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STEREOSELECTIVE 1,3-DIPOLAR CYCLOADDITION OF UNSATURATED NITRILES TO ISOQUINOLINIUM YLIDS. MOLECULAR AND CRYSTAL STRUCTURE OF 1,2-trans-3,3-DICYANO-1-CARBAMOYL-2-(3-PYRIDYL)-1,2,3,4-TETRAHYDROBENZO[f]INDOLIZINE

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The reactions of isoquinolinium ylids with arylmethylenemalononitriles proceed highly stereoselectively via a pathway involving synchronous 1,3-dipolar cycloaddition to give 1,2-trans-2-aryl-3,3-dicyano-1-carbamoyl(benzoyl)-1,2,3,4-tetrahydrobenzo[f]indolizines. The regiospecificity and stereospecificity of these reactions were confirmed by the results of PMR spectroscopy and x-ray diffraction analysis.

Reactions involving 1,3-dipolar cycloaddition of pyridinium, quinolinium, and isoquinolinium ylids to unsaturated compounds are finding extensive application in precise organic synthesis, particularly the synthesis of indolizines and related heterocyclic compounds [1-6]. Thus the reactions of azinium ylids with acetylenes have been quite thoroughly studied [1, 6]. The behavior of ethylenes in these transformations has not been adequately studied [2-6], and the determination of their regiospecificity and stereoselectivity is extremely urgent. Evidence for this is provided by the data in [2-5], which were devoted to the determination of the stereospecificity of reactions involving 1,3-dipolar addition of isoquinolinium ylids to acenaphthylene, norbornene, styrene, and some other ethylene derivatives. However, the NMR spectroscopic method used in this case does not make it possible to draw definitive conclusions regarding the stereochemistry of the process, since in the NMR spectra of the resulting tetrahydroindolizines and related cyclic azines the ^3J constants of spin-spin coupling (SSCC) between the hydrogen atoms of the hydrogenated pyrrole fragment have an intermediate value of 4-8 Hz for the cis and trans configurations; it also seems virtually impossible to form a judgment regarding the spatial orientation of the substituents bonded to the pyrrole ring of the condensed azines formed.

Considering what we have stated above, in the present research we studied the reactions of isoquinolinium ylids with arylmethylenemalononitriles and, on the basis of the results of PMR spectroscopy and x-ray diffraction analysis, established their regioselectivity and stereoselectivity. The selection of the subjects of our investigation is explained by the fact that the molecules of the starting compounds are, with respect to the electron-density distribution, highly asymmetrical and have relatively high dipole moments [7, 8]. In this connection, if the reaction in the investigated series were to proceed regio- and stereoselectively, as expected, the reaction should be preceded by, first, an intermolecular reaction (regioelectronic control), which predetermines the regiospecificity of the transformation, and, second, by steric interactions of the substituents in the starting molecules (stereo control), which determine the stereospecificity of the reactions under consideration.

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TABLE 1. Characteristics of the Synthesized IVa-e

Compound	Empirical formula	mp, °C	IR spectrum, cm^{-1}		PMR spectrum, δ, ppm							SSCC, ^3J, Hz		φ°*	Yield, %
			R	C≡N	$2\text{-}H_a$ (d)	$1\text{-}H_a$ (d)	$4\text{-}H_a$ (s)	9-He (d)	10-He (d)	NH_2 (s)**	Ar, R, 5-H, 6-H, 7-H, 8-H (m)**	$1\text{-}H_a$, $2\text{-}H_a$	9-He 10-He		
IV a	$C_{21}H_{16}N_4O$	165 ... 166	1636, 1651, 1692, 3240, 3420	2260	4,24	4,83	5,77	5,41	6,56	7,78	7,1 ... 7,6 (10H)	7,9	7,6	137	76
IV b	$C_{21}H_{15}FN_4O$	171 ... 173	1640, 1656, 1688, 3235, 3424	2258	4,45	4,79	5,75	5,41	6,55	7,77	7,1 ... 7,77 (9H)	7,8	7,7	136	88
IV c	$C_{20}H_{15}N_5O\cdot{}^1/_2C_2H_6O_2$	148 ... 149	1636, 1652, 1692, 3180, 3424	2260	4,50	4,86	5,78	5,43	6,54		7,3 ... 8,8 (10H)	7,9	7,6	137	96
IV d	$C_{27}H_{19}N_3O$	122 ... 123	1690	2245	4,64	6,07	5,58	5,48	6,88		7,1 ... 7,9 (14H)	6,8	7,7	130	98
IV e	$C_{26}H_{18}N_4O$	118 ... 120	1685	2247	4,68	6,08	5,56	5,47	6,87		7,3 ... 8,8 (13H)	6,5	7,5	129	87

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*Torsion angle $H_aC_{(1)}C_{(2)}H_a$.

