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Synthesis of a single isomer of the bis-adduct of isocyanurato-substituted azide with [60]fullerene

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A single di(azahomo)[60]fullerene isomer was prepared for the first time by the reaction between [60]fullerene and isocyanurato-substituted azide. The structure of the product was established by ^{1}H and ^{13}C NMR, UV, and IR spectroscopy.

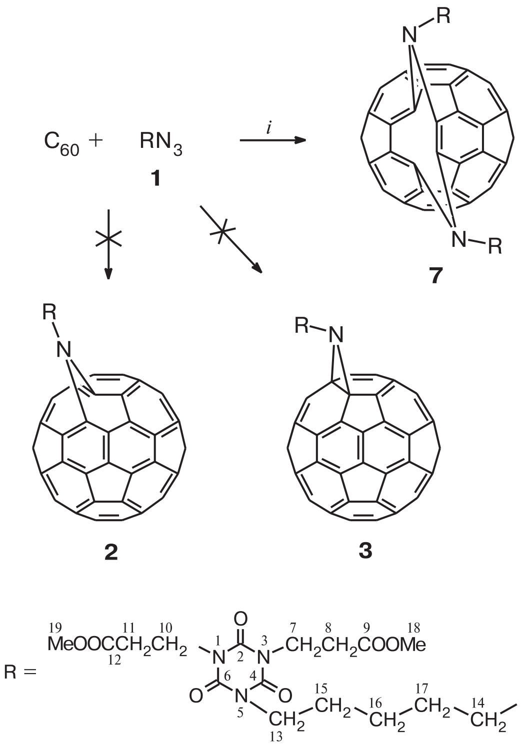
Key words: [60]fullerene, azidoalkyl isocyanurate, bis-adduct, di(azahomo)[60]fullerene.

Previously,[1,2] we demonstrated that reactions of C_{60} fullerene with 1,3-diallyl- and 1,3-di(cyanoethyl)isocyanuratoalkyl azides result in azahomo[60]fullerene, [60]fullereno[1,2-*b*]triazoline, and [60]fullereno[1,2-*b*]- and [60]fullereno[1,6-*b*]aziridine monoadducts. To continue studies of reactions of C_{60} with isocyanurato-substituted azides, we used 1,3-di(methoxycarbonylethyl)-substituted isocyanuratoalkyl azide **1**. We expected that azahomo[60]fullerene **2** will be obtained at 100 °C and [60]fullereno[1,2-*b*]aziridine **3** will be formed at 180 °C, because recently, we demonstrated[2] that kinetically controlled reactions give azahomofullerenes, while thermodynamic control results in [60]fullereno[1,2-*b*]aziridines. However, an unexpected result was obtained.

Azide **1** was synthesized by refluxing 5-(5-bromopentyl)-1,3-di(methoxycarbonylethyl) isocyanurate **4** with NaN_3 in anhydrous acetone for 6 h. Compound **4** was prepared by the reaction of excess 1,5-dibromopentane with the monosodium salt of 1,3-di(methoxycarbonylethyl)-substituted isocyanuric acid **5**, prepared by esterification of di(carboxyethyl)-substituted isocyanuric acid **6** with methanol.

The reaction of C_{60} with azide **1** was carried out by heating the reactants in *o*-dichlorobenzene for 4 h at 100 or 180 °C (Scheme 1). Column chromatography of the reaction mixture obtained at 100 °C furnished unreacted fullerene (~80% of the starting amount) and a fraction representing a mixture of fullerene adducts, whose attempted separation failed. An increase in the reaction temperature to 180 °C diminished the amount of recovered fullerene to 20%. Moreover, only one product **7** was isolated from the reaction mixture along with fullerene (the product yield was 18% relative to the fullerene used in the reaction). The elemental analysis of product **7** corresponded to the addition of two azide molecules to C_{60} with elimination of two dinitrogen molecules. A study of product **7** on a MALDI TOF laser desorption spectrometer did not give the molecular ion peak, probably, due to low volatility of the compound. Such a behavior is typical of [60]fullerene bis-adducts and, especially, di(azahomo)[60]fullerenes.[3]

Scheme 1



Reagents and conditions: *i*. *o*-Dichlorobenzene, 180 °C, 4 h.

