

π-Extended Systems Based on Tetracyclic Benzo [4,5] Imidazo [2',1':6,1] Pyrido[2,3-D] Pyrimidines



Harutyunyan Artur A^{1,2*}, Ghukasyan Gohar T¹ and Danagulyan Gevorg G^{1,2}

¹Russian Armenian (Slavonic) University, Yerevan, Armenia, Europa

²Scientific and technological center of organic and pharmaceutical chemistry, Yerevan, Europa

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*Corresponding author: Harutyunyan Artur A, Russian Armenian (Slavonic) University, Yerevan, Armenia, Europa

Introduction

Recently, we have developed a fundamentally new method of constructing the derivatives of the heterocyclic system of benzo[4',5'] imidazo[2',1':6,1]pyrido[2,3-d]pyrimidine [1], which allowed the introduction of methyl and methylene into the molecule groups. It should be noted that some derivatives of the indicated heterocyclic system revealed antibacterial and ant monoamine oxidase properties [2,3].

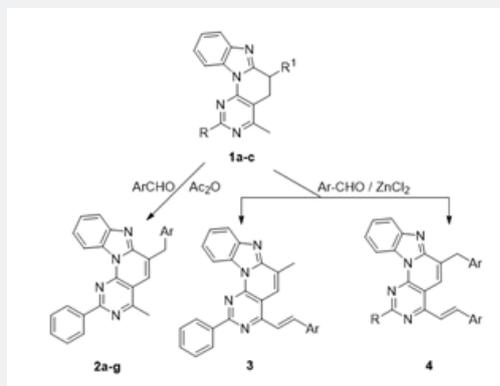


Figure 1: 1a-c: R,R1 = Ph, H (a); Ph, Me (b); OH, H (c). 2a-g: Ar = Ph (a), 2,4-Cl₂C₆H₃(b), 2-AcO-naphthalen-1-yl (c), 4-(OCH₂C₆H₅) C₆H₄ (d), 4-MeOC₆H₄ (e), thiophen-2-yl (f), CH=CHPh (g). 3,4: R = Ph, OH; Ar = 4-ClC₆H₄, 4-NO₂C₆H₄.

Taking into account, we have developed possibilities for the further functionalization of the class of compounds under discussion, in particular, for obtaining new derivatives on their basis with a lengthy system of conjugated π-bonds that are interesting in terms of biomedical and technical applications. In this context, we developed the reactions of 4-methyl-2-phenyl-5,6-dihydrobenzo[4',5'] imidazo [2',1':6,1] pyrido[2,3-d] pyrimidine 1a, (RS)-4,6-dimethyl-2-phenyl-5,6-dihydrobenzo [4',5'] imidazo[2',1':6,1] pyrido[2,3-d] pyrimidine

1b and 4-methyl-5,6-dihydrobenzo [4',5'] imidazo[2',1':6,1]pyrido[2,3-d]pyrimidin-2-ol 1c with aromatic aldehydes in various experimental conditions. This allowed us to obtain benzo [4',5'] imidazo[2',1':6,1]pyrido[2,3-d] pyrimidine derivatives, combining two structural features: an extended π-conjugated chain and a bulky aromatic substituent in the side chain, according to the scheme: (Figure 1).

It was found that under boiling conditions of the starting tetracycle 1a, even with an excess of aromatic aldehyde in acetic anhydride, the reaction proceeds exclusively with the formation of 6-arylmethyl-4-methyl derivatives 2a-g, which is the result of a 1,3-prototropic shift of the 6-benzylidene derivative originally formed and moving double bond from exocyclic position to ring. The signal of the proton H5, which manifests itself in the 7.87 ppm region, characteristic of aromatic structures and not characteristic of protons of the olefinic double bond, is in favor of the intracyclic position of the double bond. Under more severe conditions, namely, with the simultaneous heating of heterocycles 1b or 1c with 4-chloro- and 4-nitrobenzaldehydes in the presence of ZnCl₂, either only 4-methyl group is involved in the condensation reaction to form 4-styryl derivative 3, or both reaction centers to form 4-styryl-6-(4-chloro, nitro) benzyl derivatives 4. The specified course of the reaction was confirmed by IR, NMR 1H and 2D NOESY spectroscopy and X-ray crystallography.

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