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General regioselective synthesis and crystal structure of racemic 5-substituted 2,2-dimethyl-3-hydroxyimidazolidin-4-ones

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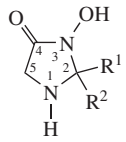
The cyclocondensation of racemic valine, leucine and β-phenylalanine hydroxamic acids with acetone regioselectively affords corresponding cyclic hydroxamic acids; the crystal structure of the 5-isobutyl derivative was determined by X-ray diffraction analysis and compared with that of its 5-methyl homologue.

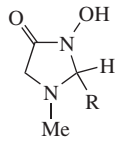
Hydroxamic acids [HAs, RC(O)N(R′)OH] possess important pharmacological properties,[1] function as NO donors[2] and good bioligands for vital metals[3] (siderophores, metalloenzyme inhibitors, *etc*.).

It is known that reactions of the simplest α-amino hydroxamic acid (glycine hydroxamic acid, GlyHA) with aliphatic or aromatic aldehydes (EtOH, Δ, 2–4 h)[4] and ketones (MeOH, Δ, 1 h)[5,6] form 3-hydroxyimidazolidin-4-one derivatives **1** (yield, 70–90%)[4] and **2** (27–57%),[5,6] respectively. Cyclic hydroxamic acids (CHAs) could also be formed in the reactions of β- and α-methyl substituted derivatives of GlyHA, namely, sarcosine hydroxamic acid (SarHA) with aldehydes (EtOH, 2–4 h, **3**, 70–90%)[4] and DL-α-alanine hydroxamic acid (DL-AlaHA) with acetone (Δ, 3 h, **4**, 83%),[7] respectively. Extensively, the analogous regioselective N,N′-cyclocondensation also occurred in the reactions of β-AlaHA with aldehydes and ketones (MeOH, Δ, 3–6 h, **5**, 26–74%).[8]

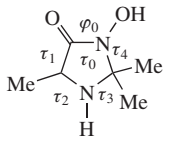
On the other hand, products obtained by the reactions of bulky substituted α-amino HAs [*e.g.*, DL-leucine HA (DL-LeuHA) and DL-β-phenylalanine HA (DL-PheHA)] with aldehydes (benzene + EtOH, Δ, 1 h) are assigned to six-membered cyclic hydroxamate structures **6** and **7** (10–45%)[9] that implies regioselective N,O-cyclocondensation.



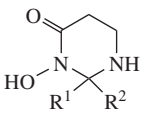
(±)-**1** R^1 = H, R^2 = Alk, Ar
(±)-**2** R^1, R^2 = Alk, Ph



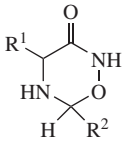
(±)-**3** R = H, Alk, Ar



(±)-**4**



5 R^1 = H, Alk, Ar
R^2 = Alk, Ar



6 R^1 = CH_2CHMe_2, R^2 = Ar
7 R^1 = CH_2Ph, R^2 = Alk, Ar

As for the mode of cyclocondensation regioselectivity (N,N′ or N,O) for reactions of sterically hindered α-amino HAs with aliphatic ketones, here we studied the reactions of DL-ValHA (**8**), DL-LeuHA (**9**) and DL-PheHA (**10**) with acetone, as its symmetry avoids the formation of diastereomeric products. In addition, we compared acids **2a** (R^1 = R^2 = Me)[5] and **4**[7] earlier characterized by an X-ray method.

The reactions of bulky α-amino HAs **8**–**10** with acetone gave the corresponding 5-substituted derivatives of 2,2-dimethyl-3-hydroxyimidazolidin-4-one **11**–**13** (Scheme 1),[†] *i.e.,* as in the reactions of ordinary α-amino HAs with aldehydes and ketones,[4–7] the regioselective N,N′-cyclocondensation occurs.