**The signals are overlapped.

***Complex with ethylene glycol (2:1), δ: 4.37 ppm (CH_2, t, ^3J = 5.1 Hz).

TABLE 2. Bond Angles in the IVc Molecule

Angle	ω°	Angle	ω°	Angle	ω°
$C_{(1)}N_{(11)}C_{(4)}$	109,8(3)	$C_{(20)}C_{(3)}C_{(22)}$	109,3(4)	$N_{(11)}C_{(10)}C_{(9)}$	122,7(5)
$C_{(1)}N_{(11)}C_{(10)}$	117,8(4)	$N_{(11)}C_{(4)}C_{(3)}$	101,1(3)	$O_{(1)}C_{(12)}N_{(13)}$	123,8(4)
$C_{(4)}N_{(11)}C_{(10)}$	118,1(4)	$N_{(11)}C_{(4)}C_{(4a)}$	114,9(4)	$O_{(1)}C_{(12)}C_{(1)}$	118,5(4)
$C_{(15)}N_{(16)}C_{(17)}$	117,1(5)	$C_{(3)}C_{(4)}C_{(4a)}$	116,9(4)	$N_{(13)}C_{(12)}C_{(1)}$	117,7(4)
$N_{(11)}C_{(1)}C_{(2)}$	106,1(3)	$C_{(4)}C_{(4a)}C_{(5)}$	120,0(4)	$C_{(2)}C_{(14)}C_{(15)}$	118,1(4)
$N_{(11)}C_{(1)}C_{(12)}$	114,0(4)	$C_{(4)}C_{(4a)}C_{(5)}$	120,3(4)	$C_{(2)}C_{(14)}C_{(19)}$	125,0(4)
$C_{(2)}C_{(1)}C_{(12)}$	110,2(4)	$C_{(5)}C_{(4a)}C_{(8a)}$	119,7(4)	$C_{(15)}C_{(14)}C_{(19)}$	116,8(4)
$C_{(1)}C_{(2)}C_{(3)}$	101,9(3)	$C_{(4a)}C_{(5)}C_{(6)}$	121,3(5)	$N_{(16)}C_{(15)}C_{(14)}$	124,1(5)
$C_{(1)}C_{(2)}C_{(14)}$	116,6(4)	$C_{(5)}C_{(6)}C_{(7)}$	119,1(5)	$N_{(16)}C_{(17)}C_{(18)}$	123,4(6)
$C_{(3)}C_{(2)}C_{(14)}$	115,6(3)	$C_{(6)}C_{(7)}C_{(8)}$	119,9(6)	$C_{(17)}C_{(18)}C_{(19)}$	118,2(6)
$C_{(2)}C_{(3)}C_{(4)}$	100,5(3)	$C_{(7)}C_{(8)}C_{(8a)}$	122,5(5)	$C_{(14)}C_{(19)}C_{(18)}$	120,3(6)
$C_{(2)}C_{(3)}C_{(20)}$	112,9(4)	$C_{(4a)}C_{(8a)}C_{(8)}$	117,4(5)	$N_{(21)}C_{(20)}C_{(3)}$	175,6(5)
$C_{(2)}C_{(3)}C_{(22)}$	111,4(3)	$C_{(4a)}C_{(8a)}C_{(9)}$	118,1(4)	$N_{(23)}C_{(22)}C_{(3)}$	177,5(5)
$C_{(4)}C_{(3)}C_{(20)}$	111,4(3)	$C_{(8)}C_{(8a)}C_{(9)}$	124,4(5)	$O_{(2)}C_{(24)}C_{(24')}$	97,2(8)
$C_{(4)}C_{(3)}C_{(22)}$	111,2(3)	$C_{(8a)}C_{(9)}C_{(10)}$	121,7(5)		