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The structure of compound **7** was established by ^{13}C and ^{1}H NMR, IR, and UV spectroscopy.

The ^{13}C NMR spectrum of adduct **7** exhibits 32 signals (28 signals with 2 C intensity and 4 signals with 1 C intensity) in the region of δ 133—147, corresponding to sp^2-hybridized carbon atoms of the fullerene cage. This implies a homofullerene structure of the spheroid with C_s symmetry of the molecule. In addition, the spectrum contains signals for the C atoms of two magnetically equivalent methoxycarbonylethyl-substituted isocyanurate fragments. The equivalence of the methoxycarbonylethyl substituents is also indicated by the ^{1}H NMR spectral data for compound **7**. The IR spectrum of this compound exhibits characteristic absorption bands for the carbonyl groups of the isocyanuric ring (1690 cm^{-1}) and for the methoxycarbonylethyl substituent in this ring (1737 cm^{-1}) and also a band at ~526 cm^{-1}, typical of fullerene derivatives. However, unlike the spectra of isocyanurato-substituted fullerene monoadducts isolated previously,**[1,2]** in the IR spectrum of compound **7**, the intensity of the last-mentioned band is much lower than those of the absorption bands corresponding to the isocyanurate fragment. This trend in the variation of band intensities of outer-sphere and fullerene fragments in the IR spectra of mono- and bis-adducts has been observed previously**[4]** for phosphorylated isoxazoline derivatives of C_{60} fullerene. The UV spectrum of compound **7** exhibited no bands with λ_{max} = 420—430 nm characteristic of the addition of a dipolarophile to the bonds common to two six-membered rings (the fullerene [6,6]-bond). Thus, spectroscopic data (first of all, the narrow lines and the number of lines in the ^{13}C and ^{1}H NMR spectra) in combination with TLC and elemental analysis data indicate that product **7** is a single compound with a di(azahomo)[60]fullerene structure.

The homofullerene structure of C_{60} adducts implies that the fullerene cage has been extended upon bond cleavage between the rings and formation of bridges from the additional atoms. In the case of monoazahomo[60]fullerene adducts, only structures formed upon cleavage of bonds common to five- and six-membered rings**[5]** ([5,6]-bonds) to give [5,6]-open adducts have been substantiated theoretically and obtained experimentally, whereas in the case of di(azahomo)fullerenes, both traditional [5,6]-open bis-adducts**[6—9]** and isomeric [6,6]-open bis-adducts,**[10]** formed upon cleavage of two [6,6]-bonds, have been reported.

A di(azahomo)[60]fullerene molecule with C_s symmetry, which is consistent theoretically with the ^{13}C NMR spectrum of adduct **7**, can exist as ten regioisomers whose formation requires cleavage of two [5,6]-bonds in C_{60} (Fig. 1, structures **A—J**) and the appearance of two imine bridges in the fullerene cage. However, the pyramidal structures of the N atoms in these bridges implies, in addition, the presence of conformers in each regioisomer **A—J** with different mutual orientations of their exocyclic bonds. As a consequence, apart from C_s symmetry, regioisomers **A—G** can have C_1 symmetry, isomers **H** and **I**, C_{2v} symmetry and isomer **J**, C_{2h} symmetry. The possible inversion of the pyramidal nitrogen N would average the molecular geometry on the NMR time scale and would thus result in degeneration of two types of symmetry for each regioisomer to the higher one. Therefore, the ^{13}C NMR spectra of isomers **H—J** would, most likely, differ from the spectrum recorded for bis-adduct **7**, because C_{2v} symmetry implies the presence of 16 signals in this spectrum (14 signals with 4 C intensity and two signals with 2 C intensity) and C_{2h} symmetry accounts for 15 signals with 4 C intensity. Therefore, structures **A—G** are the most probable for bis-adduct **7**.

