SYNTHESIS OF 1,3,4,5-SUBSTITUTED 1,3-DIHYDRO-2H-IMIDAZOL-2-ONES

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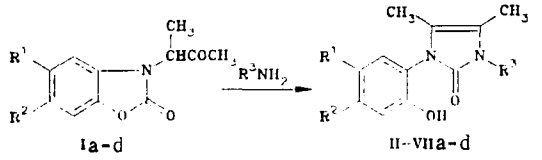
It was shown that 3-(1-methyl-2-oxopropyl)-2-benzoxazolones react with primary amines to form the corresponding 1,3-dihydro-2H-imidazol-2-ones in good yields.

It is known that substituted 1,3-dihydro-2H-imidazol-2-ones have various types of physiological activity [1-3]. In particular, they exhibit high herbicidal activity [4].

We therefore carried out the synthesis of certain novel 1,3-dihydro-2H-imidazol-2-ones, using 3-(1-methyl-2-oxopropyl)-2-benzoxazolones previously described by us [5] as starting materials [1].

The present article describes part of our investigations on the reaction of 3-(oxoalkyl)-2-benzoxazolones with nucelophilic agents [6].



Ia-d II-VIIa-d

II—VII a $R^1=R^2=H$; b $R^1=Cl$; $R^2=H$; c $R^1=H$, $R^2=Cl$; d $R^1=H$, $R^2=Br$; II $R^3=CH_3$, III $R^3=C_2H_5$; IV $R^3=C_3H_7$; V $R^3=C_4H_9$; VI $R^3=CH_2C_6H_5$; VII $R^3=C_6H_5$

We found that by the action of nucleophilic agents, such as primary amines, the reaction proceeds with the formation of several intermediate compounds, the alkylamino group of which attacks the carbonyl carbon atom of the benzoxazole ring, as the result of which 1,3-dihydro-2H-imidazol-2-ones are formed.

TABLE 1. 1,3-Dihydro-2H-imidazol-2-ones

Compound	mp, °C	Found N, %	Empirical formula	Calculated N, %	Yield, %
IIa	179...180	12.8	$C_{12}H_{14}N_2O_2$	12.8	55
IIIa	168...169	11.9	$C_{13}H_{16}N_2O_2$	12.7	63
IVa	141...142	11.2	$C_{14}H_{18}N_2O_2$	11.4	57
Va	139...140	10.8	$C_{15}H_{20}N_2O_2$	10.8	68
VIa	163...164	9.4	$C_{18}H_{18}N_2O_2$	9.5	62
VIIb	210...211	9.8	$C_{17}H_{16}N_2O_2$	10.0	76
IIb	222...223	11.0	$C_{12}H_{13}ClN_2O_2$	11.0	78
IIIb	205...206	10.4	$C_{13}H_{15}ClN_2O_2$	10.4	80
IVb	132...133	9.7	$C_{14}H_{17}ClN_2O_2$	10.0	70
Vb	152...153	9.4	$C_{15}H_{19}ClN_2O_2$	9.5	73
VIb	193...194	8.8	$C_{18}H_{17}ClN_2O_2$	8.5	67
VIIb	205...206	9.2	$C_{17}H_{15}ClN_2O_2$	8.9	77
IIc	190...191	10.9	$C_{12}H_{13}ClN_2O_2$	11.0	78
IIIc	182...183	10.4	$C_{13}H_{15}ClN_2O_2$	10.4	73
IVc	183...184	9.9	$C_{14}H_{17}ClN_2O_2$	10.0	69
Vc	149...150	9.2	$C_{15}H_{19}ClN_2O_2$	9.5	51
VIc	211...212	8.4	$C_{18}H_{17}ClN_2O_2$	8.5	55
VIIc	206...207	8.7	$C_{17}H_{15}ClN_2O_2$	8.9	41
IId	226...227	9.3	$C_{12}H_{13}BrN_2O_2$	9.4	73
IIId	160...161	9.1	$C_{13}H_{15}BrN_2O_2$	9.0	78
IVd	188...189	8.9	$C_{14}H_{17}BrN_2O_2$	8.6	72
Vd	164...165	8.2	$C_{15}H_{19}BrN_2O_2$	8.3	57
VId	226...227	7.7	$C_{18}H_{17}BrN_2O_2$	7.5	66
VIId	220...221	7.8	$C_{17}H_{15}BrN_2O_2$	7.8	59

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In the IR spectra of compounds II, in addition to the stretching vibration bands of the carbonyl group of the imidazolone ring at 1670-1665 cm^{-1}, bands of the double bond at 1650-1640 cm^{-1} were observed.

In the PMR spectrum of compound IId (in DMSO-D_6) besides the characteristic group of signals of three aromatic protons, a singlet of three methyl groups (1.97, 2.25, and 3.27 ppm) and a singlet of the hydroxyl proton at 10.48 ppm were observed. When the spectrum was run in a D_2O solution the latter singlet disappeared.

EXPERIMENTAL

The IR spectra were run on a UR-20 spectrophotometer in KBr tablet, and the PMR spectra on a BS-487C spectrometer (80 MHz) in a DMSO-D_6 solution, using TMS as a standard. The melting points were determined by the capillary method, and were not corrected.

The characteristics of the compounds are given in Table 1.

1,3-Dihydro-2H-imidazol-2-ones (IIa-d-VIIa-d). A 30-mmole portion of the corresponding amine is added to a solution of 10 mmoles of compound I in 20 ml of ethanol. The mixture is boiled for 2-6 h, the precipitate is separated, washed with water, and recrystallized from 70% aqueous ethanol.

The reaction with aniline is carried out in the absence of ethanol. The reaction mixture is cooled and neutralized with 10% hydrochloric acid.

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