TABLE 3. Coordinates of the Nonhydrogen Atoms ($\cdot 10^4$)

Atom	x	y	z	Atom	x	y	z
$O_{(1)}$	1668(1)	1763(3)	3937(2)	$C_{(7)}$	2016(3)	4310(6)	346(3)
$O_{(2)}$*	−32(5)	−5536(9)	1718(7)	$C_{(8)}$	1667(2)	5013(5)	492(3)
$N_{(11)}$	1618(1)	3308(3)	2310(2)	$C_{(8a)}$	1598(2)	4428(4)	1032(2)
$N_{(13)}$	2418(2)	3227(4)	4128(2)	$C_{(9)}$	1249(2)	5145(4)	1223(3)
$N_{(16)}$	234(2)	−2394(4)	1828(3)	$C_{(10)}$	1250(2)	4589(4)	1806(3)
$N_{(21)}$	1683(2)	−1193(4)	1273(2)	$C_{(12)}$	1844(2)	2456(4)	3627(2)
$N_{(23)}$	49(2)	1976(4)	2(2)	$C_{(14)}$	660(2)	−7(4)	1947(2)
$C_{(1)}$	1362(2)	2447(4)	2604(2)	$C_{(15)}$	741(2)	−1505(4)	2109(3)
$C_{(2)}$	1269(2)	845(3)	2262(2)	$C_{(17)}$	−383(2)	−1789(6)	1378(3)
$C_{(3)}$	1271(2)	1087(3)	1530(2)	$C_{(18)}$	−517(2)	−325(5)	1188(3)
$C_{(4)}$	1833(2)	2339(4)	2000(2)	$C_{(19)}$	16(2)	567(5)	1477(3)
$C_{(4a)}$	1906(2)	3057(4)	1435(2)	$C_{(20)}$	1483(2)	−228(4)	1373(2)
$C_{(5)}$	2271(2)	2365(5)	1298(3)	$C_{(22)}$	585(2)	1580(4)	658(2)
$C_{(6)}$	2332(2)	2985(6)	761(3)	$C_{(24)}$*	−319(3)	−5693(7)	2020(4)

*Atoms of the ethylene glycol molecule.

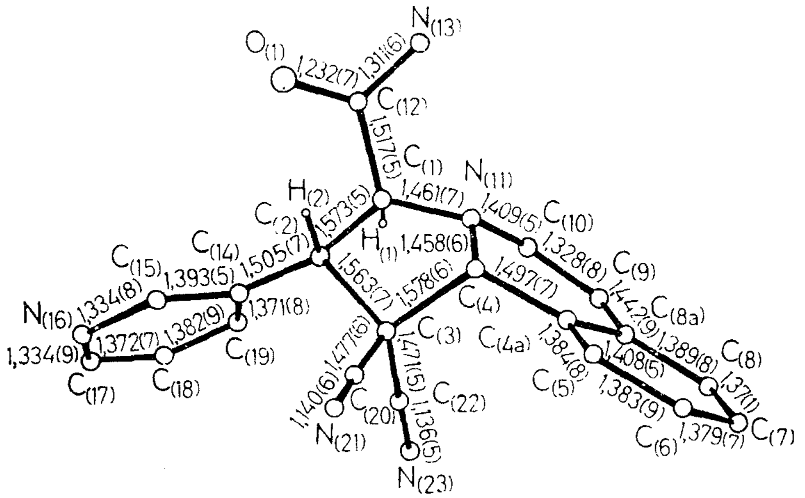
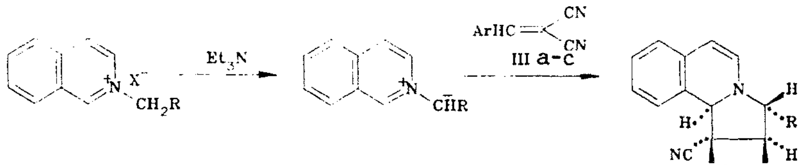


Fig. 1. Overall form of the IVc molecule and bond lengths in it. The trans orientation of the $H_{(1)}$ and $H_{(2)}$ atoms is apparent.