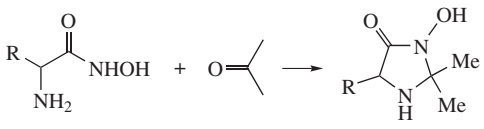
The reactions (Scheme 1) are self-catalytic and proceed under mild conditions (with an excess of ketone) leading to moderate product yields (55–67%).[†] Furthermore, the addition of a small amount of MeOH (1–2 ml), to increase the solubility of reactive zwitterionic form of reactants **8**–**10**, shortened the reaction time to 0.5–1 h. However, the addition of MeOH caused lower yields of **11**–**13** in the range of 15–25% owing to the formation of an oligomeric by-product. The most plausible mechanism for this reaction was discussed earlier.[6]

All compounds were characterized by IR and NMR spectra[†] (*cf.* data for **1**–**4**[4–7]) and elemental analysis. The broad bands observed at 2850–2550 cm^{-1} in the IR spectra (KBr), likewise the downfield resonance signals of ^{1}H NMR spectra (δ_C ~9.5 ppm, [$^2H_6$]DMSO) and the test reactions with ferric chloride confirm the presence of the exocyclic OH group in **11**–**13**. It follows that for reactions considered the formation of type **6**

[†] Reactants **8**–**10** were synthesized according to known procedures.[13]

General procedure for the synthesis of **11**–**13**. Suspension of 5 mmol of **8** (or **9**, **10**) and 30 cm^3 (0.4 mol) of dry acetone was weakly refluxed until the reagent was dissolved (1–2 h) followed by filtration and solvent evaporation in a vacuum. The residue was extracted by hot benzene [or cold CH_2Cl_2 (**12**), hot dioxane (**13**)] and filtered. The filtrate was dried in a vacuum and recrystallized from an appropriate solvent to give a white solid of **11** (or **12**, **13**). Products are soluble in cold MeOH, DMSO or hot acetone, MeCN, benzene and $CHCl_3$.

For characteristics of compounds **11**–**13**, see Online Supplementary Materials.



8 R = Pr^i (DL-ValHA)
9 R = Bu^i (DL-LeuHA)
10 R = Bn (DL-PheHA)

(±)-**11** R = Pr^i
(±)-**12** R = Bu^i
(±)-**13** R = Bn

Scheme 1

and **7** hydroxamates by concurrent N,O-cyclocondensation did not occur.

Moreover, the spectral evidences are not enough to confirm the formation of the corresponding azomethine form but rather the cyclic structure of products **11**–**13**, namely: (i) the chemical shifts of the C(2) atom (δ_C ~75 ppm) in the ^{13}C NMR spectra of **11**–**13** do not correspond to trigonal carbon of azomethine form (δ_C ~125 ppm)[8] but to the tetrahedral carbon of the cyclic form,[4–7] (ii) the presence of the NH group of cyclic structure is confirmed by IR (ν_{NH} ~3250 cm^{-1}) and ^{1}H NMR spectra (δ_{NH}, ~2.8 ppm, [$^2H_6$]DMSO).

The ^{13}C NMR spectra of acids **11**–**13** ($\delta_{C=O}$ 169–172 ppm, CD_3OD), similarly to **1**–**4**,[5–7] do not show the presence of hydroxynitrone tautomer.[10]

The structure of CHA **12** was confirmed by X-ray diffraction analysis (Figure 1).‡

The comparison of crystal structures of homologues **2a**, **4** and **12** shows that acids (±)-**4** [mp 163–164 °C (acetone); space group $P2_1/c$, $Z(Z') = 8(2)$][7] and (±)-**12** [mp 132–133 °C (acetone); C_2/c, $Z(Z') = 16(2)$]‡ form racemic crystals whose asymmetric units contain two crystallographically independent molecules (**A** and **B**), possessing similar geometric parameters (Figure 1), but parent achiral acid **2a** [mp 177–178 °C (acetone)] crystallizes as a conglomerate [$P2_12_12_1$, $Z(Z') = 4(1)$].[5]

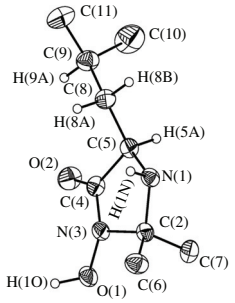


Figure 1 General view of **12** with some hydrogen atoms omitted for clarity [is illustrated by (1*R*,5*R*)-enantiomer].