In order to decide between structures **A—G**, let us consider two more criteria. The first one concerns the positions of the signals for C atoms with 1 C intensity in the ^{13}C NMR spectrum and the second one, the experimental data on the reactivity of the precursors of the bis-adducts.

In regioisomers **A—G**, planes of symmetry pass through four nonequivalent C atoms. In each regioisomer, these C atoms occur in different fragments of the fullerene sphere, which should influence the positions of their signals in the ^{13}C NMR spectra. It was shown for an osmium fullerene derivative,**[11]** in whose ^{13}C NMR spectrum all signals were assigned to definite C atoms, that the higher the geometry distortion in the spheroid region, the greater the deviation of the chemical shifts of the C atoms from a value of δ 142, which corresponds to the starting C_{60}. In addition to the change in the geometric parameters of the adduct molecule, the fragment added to fullerene perturbs the fullerene electronic system and this should affect the position of signals for the C atoms located in the immediate proximity of the addition site.

The ^{13}C NMR spectrum of bis-adduct **7** has four signals with 1 C intensity. Three of them are characterized by similar chemical shifts (δ 142.22, 143.15, and 143.46), which do not differ substantially from the chemical shift of initial C_{60}, while the fourth 1 C signal is shifted upfield (δ 137.77). This set of signals corresponding to single C atoms is better consistent with regioisomers **A—C**, because in structures **D—G**, the single C atoms located in a symmetry plane are remote from the addition sites, and, most likely, the chemical shifts of the signals due to these atoms would be similar to one another and would not differ from the chemical shift of the initial C_{60}.

In regioisomer **A**, one of the single C atoms is incorporated in an annulene ring on the surface of the fullerene sphere; in the ^{13}C NMR spectra of bis-adducts with this structure, signals at about 160 ppm are attributed to this C atom.**[6]** This low-field position (normally, the signals

