Russian Chemical Bulletin, International Edition, Vol. 50, No. 12, pp. 2411—2415, December, 2001 2411

2,4,6,8-Tetraiodo-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione as a mild and convenient reagent for iodination of aromatic compounds

V. K. Chaikovski,[a] V. D. Filimonov,[a]* A. Yu. Yagovkin,[a] and V. D. Ogorodnikov[b]

[a]*Tomsk Polytechnical University,*
30 prosp. Lenina, 634004 Tomsk, Russian Federation.
Fax: +7 (382 2) 41 5235. E-mail: filim@org.chtd.tpu.edu.ru
[b]*Institute of Petroleum Chemistry, Siberian Branch of the Russian Academy of Sciences,*
3 prosp. Akademicheskii, 634021 Tomsk, Russian Federation

2,4,6,8-Tetraiodo-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (tetraiodoglycoluril) is a convenient reagent for preparative iodination of benzene, alkylbenzenes, polycyclic hydrocarbons, aromatic amines, and phenol ethers in organic solvents under mild conditions.

Key words: iodination, aromatic compounds, tetraiodoglycoluril.

Recently,[1] we have reported the successful use of 2,4,6,8-tetraiodo-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (tetraiodoglycoluril, TIG) in 90% sulfuric acid for iodination of deactivated aromatic compounds. Together with the superelectrophilic reagent (the reagent "I^+") generated in H_2SO_4 upon the reaction of iodine chloride with silver sulfate,[2,3] the TIG—H_2SO_4 system can be regarded as one of the most powerful reagents able to iodinate deactivated aromatic compounds. For example, nitrobenzene is converted[1] into *m*-iodonitrobenzene in 75% yield over a period of 1.5 h at 0 °C. However, the use of concentrated H_2SO_4 as the medium limits the scope of application of TIG in the case of activated arenes due to side processes such as protonation, resinification, sulfonation, *etc.* For example, our experiments have shown that the use of TIG in H_2SO_4 for iodination of alkylbenzenes, aromatic amines, and polycyclic hydrocarbons is faced with serious problems of product isolation.

The purpose of this work is to study the iodination capacity of TIG in organic solvents.

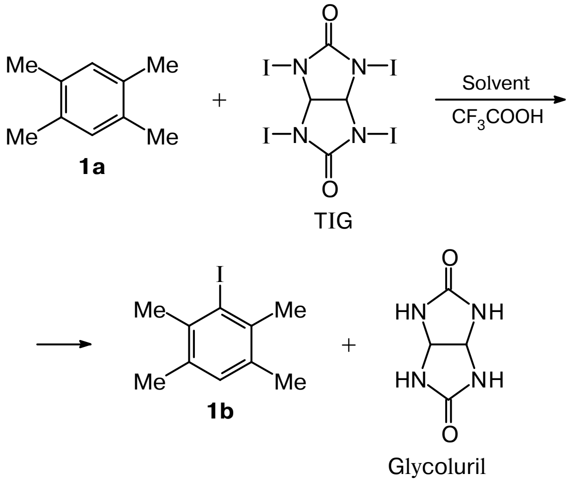
Results and Discussion

The efficacy of the reaction in various solvents was studied in relation to 1,2,4,5-tetramethylbenzene (durene) (**1a**). It was shown that in aprotic solvents at ~20 °C, iodination of arenes by TIG was very slow but the addition of concentrated H_2SO_4 or CF_3COOH markedly accelerated the reaction

In all experiments, the acid was added to a solution of the substrate and TIG (method *A*) and the reaction proceeded rather fast in most solvents. In methanol, ethanol, acetic acid, or acetonitrile, precipitation of iododurene **1b** is usually observed 1—2 min after the addition of the acid, its yield being 60—77%. In acetic acid, the reaction occurred without H_2SO_4 but even after 5.5 h the yield of iododurene **1b** was only 24%. The use of trifluoroacetic acid instead of acetic acid, also in the absence of H_2SO_4, increased substantially the reaction rate. In this case, the precipitate did appear after 1—2 min, but the product **1b** formed in a relatively high yield (64%) contained impurities that markedly decreased its melting point. Since H_2SO_4 is virtually insoluble in CCl_4 or $CHCl_3$, the experiments in these solvents were carried out using CF_3COOH. The yield of crude iododurene **1b** was 62—69%. According to TLC, the product was contaminated by the starting durene **1a** and diiododurene **1c**.