‡ *Crystallographic data*. Crystals of **12** ($C_9H_{18}N_2O_2$, M = 186.25) are monoclinic, space group $C2/c$ at 153 K: a = 14.8213(4), b = 14.8271(4) and c = 19.4308(5) Å, β = 96.957(1)°, V = 4238.6(2) Å^3, Z = 16 (Z' = 2), d_{calc} = 1.167 g cm^{-3}, μ(MoKα) = 0.83 cm^{-1}, F(000) = 1632. Intensities of 19658 reflections were measured with a Bruker P4 diffractometer [λ(MoKα) = 0.71072 Å, ω-scans, $2\theta < 56°$] and 5087 independent reflections (R_{int} = 0.0247) were used in the further refinement. The structure was solved by a direct method and refined by the full-matrix least-squares technique against F^2 in the anisotropic-isotropic approximation. The hydrogen atoms of OH and NH groups were found in difference Fourier synthesis. The H(C) atom positions were calculated. All hydrogen atoms were refined in an isotropic approximation in a riding model. For **12** the refinement converged to wR_2 = 0.1516 and GOF = 1.001 for all independent reflections [R_1 = 0.0474 was calculated against F for 3204 observed reflections with $I > 2\sigma(I)$]. All calculations were performed using SHELXTL PLUS 5.0 program.

CCDC 766522 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre *via* www.ccdc.cam.ac.uk/data_request/cif. For details, see 'Notice to Authors', *Mendeleev Commun.*, Issue 1, 2010.

The crystal structures of CHAs **2a**, **4** and **12** exhibit the same pattern of intermolecular hydrogen bonding, namely, strong O–H···N$_{sp^3}$ and weak N$_{sp^3}$–H···O=C bonds are formed (Figures 2 and 3). Moreover, in racemic crystals of **4** and **12**, the weak [N(1)–H(1N)···O(2)=C(4)] hydrogen bond connects homochiral molecules into infinite chains, but the strong bond [O(1)–H(1O)···N(1)] couples molecules of opposite handedness.

The packing arrangement of the independent molecules in **12**, however, differs from that in **4**. Checking them in detail, the alternate non-equivalent molecules in a crystal of **12** are connected into heterochiral layer together through, as mentioned above, the weak homochiral N–H···O=C hydrogen bond –$\mathbf{A}_{(S)}$–$\mathbf{B}_{(S)}$– and –$\mathbf{A}_{(R)}$–$\mathbf{B}_{(R)}$– and the strong heterochiral O–H···N$_{sp^3}$ one –$\mathbf{A}_{(S)}$–$\mathbf{B}_{(R)}$– and –$\mathbf{A}_{(R)}$–$\mathbf{B}_{(S)}$– (Figure 2). However, the independent molecules in a crystal of **4** separately form alternate heterochiral layers (sublattices) by the weak homochiral (–$\mathbf{A}_{(S)}$–$\mathbf{A}_{(S)}$– and –$\mathbf{A}_{(R)}$–$\mathbf{A}_{(R)}$– or –$\mathbf{B}_{(S)}$–$\mathbf{B}_{(S)}$– and –$\mathbf{B}_{(R)}$–$\mathbf{B}_{(R)}$–) and the strong heterochiral –$\mathbf{A}_{(S)}$–$\mathbf{A}_{(R)}$– (Figure 3) or –$\mathbf{B}_{(S)}$–$\mathbf{B}_{(R)}$– hydrogen bonds.

At the same time, the independent molecules in crystals of **4** and **12** are similarly arranged by both types of intermolecular