The isoquinolinium ylids were not isolated but were generated in the reaction mixtures by the usual method by treatment of isoquinolinium salts Ia, b in ethanol with an equimolar amount of triethylamine.



Ia,b IIa,b IVa-e

I, II a $R=CONH_2$, $X=Cl$; b $R=COC_6H_5$, $X=Br$; III a $Ar=C_6H_5$, b $Ar=4\text{-}FC_6H_4$, c $Ar=3\text{-}C_5H_4N$; IV a $R=CONH_2$, $Ar=C_6H_5$; b $R=CONH_2$, $Ar=4\text{-}FC_6H_4$; c $R=CONH_2$, $Ar=3\text{-}C_5H_4N$; d $R=COC_6H_5$, $Ar=C_6H_5$; e $R=COC_6H_5$, $Ar=3\text{-}C_5H_4N$

The subsequent reaction of the resulting isoquinolinium ylids IIa, b with arylmethylenemalononitriles IIIa-c in ethanol at 25-50°C proceeds highly regio- and stereoselectively to give substituted 1,2-trans-2-aryl-3,3-dicyano-1,2,3,4-tetrahydrobenzo[f]indolizines IVa-e (Table 1). The results obtained with respect to the regio- and stereoselectivity of the investigated reactions are in agreement with the results of IR and PMR spectroscopic studies and are unequivocally confirmed by x-ray diffraction analysis of the complex of IVc with ethylene glycol.

A number of absorption bands of deformation vibrations of amido and carbonyl groups are present in the IR spectra of IVa-c at 1636-1640, 1652-1656, and 1688-1692 cm^{-1} (see Table 1). The stretching vibrations of the NH_2 group show up in the form of two broad absorption bands at 3180-3235 and 3420-3424 cm^{-1} [9].

A characteristic peculiarity of the IR spectra of IVa-e is the presence of low-intensity absorption bands of an unconjugated nitrilo group at 2245-2260 cm^{-1}. Consequently, the molecules of the investigated indolizines IVa-e contain a hydrogenated pyrrole ring, and processes involving the elimination of hydrogen and hydrogen cyanide, which often accompany 1,3-dipolar cycloaddition reactions [1], do not occur in this case. Evidence for this is also provided by the PMR spectral data. The signals of the $C_{(1)}H$ and $C_{(2)}H$ protons of IVa-e show up in the form of two doublets at 4.79-6.08 and 4.42-4.68 ppm with SSCC ^3J = 6.5-7.9 Hz. The assignment of the indicated signals was made on the basis of a comparison of the PMR spectra of IVa-c and IVd, e; the chemical shift of the signal of the $C_{(2)}H$ proton in these compounds remains virtually unchanged. On passing to IVd, e the chemical shift of the signal of the $C_{(1)}H$ proton is shifted to weak field by a value on the order of $\Delta\delta$ = 41.2 ppm (see Table 1). In addition to this, the ^3J SSCC decreases from 7.8-7.9 Hz to 6.5-6.8 Hz, which should lead to a decrease in the trans-torsion angle $\varphi_{HC_{(1)}C_{(2)}H}$, as well as to more pronounced shielding of the $C_{(2)}H$ atom and a shift of its signal to strong field. However, the observed dependence constitutes evidence that the principal contribution to the weak-field shift of the signal of the $C_{(2)}H$ atom is made not by a steric factor but rather by an electronic factor due to polarization of the $C_{(2)}H$ atom by the stronger electron-acceptor COC_6H_5 substituent as compared with $CONH_2$. In addition, the PMR spectra of IVa-e contain, in addition to signals of protons of an aromatic substituent and an amino group, doublets of signals of $C_{(9)}H$ and $C_{(10)}H$ protons at 5.41-5.48 and 6.54-6.88 ppm with ^3J SSCC 7.5-7.7 Hz. The signal of the $C_{(4)}H$ proton shows up in the form of a singlet at 5.56-5.77 ppm; the $C_{(4)}H$ hydrogen atom is strictly oriented in space and is in a pseudoaxial orientation relative to the planar isonicotine fragment. The pseudoaxial orientation of the $C_{(4)}H$ atom under consideration leads to partial steric shielding of it by the benzoyl group and, on passing from IVa-c to IVd, e, the signal of the $C_{(4)}H$ proton in the PMR spectra is shifted by 0.2-0.22 ppm to strong field. These data constitute evidence that the substituent attached to the $C_{(1)}$ atom and the $C_{(4)}H$ hydrogen atom are located in the same plane and undergo certain steric interactions.