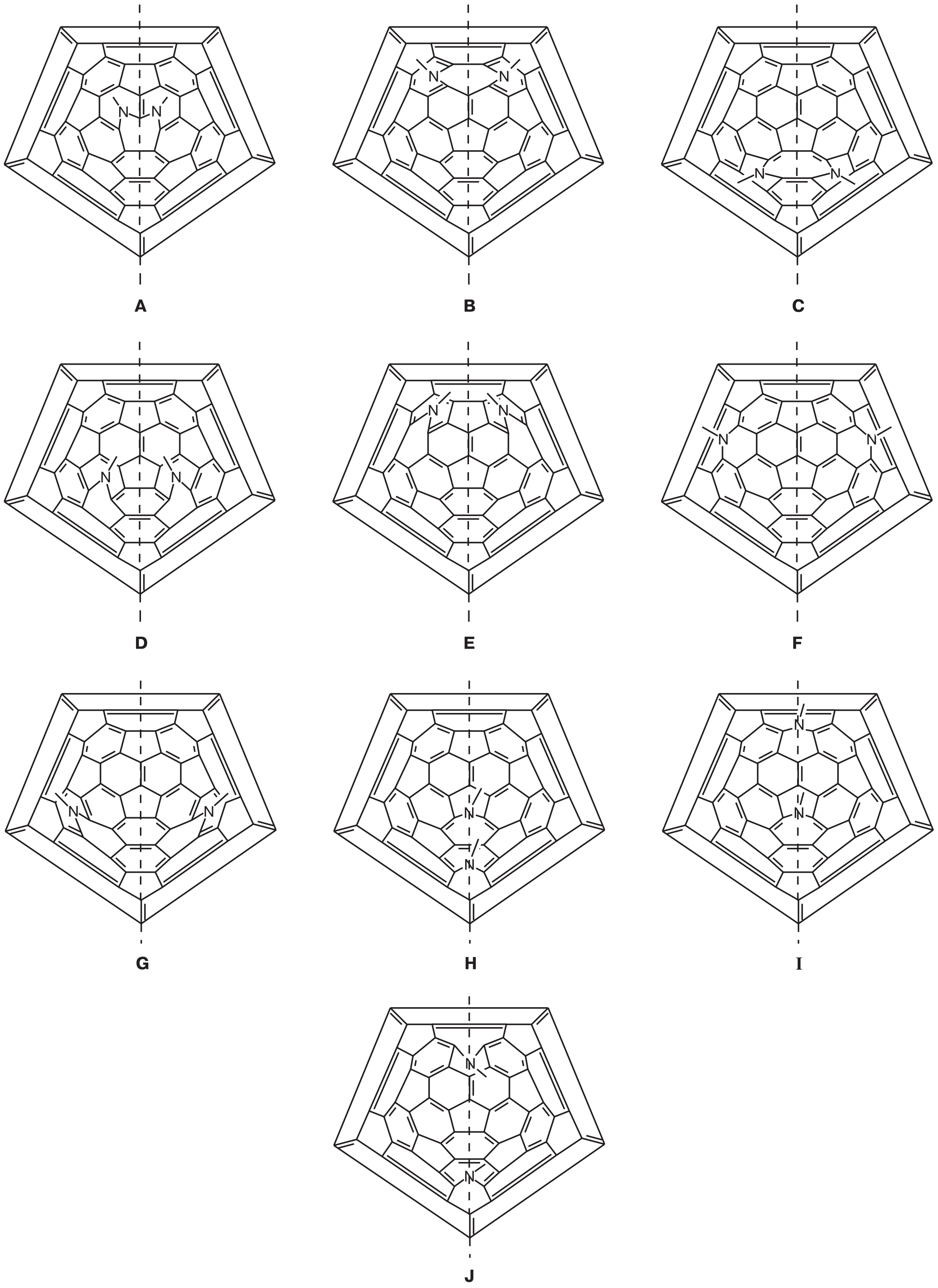


Fig. 1. Schlegel diagrams for the possible isomers of bis-adduct **7** with C_s symmetry.

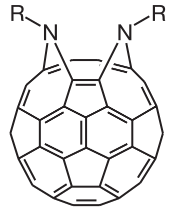
for sp^2-hybridized C atoms of the fullerene sphere are recorded at δ 133—147) is due not only to the distortion of the fullerene cage geometry but also to the presence of two electron-withdrawing N atoms in the neighborhood of the C atom under discussion. In regioisomer **B**, one of the single C atoms also belongs to the annulene ring, but ^{13}C NMR signals at δ 128—130 are usually attributed to this atom.[8,9] No signals with these chemical shifts are recorded in the ^{13}C NMR spectrum of product **7**; hence, structures **A** and **B** can be ruled out. Thus, the only regioisomer whose structure fully complies with the set of chemical shifts for single carbon atoms in the ^{13}C NMR spectrum of product **7** is regioisomer **C**.

The fact that regioisomer **C** is preferred over other structures can also be confirmed by considering the reactivity of the bis-adduct precursors. It is known[3] that the first step of reaction between C_{60} and alkyl and aryl azides gives [60]fullereno[1,2-*b*]triazolines, which undergo thermal decomposition to azahomo[60]fullerenes. On the basis of this pathway to the adducts, a pattern of formation of type **C** regioisomers in the reactions of C_{60} with diazides has been proposed.[12] According to this pattern, the second azide molecule adds to [60]fullereno[1,2-*b*]triazoline. In our opinion, this pathway to the isomers is unlikely due to the low thermal stability of [60]fullereno[1,2-*b*]triazolines. In view of the high temperature of the reaction between C_{60} and azide **1**, we are more disposed to the interpretation based on the addition of the second azide molecule to azahomo[60]fullerene.