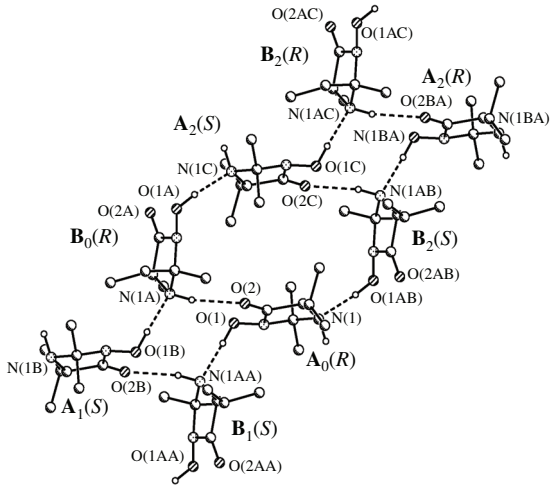


Figure 2 Fragment of the crystal structure of **12**. Hydrogen atoms that do not form H-bonds are omitted for clarify. The H-bonds are shown by dashed lines. The geometric parameters of H-bonds are as follows: O(1A)–H(1OA) 0.96 Å, H(1OA)···N(1C) 1.81 Å, O(1A)···N(1C) 2.764(2) Å, O(1A)–H(1OA)···N(1C) 173°; O(1)–H(1O) 0.94 Å, H(1O)···N(1AA) 1.82 Å, O(1)···N(1AA) 2.767(2) Å, O(1)–H(1O)···N(1AA) 178°; N(1A)–H(1NA) 0.97 Å, H(1NA)···O(2) 2.31 Å, N(1A)···O(2) 3.251(2) Å, N(1A)–H(1NA)···O(2) 166°; N(1)–H(1N) 0.99 Å, H(1N)···O(2BB) 2.30 Å, N(1)···O(2BB) 3.264(2) Å, N(1)–H(1N)···O(2BB) 167°. The symmetry transformation used to generate the atoms from the basic ones: –x, –y + 1, –z + 1 for N(1C); –x + 0.5, –y + 0.5, –z + 1 for N(1AA); x, y + 1, z for O(2); x + 0.5, y – 0.5, z for O(2BB).

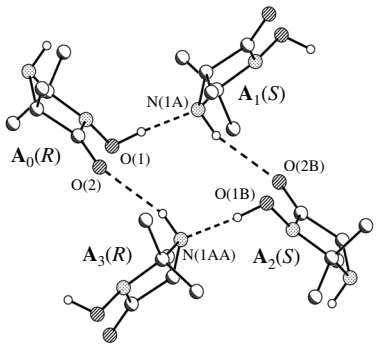


Figure 3 Fragment of the crystal packing of **4** showing the H-bonds within the **A** sublattice. The geometric parameters of the H-bonds are as follows: O(1)–H(1O) 0.94 Å, H(1O)···N(1A) 1.77 Å, O(1)···N(1A) 2.712(3) Å, O(1)–H(1O)···N(1A) 173°; N(1A)–H(1NA) 0.93 Å, H(1NA)···O(2B) 2.26 Å, N(1A)···O(2B) 3.145(3) Å, N(1A)–H(1NA)···O(2B) 159°. The symmetry transformation used to generate the atoms from the basic ones: x, –y + 0.5, z + 0.5 for N(1A); –x + 1, –y + 1, –z + 1 for O(2).

hydrogen bonds into cyclic heterochiral tetramers of two species, namely, as 14-membered [–H(1N)···O=C–N–O–H···N(1)–]$_2$ and 20-membered [–H(1N)···O=C–C(5)–N(1)···H(1O)–N(3)–C(2)–N(1)–H(1N)–]$_2$ rings (Figure 2). However, both kinds of tetramers in a crystal of **12** are formed as $–\mathbf{A}_{(R)}–\mathbf{B}_{(R)}–\mathbf{A}_{(S)}–\mathbf{B}_{(S)}–$ (Figure 2), but those of acid **4** as $–\mathbf{A}_{(R)}–\mathbf{A}_{(S)}–\mathbf{A}_{(S)}–\mathbf{A}_{(R)}–$ (Figure 3) or $–\mathbf{B}_{(R)}–\mathbf{B}_{(S)}–\mathbf{B}_{(S)}–\mathbf{B}_{(R)}–$ associates. Moreover, the corresponding tetramers of **4** and **12** adopt the similar forms.

