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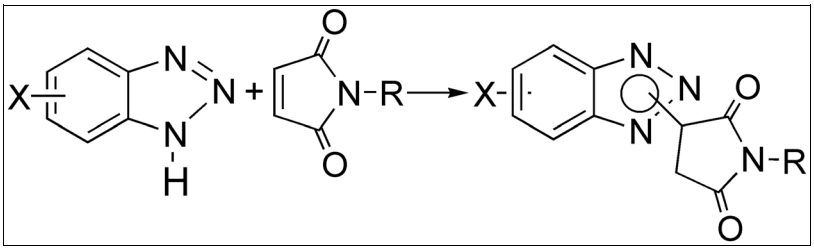

The one-step interaction of 1H-benzotriazoles with maleimides in melt at elevated temperatures has resulted in benzotriazolylsuccinimides. According to spectroscopy data, benzotriazolylsuccinimides obtained contain both major (benzotriazol-1-yl)-succinimide residue and minor (benzotriazol-2-yl)-succinimide residue. An effect of substituents in the initial reagents on the fine splitting of carbon peaks in ^{13}C NMR spectrums of benzotriazolylsuccinimides is discussed. The influence of structure of benzotriazolylsuccinimides on their crystallinity degree is also discussed.

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INTRODUCTION

1H-Benzotriazole is known to be one of interesting aromatic heterocyclic condensed compounds. In practice, 1H-benzotriazole is used as the antifogging agent in the photography and the corrosion inhibitor [1–3] as well as for promoting the urea nitrification process in soil [4]. For derivatives of 1H-benzotriazole, 2-aryl-substituted 1,2,3-benzotriazoles are utilized as the UV-stabilizers of polymers [5]. Polymers containing 2-aryl-substituted 1,2,3-benzotriazole units within a macromolecule are used in the elctrochromic devices [6]. 1H-Benzotriazole possesses high N-H acidity (pKa 8.2 in water [7]), and the excellent ability to form complex coordination compounds [8–10].

1H-benzotriazole and its derivatives take part in such reactions as N_2 abstracting followed by the cyclization [11]; the nucleophilic substitution [12–14] and the Michael nucleophilic addition to the electron-deficient π-bonds [7]. For information about the nucleophilic Michael addition of derivatives of 1H-benzotriazole to the maleinimide olefinic bond, there is an example of reaction of *N*-methylmaleimide with (benzotriazol-1-yl)methylene((benzotriazol-1-yl)methyl)amine under heating in toluene medium for 48 h and in the presence of trifluoroacetic acid. Such reaction has resulted in *N*-methyl-3-(1-benzotriazolyl)-pyrrolidin-2,5-dione formation [15]. Besides, *N*-methyl-3-(1-benzotriazolyl)-pyrrolidin-2,5-dione together with *N*-methyl-3-(2-benzotriazolyl)-pyrrolidin-2,5-dione were obtained by those authors as by-products when *N*-methylmaleimide was interacted with bis(benzotriazole-1-ylmethyl)hydroxylamine in toluene medium for 24 h and without trifluoroacetic acid. Flammable (toluene), irritant, and highly corrosive (trifluoroacetic acid) liquids were used in the both cases. Bisderivatives of 1H-benzotriazole, where one 1,2,3-benzotriazole unit was only consumed in reaction, were also utilized. In addition to this, a long period of time of reaction was required. We have offered the one-step synthesis of benzotriazolylsuccinimides in melt by means of the convenient reaction of the nucleophilic Michael addition of 1H-benzotriazoles to maleinimides. The absence of toxic, flammable, and corrosive solvents, catalysts as well as a lack of by-products evolution are the advantages of such reaction in terms of the green chemistry and the atom efficiency. To the best of our knowledge, information about such melt reaction between 1H-benzotriazoles and maleimides in the scientific literature is absent, excepting the works of the authors of the presented article and other authors [16] who referenced to our previous work [17]. This article is devoted to the continuation of our work concerning the melt synthesis and the investigation of benzotriazolylsuccinimides.