The reactivity of homo- and azahomofullerenes has been little studied. Hydrogenation of homofullerenes,[13] oxidation of azahomofullerenes,[14] and the addition of a dipolarophile to azahomofullerenes[15] have been described. The structures attributed to the bis-adducts formed in these reactions imply cleavage of bonds located in the close vicinity of the site of attachment of the first addend molecule. It is believed that small reagents (for example, H_2 or O_2) add to the C(1)=C(2) bond, while the bulky substituents add to the C(3)=C(4) bond. However, none of the publications cited discusses other types of structures consistent with the set of spectroscopic parameters found for the isolated bis-adducts. In the case of reactions of C_{60} with organic azides,[6] the probability of formation of type **A** regioisomers was substantiated by quantum-chemical calculations, according to which the most positive charge in the fullerene cage of monoazahomo[60]fullerene falls to the C atoms at the imine bridge (C(1) and C(6) atoms), which dictates the site of the attack by the second azide molecule and the formation of type **A** regioisomer. However, this consideration does not account for the formation of other types of regioisomers.

Proceeding from the highest reactivity of the bonds in the annulene fragment of azahomo[60]fullerene and taking into account the bulk of the isocyanuric ring, one can suggest that the second molecule of azide **1** would add to the C(3)=C(4) bond of azahomo[60]fullerene **2** and give triazolinoazahomofullerene **8**, which undergoes thermal decomposition to give bis-adduct **9**, which is isomerized into product **7** with a type **C** structure (Scheme 2).

As noted above, homofullerene bis-adducts may also be formed by opening of two [6,6]-bonds in C_{60} ([6,6]-open bis-adduct). Only one example of isolation of this type of compound has been reported;[10] structure **K** was attributed to this adduct, which does not contradict the above symmetry analysis and the assumption concerning the highest reactivity of the bonds in the annulene ring in the bis-adduct precursor.

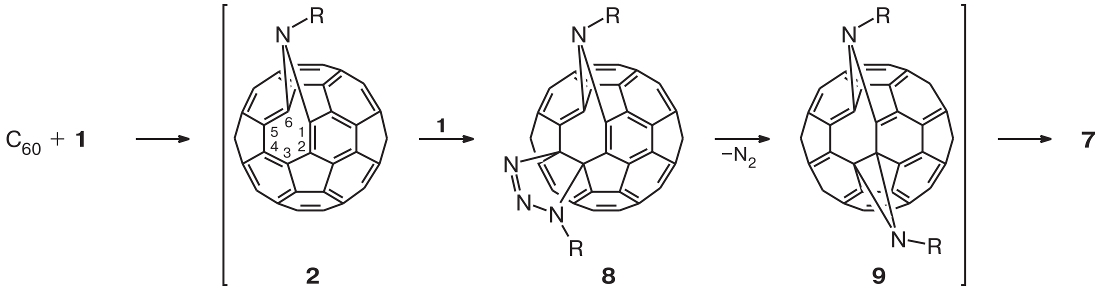


K

In the ^{13}C NMR spectrum of the described bis-adduct,[10] the signals of the C atoms linked directly to the N atoms of the imine bridges are located at δ 114 and 128. The spectrum of compound **7** does not exhibit signals with this chemical shift; hence, it cannot have the structure of type **K** regioisomer.

Thus, the reaction of C_{60} with di(methoxycarbonylethyl)isocyanuratoalkyl azide **1** at 180 °C, unlike the reactions with diallyl- and di(cyanoethyl)-substituted isocyanuratoalkyl azides,[1,2] gives rise to an individual bis-adduct isomer formed with opening of two [5,6]-bonds in one six-membered ring (type **C** structure).

Scheme 2



Experimental

Infrared spectra were recorded on a Bruker IFS-113V FT spectrometer in pellets with KBr. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker MSL-400 instrument in $CDCl_3$ (400.13 and 100.62 MHz, respectively). The δ values were calculated relative to $CDCl_3$. UV spectra were recorded on a Specord UV-VIS instrument. *o*-Dichlorobenzene was dehydrated by distillation over P_2O_5, and MeOH was distilled over MeONa. [60]Fullerene was prepared at the G. A. Razuvaev Institute of Organometallic Chemistry of the Russian Academy of Sciences (Nizhny Novgorod). 1,3-Di(2´-carboxyethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (**6**) was synthesized by a previously reported procedure.^{16}