The increase of the molecule size is accompanied by the weakening of both hydrogen bonds [O(1)···N(1) 2.68 Å and N(1)···O(2) 3.04 Å (**2a**)[5], (**4**) (Figure 3), (**12**) (Figure 2)]. Accordingly, the stretching frequencies of the characteristic O–H and N–H bonds in the IR spectra of these homologues tend to increase {**2a** [3214 cm^{-1} (ν_{NH}); 2719, 2631, 2533 cm^{-1} (ν_{OH})][5], **4** [3240 cm^{-1} (ν_{NH}); 2735, 2583 cm^{-1} (ν_{OH})],[7] and **12** [3251 cm^{-1} (ν_{NH}); 2821, 2730 cm^{-1} (ν_{OH})]}.

The H(1N) hydrogen atom is pseudo-axial [$\varphi_{H(1N)–N(1)–C(2)–N(3)}$ = = 82.3° (**2a**);[5] 80.1° ($\mathbf{A}_0$) and –81.3° ($\mathbf{B}_0$) (**4**);[7] 88.7° ($\mathbf{A}_0$) and 85.1° ($\mathbf{B}_0$) (**12**) (Figure 1)]. It follows that (i) the (1*R*) or (1*S*) absolute configuration of asymmetric amine N(1) atom in homologues **2a**, **4** and **12** is related to the corresponding enantiomeric N- ($\tau_2 > 0$) or S-type ($\tau_2 < 0$)[5,11] form of imidazolidine ring, (ii) the basic independent homochiral molecules ($\mathbf{A}_0$ and $\mathbf{B}_0$) in crystal of **12** can be presented as (1*R*,5*R*) (Figures 1, 2) or (1*S*,5*S*) as compared to those of opposite handedness for **4** [(1*R*,5*R*) (Figure 3) and (1*S*,5*S*)].

According to calculated puckering parameters (pseudorotation angle P and amplitude of puckering τ_m),[5,11] the form of N-type heterocycle of (1*R*,5*R*)-enantiomer **12** [P_N = 48.5°, τ_m = 30.9°, τ_1 = –2.8° ($\mathbf{A}_0$); P_N = 49.4°, τ_m = 31.3°, τ_1 = –2.3° ($\mathbf{B}_0$)] is close to an ideal envelope $_{C(2)}$E (P_N = 54°, τ_1 = 0, $|\tau_0| = \tau_2$, $|\tau_3| = \tau_4 = \tau_m$) (*quod vide*[5]). On the other hand, the ring of (1*R*,5*R*)-**4** [P_N = 39.5°, τ_m = 35.5° ($\mathbf{A}_0$); P_N = 42.3°, τ_m = 34.7° ($\mathbf{B}_0$)] is almost half-chair $^{N(1)}_{C(2)}$T (P_N = 36°, $\tau_0 = \tau_1 < 0$, $\tau_2 = \tau_4 > 0$, $\tau_3 < 0$) and more puckered than that of **12**. Note that the ring of parent molecule **2a** for (1*R*)-enantiomer (P_N = 44.4°, τ_m = 31.4°) is intermediate between ideal $_{C(2)}$E and $^{N(1)}_{C(2)}$T forms[5] and more flattened as compared to that of **4**, but similar to **12**.

This analysis allowed us to conclude that methyl and isobutyl groups at the C(5) atom have an opposite effect upon change of ring skeleton as compared to that of **2a**. Nevertheless, the rings in **2a**, **4** and **12** adopt a quite narrow range of forms.

The form (P) and puckering (τ_m) of **4** and **12** rings are related to their values of a mean ring torsion angle [$\tau_{mid} = \sum\tau_i/5$ = 22.9° (**A**), 22.3° (**B**) (**4**); 19.5° (**A**), 19.6° (**B**) (**12**)] and a mean ring bond angle [$\tau_{mid} = \sum\omega_i^{endo}/5$ = 105.6° (**A**), 105.8° (**B**) (**4**); 106.2° (**A**), 106.1° (**B**) (**12**)].