It is important that only the use of CF_3COOH allows one to isolate unsubstituted 2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (glycoluril), which precipitates after dilution of the reaction mixture with water and can be recovered in an almost quantitative yield by filtration (Scheme 1).

Scheme 1



Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 12, pp. 2302—2306, December, 2001.

When the reaction is carried out in H_2SO_4,[1] or when H_2SO_4 is added to the reaction mixture in a solvent, the glycoluril formed passes during the workup to the aqueous phase as the sulfate and can be isolated upon neutralization.

Dioxane, exhibiting a high solubilizing capacity with respect to both the starting alkylbenzenes and iodination products, proved to be the solvent of choice for the introduction of one, two, or three iodine atoms in the substrates (S) and their complete conversion; the ratios [S] : [TIG] being 4 : 1, 2 : 1, and ~1.3 : 1, respectively. In dioxane, 1,4-diiododurene (**1c**) was obtained as readily as monoiododurene **1b** (Table 1). Similarly, *p*- and *m*-xylenes (**2a** and **3a**) are easily converted into mono- and disubstitution products, namely, 1-iodo-2,5-dimethylbenzene (**2b**), 1,4-diiodo-2,5-dimethylbenzene (**2c**), 1-iodo-2,4-dimethylbenzene (**3b**), and 1,5-diiodo-2,4-dimethylbenzene (**3c**). Mesitylene (**4a**) gives mono-, di-, and triiodomesitylenes (**4b**, **4c**, and **4d**, respectively) in good yields. The last-mentioned product can be obtained by using 1 mmol of TIG per mmol of substrate **4a**.

Some polycyclic aromatic hydrocarbons can be iodinated quite successfully using TIG in dioxane. It is known that iodination of this type of compounds is often faced with problems related to the possibility of one-electron transfer[12] and generation of radical ions, giving rise to side products. It is also known that polycyclic hydrocarbons react with iodine to give charge transfer complexes, which inhibit the iodination process. In some cases, this can be avoided by adding small amounts of n-electron-donating compounds (dioxane, water, THF, DMF).[8,13]

In the dioxane—H_2SO_4 system, TIG readily iodinates biphenyl (**5a**), *p*-terphenyl (**6a**), and fluorene (**7a**), as well as naphthalene (**8a**) and pyrene (**9a**) (see Table 1). In all cases, except for naphthalene (**8a**), which was converted into 1-iodonaphthalene (**8b**), much better results were attained in the synthesis of diiodinated derivatives: 4,4′-diiodobiphenyl (**5b**), 4,4′-diiodo-*p*-

Table 1. Iodination of aromatic compounds with tetraiodoglycoluril in organic solvents