1,3-Di(2-methoxycarbonylethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (5). 1,3-Di(2´-carboxyethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (**6**) (11 g, 0.04 mol) was heated in 50 mL of anhydrous MeOH in the presence of 9.1 mL of concentrated HCl for 2 h. Volatile compounds were evaporated *in vacuo* and the residue was recrystallized from benzene to give 11.5 g (95%) of compound **5**, m.p. 117 °C. Found (%): C, 44.61; H, 5.16; N, 14.10. $C_{11}H_{15}N_3O_7$. Calculated (%): C, 43.85; H, 5.02; N, 13.95. IR, ν/cm $^{-1}$: 1710 (C=O), 1740 (COOMe), 3070 (NH). ^{1}H NMR, δ: 6.86 (br.s, 1 H, NH); 3.83 (m, 4 H, C(7)H_2, C(10)H_2, $^3J_{H,H}$ = 7.0 Hz*); 3.31 (s, 6 H, C(18)H_3, C(19)H_3); 2.51 (m, 4 H, C(8)H_2, C(11)H_2, $^3J_{H,H}$ = 7.0 Hz).

5-(5-Bromopentyl)-1,3-di(2-methoxycarbonylethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (4). Compound **5** (5 g, 0.016 mol) was added to a solution of NaOH (0.66 g, 0.016 mol) in 60 mL of water. The mixture was stirred for 1 h at 60—70 °C. Water was removed *in vacuo*, the residue was diluted with toluene, which was distilled off, first, under atmospheric pressure and at the end, *in vacuo*. A solution of 1,5-dibromopentane (15 g, 0.065 mol) in 100 mL of DMF was added to the resulting dry monosodium salt of 1,3-di(2-methoxycarbonylethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione. The mixture was stirred for 3 h at 50—60 °C until it was neutral. The solvent was removed *in vacuo*, 50 mL of benzene was added to the residue, the precipitate was filtered off, and the residue was dried *in vacuo* and chromatographed on a column with silica gel (elution with a petroleum ether—$CHCl_3$ mixture, 1 : 1) to give 4.8 g (64%) of compound **4** as an oil, R_f 0.74 (Silufol, elution with $CHCl_3$). Found (%): C, 42.64; H, 5.29; Br, 17.70; N, 8.81. $C_{16}H_{24}BrN_3O_7$. Calculated (%): C, 42.68; H, 5.37; Br, 17.75; N, 9.33. IR, ν/cm^{-1}: 1690 (C=O), 1734 (COOMe), 510 (CBr). ^{1}H NMR, δ: 4.16 (m, 4 H, C(7)H_2, C(10)H_2, $^3J_{H,H}$ = 7.0 Hz); 3.86 (m, 2 H, C(13)H_2, $^3J_{H,H}$ = 7.0 Hz); 3.66 (s, 6 H, C(18)H_3, C(19)H_3); 3.41 (m, 2 H, C(14)H_2, $^3J_{H,H}$ = 7.0 Hz); 2.65 (m, 4 H, C(8)H_2, C(11)H_2, $^3J_{H,H}$ = 7.0 Hz); 2.00—1.60 (m, 6 H, C(15)H_2, C(16)H_2, C(17)H_2).