RESULTS AND DISCUSSION

Syntheses and NMR investigations of benzotriazolylsuccinimides. ***1-(2,5-Dioxo-1-phenylpyrrolidin-3-yl)-benzotriazole 1 and bis[3-(benzotriazolyl)succinimides] 2a-c.*** Previously [17], we carried

out the melt one-step condensation of 1H-benzotriazole with *N*-phenylmaleimide as well as bismaleimides with 2 mol of 1H-benzotriazole in a fluoroplast reactor according to reactions 1 and 2 (Scheme 1), respectively. Purified **2a–c**, in contrast to crystalline **1**, are white amorphous powders and cannot be crystallized. If compared the ^{13}C NMR spectrum data of **1** with the ones of **2b**, which are summarized in Table 1, the peaks of the minor (benzotriazol-2-yl)-succinimide residue appear in the ^{13}C NMR spectrum of **2b** as the ones of the smaller intensity at δ 36.27, 63.71, 118.20, 127.51, 144.21, 171.40, and 173.24 ppm. The similar peaks of the minor (benzotriazol-2-yl)-succinimide residues appear also in the ^{13}C NMR spectra of **2a** and **2c**. For **1a**, this compound was investigated by authors [16] who observed the ^{13}C NMR peaks of **1a** that are similar to the ^{13}C NMR ones of the minor (benzotriazol-2-yl)-succinimide residues of **2b-c**. The ratios of an integral intensity of the C(3′)H and C(3‴)H protons in the ^{1}H NMR spectrums of **2a–c** were ascertained to be 7.0 (for **2a**), 4.5 (for **2b**), and 7.0 (for **2c**), proving additionally the lesser content of the (benzotriazol-2-yl)-succinimide residues.

Thus, the NMR investigations of **1** and **2a–c**, along with a comparison with the NMR data of **1a** [16], have revealed that 1H-benzotriazoles interact with maleimides in melt at high temperatures to afford benzotriazolylsuccinimides comprising both major (benzotriazol-1-yl)-succinimide residue and minor (benzotriazol-2-yl)-succinimide residue.

Scheme 1. Synthesis of benzotriazolylsuccinimides in melt (**1** and **2a–c** were synthesized by us previously [17]).

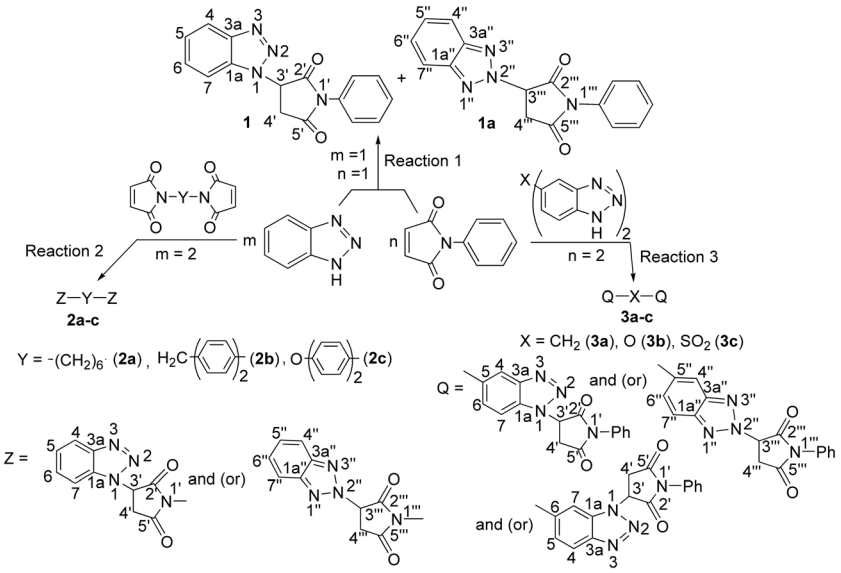


Table 1

Characteristics of benzotriazolylsuccinimides.