Considering the $^3J_{1\text{-}H,2\text{-}H}$ SSCC one may note that the reaction to form IVa-e proceeds highly stereoselectively. However, in the case of such SSCC the torsion angles calculated from the Karplus—Conroy equation [10] have two values, viz., $\varphi^1_{H_aC(1)C(2)H_a}$ 129-137° and $\varphi^2_{H_aC(1)C(2)H_e}$ 35-44°. Consequently, in the reaction of II and III one might expect the formation of 1,2-trans- or 1,2-cis-1,2,3,4-tetrahydrobenzo[f]indolizines. To unequivocally establish the regio- and stereoselectivity of the reactions of isoquinolinium ylids II with arylmethylenemalononitriles III we carried out an x-ray analysis of IVc.

The overall form of the IVc molecule and the bond lengths in it are shown in Fig. 1, while the bond angles are presented in Table 2. In the IVc molecule the torsion angles $C_{(12)}C_{(1)}C_{(2)}C_{(14)}$ 92.3(4)° and $H_{(1)}C_{(1)}C_{(2)}H_{(2)}$ 152(5)° indicate a cis-pseudoequatorial orientation of the carbamoyl and pyridine substituents and a trans-pseudoaxial orientation of the $H_{(1)}$ and $H_{(2)}$ atoms.

The five-membered heteroring of the IVc molecule has a twist conformation, as evidenced by the values of the modified [11] Cramer—Pople parameters [12] (φ = 249.4° and q_2 = 0.614 Å). The substituted dihydropyridine ring exists in a half-chair conformation: the $N_{(11)}$ and $C_{(4)}$ atoms deviate from the plane of the remaining four $C_{(4a)}C_{(8a)}C_{(9)}C_{(10)}$ ring atoms [plane I, with an accuracy of 0.022(5) Å] by −0.144(3) Å and 0.144(4) Å, respectively. Fragment I enters into the composition of a planar system that also includes the $C_{(4a)}\ldots C_{(8a)}$ benzene ring (the dihedral angle between them is 4.1°), which is not coplanar with respect to the five-membered ring. The twisted character of the IVc molecule along the $C_{(4)}$—$N_{(11)}$ bond is characterized by torsion angles $C_{(3)}C_{(4)}N_{(11)}C_{(1)}$ 36.1(3)°, $C_{(10)}N_{(11)}C_{(4)}C_{(4a)}$ 24.0(3)°, $C_{(4a)}C_{(4)}N_{(11)}C_{(1)}$ 162.9(4)°, and $C_{(10)}N_{(11)}C_{(4)}C_{(3)}$ 102.6(4)°.

The IVc molecule contains a number of shortened nonvalence contacts, which, in particular, determine rotation of the carbamoyl group, i.e., the coordination plane of the $C_{(12)}$ atom [the $C_{(12)}$ atom deviates from the plane of the $C_{(1)}O_{(1)}N_{(13)}$ atom by only 0.008(5) Å] relative to the plane of the $N_{(11)}C_{(1)}C_{(2)}$ fragment of the five-membered heteroring by 58.1°, as well as rotation of the pyridine ring relative to the $C_{(1)}C_{(2)}C_{(3)}$ plane by 75.4°. At the same time, the dihedral angle between the plane drawn through the $N_{(21)}C_{(20)}C_{(3)}C_{(22)}N_{(23)}$ atoms with an accuracy within the limits 0.031(5) Å and the plane of the $C_{(2)}C_{(3)}C_{(4)}$ fragment is 88.5°, i.e., it is close to the ideal value of 90°C.