5-(5-Azidopentyl)-1,3-di(2-methoxycarbonylethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (1). Compound **4** (1.6 g, 0.0035 mol) was added to a suspension of NaN_3 (0.5 g, 0.077 mol) in 20 mL of anhydrous acetone. The mixture was stirred for 4 h, NaBr was filtered off, and the filtrate was concentrated *in vacuo* and chromatographed on a column with silica gel (elution with a hexane—$CHCl_3$ mixture, 1 : 1) to give 0.9 g (61%) of compound **1**. Found (%): C, 46.64; H, 5.47; N, 19.89. $C_{16}H_{24}N_6O_7$. Calculated (%): C, 46.66; H, 5.82; N, 20.38. IR, ν/cm^{-1}: 1690 (C=O), 1710 (COOMe), 2105 (N_3). ^{1}H NMR, δ: 4.01 (m, 4 H, C(7)H_2, C(10)H_2, $^3J_{H,H}$ = 7.2 Hz); 3.74 (m, 2 H, C(13)H_2, $^3J_{H,H}$ = 7.3 Hz); 3.54 (s, 6 H, C(18)H_3, C(19)H_3); 3.29 (m, 2 H, C(14)H_2, $^3J_{H,H}$ = 6.7 Hz); 2.52 (m, 4 H, C(8)H_2, C(11)H_2, $^3J_{H,H}$ = 7.2 Hz); 1.77 (m, 2 H, C(17)H_2); 1.54 (m, 2 H, C(15)H_2); 1.35 (m, 2 H, C(16)H_2). ^{13}C NMR, δ: 170.57 (m, C(9), C(12), $^2J_{C,H}$ = 3.7 Hz); 148.21 (s, C(2), C(6)); 148.12 (s, C(4)); 51.38 (q, C(18), C(19), $^1J_{C,H}$ = 147.0); 42.24 (tm, C(13), $^1J_{C,H}$ = 142.2 Hz, $^2J_{C,H}$ = 4.3 Hz); 38.30 (tt, C(7), C(10), $^1J_{C,H}$ = 145.7 Hz, $^2J_{C,H}$ = 5.2 Hz); 32.93 (tm, C(14), $^1J_{C,H}$ = 144.0 Hz); 31.90 (tt, C(8), C(11), $^1J_{C,H}$ = 131.2 Hz, $^2J_{C,H}$ = 3.2 Hz); 31.68 (tt, C(17), $^1J_{C,H}$ = 130.0 Hz, $^2J_{C,H}$ = 4.7 Hz); 26.65 (tm, C(15), $^1J_{C,H}$ = 124.3 Hz); 24.65 (tm, C(16), $^1J_{C,H}$ = 123.0 Hz).

Reaction of C_{60} with 5-(5-azidopentyl)-1,3-di(2-methoxycarbonylethyl)-(1*H*,3*H*,5*H*)-1,3,5-triazine-2,4,6-trione (1). A mixture of C_{60} (0.104 g, 0.144 mmol) and azide **1** (0.18 g, 0.433 mmol) in 25 mL of anhydrous degassed *o*-dichlorobenzene was heated at reflux for 4 h. The solvent was evaporated under atmospheric pressure and the residue was chromatographed on a column with silica gel to give 0.020 g of C_{60} (elution with toluene) and 0.040 g (18%) of bis-adduct **7** (elution with a toluene—$CHCl_3$ mixture, 1 : 1), R_f 0.16 (Silufol, elution with $CHCl_3$). Found (%): C, 73.61; H, 2.96; N, 7.63. $C_{92}H_{48}N_8O_{14}$. Calculated (%): C, 74.19; H, 3.25; N, 7.52. ^{1}H NMR, δ: 4.19 (m, 4 H, C(7)H_2, C(10)H_2, $^3J_{H,H}$ = 7.3 Hz); 3.88 (m, 2 H, C(13)H_2, $^3J_{H,H}$ = 7.3 Hz); 3.68 (s, 6 H, C(18)H_3, C(19)H_3); 3.40 (m, 2 H, C(14)H_2, $^3J_{H,H}$ = 6.7 Hz); 2.66 (m, 4 H, C(8)H_2, C(11)H_2, $^3J_{H,H}$ = 7.2 Hz); 1.90 (m, 2 H, C(17)H_2); 1.68 (m, 2 H, C(15)H_2); 1.49 (m, 2 H, C(16)H_2). ^{13}C NMR, δ: 170.98 (C(9), C(12)); 148.54 (C(2), C(6)); 148.61 (C(4)); 51.81 (C(18), C(19)); 33.16 (C(14)); 42.69 (C(13)); 38.70 (C(7), C(10)); 32.02 (C(17)); 32.08 (C(8), C(11)); 26.72 (C(15)); 25.02 (C(16)); 133.67, 135.77, 136.16, 137.09, 137.28, 137.98, 138.43, 139.14, 140.63, 141.35, 142.58, 142.63, 142.71, 142.83, 143.03, 143.31, 143.57, 143.76, 144.03, 144.07, 144.20, 144.23, 144.37, 144.49, 144.64, 144.95, 146.88, 147.71 (C_{60}N, 2 C); 137.77, 142.22, 143.15, 143.46 (C_{60}N, 1 C).

