidazolone.	65	165
2	1-methyl-3-H-4-(4-dimethylaminophenyl)-5-(4-dimethylaminophenyl)-2-imidazolone	68	160
3	1-phenyl-3-H-4-(4-dimethylaminophenyl)-5-(4-dimethylaminophenyl)-2-imidazolone.	70	175
4	1, 3,-dihydro-4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone.	66	145
5	1-methyl-3-H--4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone.	62	180
6	1-phenyl-3-H -4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone.	58	138
7	1, 3-dihydro-4-(2-hydroxyphenyl)-5-(2-hydroxyphenyl)-2-imidazolone	66	190
8	1-methyl-3-H-4-(2-hydroxyphenyl)-5-(2-hydroxyphenyl)-2-imidazolone	62	168
9	1-phenyl-3-H-4(2-hydroxyphenyl)-5-(2-hydroxyphenyl)-2-imidazolone	63	185

CONCLUSION

Thus, we could succeed in synthesizing variedly substituted-2-imidazolone with simple and easy to workout methodology. Use of Zeolite as a catalyst enabled us rapid route for the synthesis of 2-imidazolones which could reduce reflux time to as low as two and half hours. The catalyst is insoluble in solvent due to which isolation of the product became much easy.

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