The $C_{(1)}$—$N_{(11)}$ [1.461(7) Å] and $C_{(4)}$—$N_{(11)}$ [1.458(6) Å] bond lengths are only slightly shorter than the standard value of the $C_{(sp^3)}$—$N_{(sp^3)}$ bond length of 1.475 Å in tetrahydropyrrole [13] and are close to the corresponding values in the unsubstituted tetrahydropyrrole substituent of 2-(1-pyrrolidinyl)benzylidenemalononitrile (V) (1.475 and 1.468 Å) [14] and 1,5-di(1-pyrrolidinyl)-4-hexene-1,3-dione (VI) [1.472(3), 1.472(3) and 1.473(3), 1.452(3) Å [15]], as well as comparable with the values found in the sterically strained alkaloid gemantamine (VII) [1.484(4) and 1.472(4) Å] [16]. At the same time, the $C_{(1)}$—$C_{(2)}$ [1.573(5) Å], $C_{(2)}$—$C_{(3)}$ [1.563(7) Å], and $C_{(3)}$—$C_{(4)}$ [1.578(6) Å] bond lengths are increased as compared with the standard $C_{(sp^3)}$—$C_{(sp^3)}$ bond length, as, for example, in cyclopentane (1.543 Å [13]), and substantially surpass the values of the corresponding bond lengths in V (1.502, 1.502, 1.511 Å) and VI [1.478(5), 1.405(6), 1.506(6) Å and 1.508, 1.498(5), 1.527(5) Å], remaining comparable with the values established in VII [1.540(3), 1.571(4), 1.562(4) Å]. The increase in the indicated bond lengths in the heteroring of the IVc is probably due to its strained character and steric effects that arise when the ring is formed, as well as with the presence of electron-acceptor CN groups attached to the $C_{(3)}$ atom.

Conjugation leading to an appreciable increase in the length of the C(12)═$O_{(1)}$ bond to 1.232(7) Å [which actually coincides with the standard value of the length of the conjugated $C_{(sp^2)}$═O bond in amides (1.234 Å) [13]] and to substantial shortening of the length of the $C_{(sp^2)}$—N bond of 1.325 Å in amides [13], is observed in the carbamoyl group of the IVc molecule.

EXPERIMENTAL

The IR spectra of KBr pellets of the compounds were recorded with a Perkin—Elmer 577 spectrometer. The PMR spectra of solutions of the compounds in d_6-DMSO were recorded with a Bruker WM-250 spectrometer with tetramethylsilane (TMS) as the internal standard.

X-ray Diffraction Study of 1-Carbamoyl-3,3-dicyano-2-(3-pyridyl)benzo[f]indolizine (IVc). Compound IVc was investigated in the form of the crystal solvate with ethylene glycol (1:0.5 $C_2H_6O_2$). The crystals were monoclinic and had the following parameters at 20°C: $a = 27.5645(35)$, $b = 9.1603(50)$, $c = 21.102(3)$ Å, $\beta = 135.953(8)°$, $V = 3704(2)$ Å^3, $d_{calc} = 1.342$ g/cm^3, $Z = 8$, space group C2/c. The cell parameters and the intensities of 2151 independent reflections were measured with a Hilger—Watts four-circle automatic diffractometer (λ_{MoK_α}, graphite monochromator, $\theta/2\theta$ scanning up to $\theta_{max} = 30°$). The structure was decoded by the direct method by means of the MULTAN program and was refined within the totally matrix method of least squares within the anisotropic approximation for the nonhydrogen atoms with respect to 1833 reflections with $I \geq 3\sigma$. The hydrogen atoms, objectively revealed by differential synthesis, were refined isotropically with fixed $B_{iso} = 6.0$ Å^2. The final divergence factors R = 0.058 and R = 0.067. All of the calculations were carried out with an Eclipse S/200 computer by means of the INEXTL programs [18]. The coordinates of the nonhydrogen atoms are presented in Table 3.

Substituted 2-Aryl-3,3-dicyano-1,2-trans-1,2,3,4-tetrahydrobenzo[f]indolizines IVa-e. A 10-ml sample of triethylamine was added with stirring at 25-50°C to a suspension of 10 mmole of Ia, b and 10 mmole of arylmethylenemalononitrile IIIa-c in 25-40 ml of ethanol, after which the reaction mixture was stirred at 25°C for 8 h, and the precipitate was removed by filtration and washed with ethanol and hexane. Compounds IVa-e were recrystallized from ethanol.

The complex of IVc with ethylene glycol (1:0.5) was obtained by recrystallization of IVc from an ethanol—ethylene glycol system (1:1).

The results of elementary analysis for C, H, and N for IVa-e were in agreement with the calculated values.

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