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References

1. I. P. Romanova, G. G. Yusupova, S. G. Fattakhov, A. A. Nafikova, V. I. Kovalenko, V. V. Yanilkin, V. E. Kataev, N. M. Azancheev, V. S. Reznik, and O. G. Sinyashin, *Izv. Akad. Nauk, Ser. Khim.*, 2001, 426 [*Russ. Chem. Bull., Int. Ed.*, 2001, **50**, 445].
2. O. G. Sinyashin, I. P. Romanova, G. G. Yusupova, A. A. Nafikova, V. I. Kovalenko, N. M. Azancheev, S. G.

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* For protons of all methylene groups, $^3J_{H,H} = 0.5(^3J_{AX} + {}^3J_{AX'})$ of an AA´XX´-system spectrum.

Fattakhov, and V. S. Reznik, *Izv. Akad. Nauk, Ser. Khim.*, 2001, 2064 [*Russ. Chem. Bull., Int. Ed.*, 2001, **50**, 2162].
3. A. Hirsch and B. Nuber, *Acc. Chem. Res.*, 1999, **32**, 795.
4. O. G. Sinyashin, I. P. Romanova, F. R. Sagitova, V. A. Pavlov, V. I. Kovalenko, Y. V. Badeev, N. M. Azancheev, A. V. Ilyasov, A. V. Chernova, and I. I. Vandyukova, *Mendeleev Commun.*, 1998, 79.
5. M. Cases, M. Duran, J. Mestres, N. Martin, and M. Sola, *J. Org. Chem.*, 2001, **66**, 433.
6. T. Grosser, M. Prato, V. Lucchini, A. Hirsch, and F. Wudl, *Angew. Chem., Int. Ed. Engl.*, 1995, **34**, 1343.
7. G.-X. Dong, J.-S. Li, and T.-H. Chan, *J. Chem. Soc., Chem. Commun.*, 1995, 1725.
8. P. P. Kanakamma, S.-L. Huang, C.-G. Juo, G.-R. Her, and T.-Y. Luh, *Chem. Eur. J.*, 1998, **4**, 2037.
9. L.-L. Shiu, K.-M. Chien, T.-Y. Li, G.-R. Her, and T.-Y. Luh, *J. Chem. Soc., Chem.Commun.*, 1995, 1159.
10. G. Schick, A. Hirsch, H. Mauser, and T. Clark, *Chem. Eur. J.*, 1996, **2**, 935.
11. J. M. Hawkins, S. Loren, A. Meyer, and R. Nunlist, *J. Am. Chem. Soc.*, 1991, **113**, 7770.
12. C. K.-F. Shen, H.-H. Yu, C.-G. Juo, K.-M. Chien, G.-R. Her, and T.-Y. Luh, *Chem. Eur. J.*, 1997, **3**, 744.
13. F. Djojo, A. Herzog, I. Lamparth, F. Hampel, and A. Hirsch, *Chem. Eur. J.*, 1996, **2**, 1537.
14. J. C. Hummeln, M. Prato, and F. Wudl, *J. Am. Chem. Soc.*, 1995, **117**, 7003.
15. L. Ulmer, G. Torres-Cargia, H. Luftmann, and J. Mattay, *J. Inf. Recording*, 1998, **24**, 243.
16. T. C. Frazier, E. D. Little, and B. E. Lloyd, *J. Org. Chem.*, 1960, **25**, 1945.

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