	^{13}C NMR chemical shifts of succinimide carbons (DMSO-d_6), ppm[a]									
Compound	C(2′)	C(2‴)	C(5′)	C(5‴)	C(3′)	C(3‴)	C(4′)	C(4‴)	Yield[b] (%)	CD[c] (%)
1	172.33	–	173.19	–	56.34	–	35.43	–	71	59[d]
2a	173.13	172.20	174.22	174.28	55.80	63.40	35.33	36.16	85	–
2b	172.31	171.40	173.19	173.24	56.28	63.71	35.40	36.27	88	5[e]
2c	171.59	170.66	172.42	172.55	55.72	63.20	35.13	36.01	91	–
3a	172.26	171.39	173.13	173.22	56.23	63.61	35.35	36.25	75	6[e]
	172.32	–	173.15	–	56.37	–	35.37	–	–	–
3b	Seven peaks at	171.25	Eight peaks at	173.21	55.94	63.56	35.28	36.20	72	5.5[e]
	172.06–172.15	–	172.93–173.08	–	56.01	–	35.31	–	–	–
3c	171.80	170.74	172.87	173.08	56.52	64.31	35.25	36.32	68	10[e]
	171.95	–	172.92	–	56.60	–	35.28	–	–	–

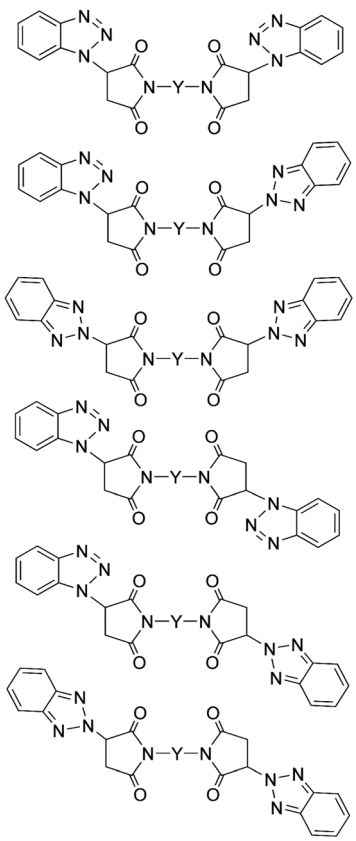
[a]Carbon indexes correspond to ones on Scheme 1.
[b]Purified compounds.
[c]CD is crystallinity degree that has been calculated as the ratio of an amorphous area to a total (amorphous + crystalline) one on the X-ray powder diffractograms.
[d]mp 145–147°C.
[e]Amorphous.