Conformational changes of ring (from half-chair in **4** to envelope in **12**) also agree well with the higher tendency to non-planarity of the exocyclic hydroxamic moiety [$\varphi^{exo}_{O(1)–N(3)–C(4)–O(2)}$ = 18.6° ($\mathbf{A}_0$), –20.5° ($\mathbf{B}_0$) (**4**); 20.4° ($\mathbf{A}_0$), 21° ($\mathbf{B}_0$) (**12**)] and its conjugated amide bond torsion angle [$\tau_{0\ C(2)–N(3)–C(4)–C(5)}$ = –13.7° ($\mathbf{A}_0$), 15.2° ($\mathbf{B}_0$) (**4**); –16.5° ($\mathbf{A}_0$), –17.1° ($\mathbf{B}_0$) (**12**) (Figure 1)]. Accordingly, the more pronounced pyramidalization of the amide N(3) atom in **12** [$\sum\omega_{N(3)}$ = 348.7° (**A**), 347.8° (**B**)] as compared to that for **4** [$\sum\omega_{N(3)}$ = 351.2° (**A**), 349.5° (**B**)] is observed, corresponding to decrease in the *p*-character of the lone electron pair (LP)[12] of the atom N(3). On the other hand, the decrease in pyramidality degree for the amine N(1) atom in **12** [$\sum\omega_{N(1)}$ = = 323.6° (**A**), 322.3° (**B**)] (Figure 1) as compared to that for **4** [319.8° (**A**), 321° (**B**)] corresponds to increase in the *p*-character of *pseudo-e*-$LP_{N(1)}$ in **12**. As result, the mutual *trans* orientation of $LP_{N(1)}$ and $LP_{N(3)}$ in **12** [$\varphi_{LP_{N(1)}N(1)\cdots N(3)LP_{N(3)}}$ = = 155.4° ($\mathbf{A}_0$), 154.5° ($\mathbf{B}_0$)] increases with respect to that of **4** [150.7° ($\mathbf{A}_0$), –151.7° ($\mathbf{B}_0$)].

According to the ^{1}H NMR spectra, the dominant conformation of the molecule of **12** in solution is consistent with that in a crystal (Figure 1). In particular, the vicinal coupling constant of H(1N) and H(4) protons (^3J = 8.0 Hz, [$^2H_6$]DMSO) corresponds to their *trans* orientation in crystal [$\varphi_{H(1N)–N(1)–C(5)–H(5A)}$ = 149.4° ($\mathbf{A}_0$), 148.7° ($\mathbf{B}_0$)]. Moreover, a sequence of the mutual orientations for each of the methylene protons H(8A) and H(8B) with protons H(5A) and H(9A) [namely, *trans–cis* orientation for H(8A): $\varphi_{H(5A)–C(5)–C(8)–H(8A)}$ = 176° ($\mathbf{A}_0$), 176.4° ($\mathbf{B}_0$); $\varphi_{H(8A)–C(8)–C(9)–H(9A)}$ = –65.6° ($\mathbf{A}_0$), –65.3° ($\mathbf{B}_0$) and *cis–trans* orientation for H(8B): $\varphi_{H(5A)–C(5)–C(8)–H(8B)}$ = –68.5° ($\mathbf{A}_0$), –68.2° ($\mathbf{B}_0$); $\varphi_{H(8B)–C(8)–C(9)–H(9A)}$ = –178.9° ($\mathbf{A}_0$), 179.4° ($\mathbf{B}_0$)] (Figure 1) agrees with a sequence of small and large vicinal couplings constants in the ^{1}H NMR spectrum of **12** in $CDCl_3$ (or in [$^2H_6$]acetone): $^3J_{H(8A),H(5)}$ 10.0 (or 10.1) Hz, $^3J_{H(8A),H(9)}$ 5.2 (5.2) Hz, $^3J_{H(8B),H(5)}$ 3.9 (4.0) Hz, $^3J_{H(8B),H(9)}$ 9.1 (9.0) Hz.

In summary, according to the results of X-ray analysis, IR and NMR spectroscopy, acids **11**–**13**, similarly to **1**–**4**,[5–7] exist in crystal and in solution as a strongly predominant hydroxyamide tautomer. Taking into account published data,[5–7] we concluded that the regioselective N,N′-cyclocondensation is the general tendency for reactions of neutral aliphatic and aromatic α-amino HAs with aliphatic ketones.

Online Supplementary Materials

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mencom.2010.03.014.

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