bis[(N-phenylsuccinimid-3-yl)benzotriazoles] 3a–c. Next, it was interesting to carry out the syntheses of benzotriazolylsuccinimides via the interaction of 5,5′-bisbenzotriazoles with 2 mol of N-phenylmaleimide. 5,5′-Bisbenzotriazoles contain, in contrast to 1H-benzotriazole, the two nonequivalent 1-nitrogen atoms because of bridge groups located between the benzene rings. In more detail, 5,5′-bisbenzotriazoles possess all the three nonequivalent nitrogen atoms in each 1H-benzotriazole residue that must contribute to specific NMR spectral patterns and other properties of reaction products. Hence, as the continuation of our work, we have realized the syntheses of methylene-**3a**, oxa-**3b**, and dioxothia-**3c** bis[(N-phenylsuccinimid-3-yl)-benzotriazoles] via the condensation of 5,5′-bisbenzotriazoles such as 5,5′-bisbenzotriazol methane, 5,5′-bisbenzotriazol oxide, and 5,5′-bisbenzotriazol sulfone with 2 mol of *N*-phenylmaleimide according to reaction 3 (Scheme 1). Purified **3a–c** are white amorphous powders and cannot be crystallized. The values of the ^{13}C NMR resonance peaks of the succinimide carbons of **3a-c** are summarized in Table 1. The intensive resonances of the carbonyl, methine, and methylene succinimide carbons of **3a** that correspond to the carbons of the major (benzotriazol-1-yl)-succinimide residue are split onto the two peaks. This fact may be explained in terms of nonequivalence of the 1-nitrogen atoms in the bisbenzotriazolyl residues of **3a** because of the bridge group of CH_2 located between the benzene cycles. The similar situation is observed for compound **3c** with the sulfone bridge group. Signals for the carbons of the (benzotriazol-2-yl)-succinimide residues in the ^{13}C NMR spectrums of **3a–c** are not split. This is due to the symmetrical character of the benzotriazol-2-yl residue. The resonance peaks for the carbonyl and methine carbons of the succinimide cycle of the (benzotriazol-1-yl)-succinimide residues of **3c** are split analogously to the similar carbons of **3a**. The peaks of the carbonyl carbons of the nonsymmetrical (benzotriazol-1-yl)-succinimide residues of **3b** are split on seven and eight peaks. That is, they are split to the greater extent than the analogous peaks of **3a** and **3c**. This phenomenon appears to be due to the different electronic influences of the oxygen (the electron donor), methylene (the very weak electron donor because of absense of a lone pair of electrons) and sulfone (the electron acceptor) bridge groups on the benzotriazolyl nitrogens. The π-donating properties of the lone pair of electrons of the bridge oxygen in **3b** appear to be influenced by a *metha*-position or *para*-position of the N=N electron-acceptor group in the benzotriazol-1-yl residues or two C=N electron-acceptor groups in the benzotriazol-2-yl residue. Hence, a mutual π-electronic influence of one benzotriazolylsuccinimide residue on another benzotriazolylsuccinimide residue, which is transmitted via the bridge oxygen, should take place. Such mutual influence may be the cause of the fine splitting of the ^{13}C NMR signals of the carbonyl succinimide carbons of the nonsymmetrical (benzotriazol-1-yl)-succinimide residues in **3b**.

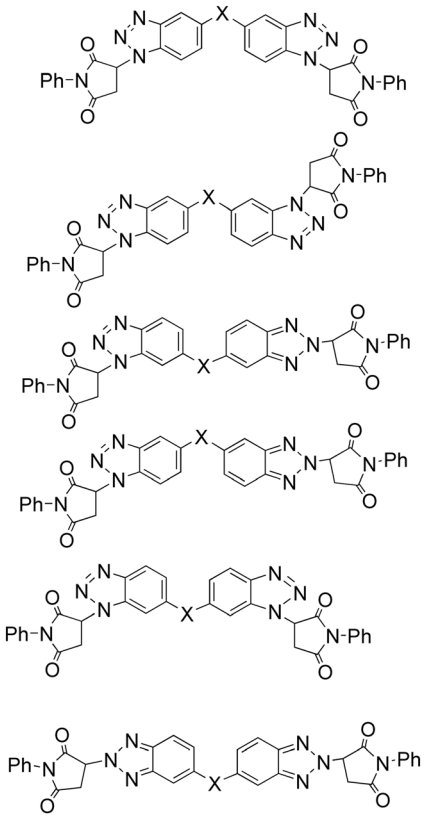
Crystallinity degree of bezotriazolylsuccinimides. A crystallinity degree of bezotriazolylsuccinimides is presented in Table 1. **1** possesses the higher value of this parameter of 59%, probably because of the single isomer presence. For other compounds **2b** and **3a–c**, a value of the crystallinity degree varies from 5 to 10 %, evidencing their amorphness. Amorphous nature of these compounds appears to be due to the presence of the six probable isomers of a benzotriazol-1-yl residue and (or) benzotriazol-2-yl residue position with respect to bissuccinimide (Scheme 2) bridge groups or a *N*-phenylsuccinimid-3-yl residue position with respect to a bridge group (−X−) located between the benzene rings of the bisbenzotriazolyl fragments (Scheme 3). In addition, every position isomer contains the two asymmetrical succinimide carbons, rendering possible the presence of the DD-stereoisomer, LL-stereoisomer, and DL-stereoisomer in one of position isomers. All this hinders the packing of molecules onto crystalline structure and favors the formation of amorphous powders.

Proposed reaction mechanism of benzotriazolylsuccinimides formation. The proposed reaction mechanism of the benzotriazolylsuccinimides formation is presented on the Scheme 4. Reaction begins with the nucleophilic attack of the lone pair of electrons of the sp^2-hybridized "pyridine"

Scheme 2. Probable isomers of the benzotriazol-1-yl residue and (or) benzotriazol-2-yl residue position with respect to the bissuccinimide bridge groups.



Scheme 3. Probable isomers of the *N*-phenylsuccinimid-3-yl residue position with respect to bridge groups located between the benzene rings of the bisbenzotriazolyl groups.

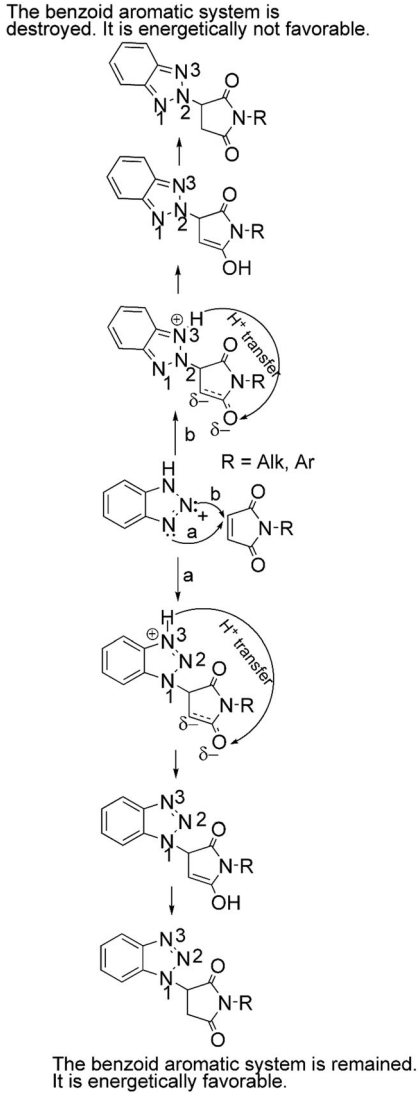


nitrogens of 1H-benzotriazole on the electron-deficient C=C bond of maleimides followed by the charged intermediates formation. These charged intermediates are transformed to the "enolic" neutral intermediates across a proton transfer. After, the enolic neutral intermediates are isomerized into benzotriazol-1-yl-succinimides or benzotriazol-2-yl-succinimides. The benzotriazol-2-yl residue contains the *ortho*-benzoquinoid fragment that is energetically less favorable than the benzoid fragment of the benzotriazol-1-yl residue. This fact explains the higher content of the nonsymmetrical benzotriazol-1-yl residues in reaction products.

CONCLUSIONS

1H-Benzotriazoles react with maleimides in melt via addition to the double bond of the latter to afford both major (benzotriazol-1-yl)succinimide and minor (benzotriazol-2-yl)succinimide. For the products of an interaction of 5,5′-bisbenzotriazoles with 2 mol of *N*-phenylmaleimide, they contain two kinds of the nonsymmetrical (benzotriazol-1-yl)-succinimide residues and single kind of the symmetrical (benzotriazol-2-yl)-succinimide residues. These results are evidence that all the three nitrogen atoms of benzotriazoles participate in the nucleophilic addition to maleimides.

Scheme 4. The proposed reaction mechanism of benzotriazolylsuccinimides formation.



The amorphous character of bisbenzotriazolylsuccinimides has been proved to be the consequence of the presence of the six probable isomers of a position of *N*-phenylsuccinimid-3-yl or benzotriazol-1-yl residue and (or) benzotriazol-2-yl residue with regard to the bridge groups in the bisbenzotriazolyl or bissiccinimidyl residues, respectively. The additional cause of the amorphous character of such compounds may be that every position isomer contains the two asymmetrical succinimide carbons, rendering possible the presence of the DD-stereoisomer, LL-stereoisomer, and DL-stereoisomer in one of position isomers.

EXPERIMENTAL

Fourier transform ir spectra were recorded with a "IFS25" instrument in KBr tablets. ^{13}C and ^{1}H NMR

spectra were obtained on a Varian VXR-500S spectrometer at 126.7 and 500 MHz, respectively, in DMSO-d_6 and $CDCl_3$; the signals of the residual protons for DMSO-d_6 ($\delta = 2.5$ ppm), $CDCl_3$ ($\delta = 7.24$ ppm) in ^{1}H NMR spectra and for the carbon atoms of DMSO ($\delta = 39.7$ ppm) in ^{13}C NMR spectra served as the internal standard. Melting points were determined on a "IA9100" instrument and not corrected. X-ray powder diffractograms were run on a "D8 ADVANCE" diffractometer of "Brucker ACX," Germany. The conditions of the diffractograms runs were CuKα-radiation and a "Vantec-1" detector.

Ice CH_3COOH, HCOOH, $NaHSO_3$, $NaNO_2$, and NaOH of the chemical pure grade were used as received. N-phenylmaleimide was prepared according to the method cited in the article [18]. All the solvents applied were purified according to the known procedures [19]. 1H-Benzotriazole was crystallized from benzene, mp 98–99°C. Bismaleimides were synthesized similar to the procedures [20].

3,3′,4,4′-tetraaminodiphenyl methane was purified by refluxing in distilled water in the presence of $NaHSO_3$ and charcoal. After a hot filtration and cooling, precipitated crystals were filtered off and rinsed with distilled water and dried *in vacuo* at 40–50°C, mp 140–142°C. 3,3′,4,4′-Tetraaminodiphenyl ether was purified as 3,3′,4,4′-tetraaminodiphenyl methane, mp 150–151°C. 3,3′,4,4′-Tetraaminodiphenyl sulfone was purified as 3,3′,4,4′-tetraaminodiphenyl methane and 3,3′,4,4′-tetraaminodiphenyl ether, mp 173–174°C.

5,5′-Bisbenzotriazol methane. The synthesis must be carried out in a fume chamber because of an evolution of nitrous oxides. A 0.5 L three-neck flask equipped with a stirrer, an inlet and an outlet of argon, and a thermometer was charged with mixture of ice CH_3COOH (110 mL) and distilled water (55 mL), and then 3,3′,4,4′-tetraaminodiphenyl methane (22 g, 0.0964 mol) was added. Flask content was heated under stirring until complete dissolution of solids. Solution was cooled to 13–14°C, and then the cooling bath was removed. After that, solution of $NaNO_2$ (18 g, 0.261 mol) in distilled water (40 mL) was quickly added to the flask content under stirring. Temperature of flask content rose up to 60–70°C. The content was stirred for about 1 h, with a suspension precipitating throughout this period. The suspension was poured into cold distilled water (500–600 mL) and stirred. The product was filtered off and rinsed with distilled water and dried *in vacuo* at 50–60°C. Crude 5,5′-bisbenzotriazol methane was dissolved in ice CH_3COOH under heating and refluxed in the presence of small quantities of distilled water and charcoal. After a hot filtration, filter liquor was poured into cold distilled water (500–600 mL) and stirred. Precipitated 5,5′-bisbenzotriazol methane was filtered off, rinsed several times with distilled water, and dried *in vacuo* at 70–80°C. 19.02 g (85%), mp 240–241°C; ^{1}H NMR (DMSO-d_6), δ 4.29 (s, 2H, CH_2), 7.34–7.42 (d, $J = 19.5$ Hz, 2H, phenyl protons), 7.78 (s, 2H, phenyl protons), 7.81–7.89 (d, $J = 19.5$ Hz, 2H, phenyl protons), 15.46 ppm (br s, 2H, NH). ir: CH_2 1400, 3070, 2950, NH 3333–3400 cm^{-1}. *Anal.* Calcd for $C_{13}H_{10}N_6$: C, 62.39; H, 4.03; N, 33.58. Found: C, 62.15; H, 3.90; N, 33.16.

5,5′-Bisbenzotriazol oxide was synthesized similar to 5,5′-bisbenzotriazol methane, with the charge of 3,3′,4,4′-tetraaminodiphenyl ether being 22.2 g (0.0964 mol). 15.72 g (82%), mp 263–264°C. ^{1}H NMR (DMSO-d_6), δ 7.34–7.42 (d, $J = 19.5$ Hz, 2H, phenyl protons), 7.78 (s, 2H, phenyl protons), 7.81–7.89 (d, $J = 19.5$ Hz, 2H, phenyl protons), 15.54 ppm (br s, 2H, NH). ir: NH 3325–3410, −O– 1247 cm^{-1}. *Anal*. Calcd for $C_{12}H_8N_6O$: C, 57.14; H, 3.20; N, 33.32. Found: C, 57.03; H, 3.28; N, 33.12.

5,5′-Bisbenzotriazol sulfone. The synthesis must be carried out in a fume chamber because of an evolution of nitrous oxides. 3,3′,4,4′-Tetraaminodiphenyl sulfone (10 g, 0.0336 mol) and ice CH_3COOH (85 mL) were placed into a 1.5 L three-neck flask equipped with a stirrer, an inlet and an outlet of argon, and a thermometer. Flask content was heated up to 50°C under stirring until the complete dissolution of solids. Then, at 50°C, solution of $NaNO_2$ (7 g, 0.102 mol) in distilled water (20 mL) was quickly added to the flask content intensively stirred. After addition of $NaNO_2$ solution, reaction masse boils up sharply ("Careful! Eye protectors and protective gloves must be used!")

The content was gradually cooled to room temperature, with a suspension precipitating throughout this period. Then the suspension was filtered off and rinsed with distilled water several times and dried *in vacuo* at 50–60°C. Crude 5,5′-bisbenzotriazol sulfone was dissolved in HCOOH under heating and refluxed in the presence of charcoal. After a hot filtration, filter liquor was cooled gradually to room temperature. Fine crystals were filtered off and rinsed with HCOOH. The operation of the purification was carried out once again. After purification, 5,5′-bisbenzotriazol sulfone was dried *in vacuo* at 70–80°C. 8.42 g (85%), mp 292–294°C. ^{13}C NMR (DMSO-d_6) δ 115.18, 118.00, 124.2, 132.66, 135.72, 140.22 ppm (aromatic carbons). *Anal.* Calcd for $C_{12}H_7N_6O_2S$: C, 48.00; H, 2.69; N, 27.99. Found: C, 47.88; H, 2.78; N, 27.82.

1-(2,5-dioxo-1-phenylpyrrolidin-3-yl)-benzotriazole 1. 1H-Benzotriazole (1.1913 g, 0.01 mol) and *N*-phenylmaleimide (1.7937 g, 0.01 mol) were carefully mixed and put in a fluoroplast reactor. Then the reactor was placed into a bath heated up to 120°C. Temperature of the bath was risen up to 220°C. Reaction mixture was held at 220°C for 5 min, then cooled to ambient temperature, and removed from the reactor. The product was dissolved in a mixture of HCOOH and H_2O, refluxed for 10 min in the presence of activated charcoal, then filtered off. The pH of mother solution was adjusted to about 5 with 0.5% aqueous NaOH. Precipitate was filtered off, dissolved in boiling ethanol, and gradually cooled. The fine crystals were filtered off and dried

in vacuo at 50–70°C. 2.12 g (71%), mp 145–147°C. ir: CO 1717.5, C–N–C 1187.5, CH_2 and CH 2930, 2979 cm^{-1}. *Anal*. Calcd for $C_{16}H_{12}N_4O_2$: C, 65.74; H, 4.14; N, 19.17. Found: C, 65.90; H, 4.15; N, 18.98. ^{13}C NMR peaks are shown in Table 1.

General procedure for synthesis of bis[3-(benzotriazolyl) succinimides] 2a–c. 1H-Benzotriazole (2.3826 g, 0.02 mol) and either *N*,*N*′-hexamethylene–bismaleinimide (2.7629 g, 0.01 mol) (for **2a**), or 4,4′-diphenylmethane–bismaleinimide (3.5835 g, 0.01 mol) (for **2b**), or 4,4′-bis(maleimido)-diphenyl ether (3.6032 g, 0.01 mol) (for **2c**) were placed in a fluoroplast reactor. Reaction was carried out as described in the preceding text for **1**. Purification of **2a-c** was also carried out as described for **1**. The products were precipitated as amorphous powders after purification, then filtered off, and dried *in vacuo* at 50–70°C. ^{13}C NMR peaks of **2a–c** are shown in Table 1. **2a**. 4.37 g (85%). ir: CO 1714, CH_2 and CH 2941, 2860, *ortho*-phenylene 746 cm^{-1}. *Anal*. Calcd for $C_{26}H_{26}N_8O_4$: C, 60.69; H, 5.09. Found: C, 60.49; H, 4.98. **2b**. 5.25 g (88%). ir: CO 1721, 1731, CH_2, and CH 2944, 2854, *ortho*-phenylene 746, C–N–C 1386 cm^{-1}. *Anal*. Calcd for $C_{33}H_{24}N_8O_4$: C, 66.44; H, 4.05. Found: C, 66.53; H, 4.13. **2c**. 5.45 g (91%). ir: CO 1715, 1732, −O– 1242, *ortho*-phenylene 746, C–N–C 1393 cm^{-1}. *Anal*. Calcd for $C_{32}H_{22}N_8O_5$: C, 64.21; H, 3.70. Found: C, 64.46; H, 3.74.

General procedure for synthesis of bis[(N-phenylsuccinimid-3-yl)benzotriazoles] 3a–c. A fluoroplast reactor was charged with 5,5′-bisbenzotriazol methane (for **3a**) (1.2513 g, 0.005 mol) and either 5,5′-bisbenzotriazol oxide (for **3b**) (1.2612 g, 0.005 mol) or 5,5′-bisbenzotriazol sulfone (for **3c**) (1.5015 g, 0.005 mol), and *N*-phenylmalrimide (1.7317 g, 0.01 mol) were placed in a fluoroplast reactor. Reaction was carried out as described in the preceding text for **1**. Purification of **3a-c** was also carried out as described for **1**. The products were precipitated as amorphous powders after purification, then filtered off, and dried *in vacuo* at 50–70°C. **^{13}C** NMR peaks of **3a–c** are shown in Table 1.

Compound **3a**. 2.24 g (75%). ir: CO 1724, 1795, CH_2 and CH 2787–3062 cm^{-1}. *Anal*. Calcd for $C_{33}H_{24}N_8O_4$: C, 66.44; H, 4.05. Found: C, 66.68; H, 4.13. Compound **3b**. 2.15 g (72%). *Anal*. Calcd for $C_{32}H_{22}N_8O_5$: C, 64.21; H, 3.70. Found: C, 64.45; H, 3.68. Compound **3c**. 2.20 g (68%). *Anal*. Calcd for $C_{32}H_{22}N_8O_6S$: C, 59.44; H, 3.43. Found: C, 59.62; H, 3.42.

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