Substrate	Method	Solvent	*T* /°C	τ /min	Product	Yield (%)	M.p./°C (solvent) or [b.p./°C]*	
							Experiment	Published data
Durene (**1a**)	*A*	Dioxane	20	15	Iododurene (**1b**)	74	78—79 (EtOH)	78—79[4]
				20	Diiododurene (**1c**)	75	138—139 (EtOH)	139[4]
p-Xylene (**2a**)	*A*	Dioxane	20	15	1-Iodo-2,5-dimethylbenzene (**2b**)	65	[227—230]	[230][5]
				30	1,4-Diodo-2,5-dimethylbenzene (**2c**)	53	101—102 (EtOH)	101—102[6]
m-Xylene (**3a**)	*A*	Dioxane	20	15	1-Iodo-2,4-dimethylbenzene (**3b**)	67	[229—232]	[232][5]
				30	1,5-Diiodo-2,4-dimethylbenzene (**3c**)	45	70—71 (EtOH)	70—71[6]
Mesitylene (**4a**)	*A*	Dioxane	20	15	Iodomesitylene (**4b**)	63	30 (MeOH)	30[6]
				20	Diiodomesitylene (**4c**)	72	73—74 (EtOH)	74—75[6]
				30	Triodomesitylene (**4d**)	51	207—208 (DMF)	207—209[6]
Biphenyl (**5a**)	*A*	Dioxane	20	20	4,4′-Diiododiphenyl (**5b**)	85	203—204 (PrOH)	204[4]
Terphenyl (**6a**)	*A*	Dioxane	20	20	4,4′-Diiodoterphenyl (**6b**)	80	305—307 (toluene)	305—307[7]
Fluorene (**7a**)	*A*	Dioxane	20	20	2,7-Diiodofluorene (**7b**)	75	208—209 (toluene)	210[4]
Naphthalene (**8a**)	*A*	Dioxane	20	30	1-Iodonaphthalene (**8b**)	52	[302—305]	[305][5]
Pyrene (**9a**)	*A*	Dioxane	20	20	1,6-Diiodopyrene (**9b**)	15	261—263 (chlorobenzene)	263—264[8]
Anthracene (**10a**)	*Б*	Benzene**	20	30	9-Iodoanthracene (**10b**)	25	78—80 (benzene—MeOH, 1 : 2)	78—80[9]
		Chloroform**	20	30	9,10-Diiodoanthracene (**10c**)	17	250—252 (toluene)	254—255[10]
Benzene (**11a**)	*A*	Dioxane	20	20	Iodobenzene (**11b**)	56	[185—188]	[186—187.5][4]
		Benzene**	20	20	Iodobenzene (**11b**)	44	[185—188]	[186—187.5][4]
Aniline (**12a**)	*Б*	Ethanol	0	10	4-Iodoaniline (**12b**)	52	67—68 (hexane)	67—68[5]
Diphenylamine (**13a**)	*Б*	Ethanol	0	10	4,4′-Diiododiphenylamine (**13b**)	51	122—124	125—126[11] (80% EtOH)
Acetanilide (**14a**)	*Б*	Ethanol	20	20	4-Iodoacetanilide (**14b**)	67	183—184 (EtOH)	183—184[5]
p-Nitroaniline (**15a**)	*Б*	Ethanol	20	15	2-Iodo-4-nitroaniline (**15b**)	71	114—115 (Pr^iOH)	115[5]
Anisole (**16a**)	*Б*	Ethanol	20	10	4-Iodoanisole (**16b**)	73	51—52 (MeOH)	51—52[5]
Diphenyl ether (**17a**)	*Б*	Ethanol	20	10	4,4′-Diiododiphenyl ether (**17b**)	38	138—139 (EtOH)	140[4]
Dibenzofuran (**18a**)	*A*	Acetic acid	20	10	3,6-Diiododibenzofuran (**18b**)	36	172—173 (EtOH)	172—173[4]

* Under a pressure of 760 Torr.

** CF_3COOH was used as the catalyst.

terphenyl (**6b**), and 2,7-diiodofluorene (**7b**). The attempts at monoiodination (molar ratio [S] : [TIG] = 4 : 1) resulted in mixtures of mono- and diiodo-substituted products. According to TLC, pyrene **9a** gave a mixture of two diiodopyrene isomers, presumably,[8] 1,6- and 1,8-disubstituted ones, in 82% yield. By recrystallization of this mixture from chlorobenzene, individual 1,6-diiodopyrene (**9b**) was isolated (15%).

The iodination of anthracene (**10a**) is known to be especially difficult as this substrate not only forms nonpolar complexes with iodine, which hamper iodination, but is also oxidized rather easily to give anthraquinone.[14] The reaction with iodine chloride in organic solvents proceeds as selective chlorination instead of iodination.[12] Therefore, iodoanthracenes are usually prepared by substitution of iodine for the bromine atoms in bromoanthracenes.[10,15] Direct synthesis of 9-iodoanthracene (**10b**) was performed by treatment of anthracene with iodine in benzene in the presence of Cu^{II} salts on alumina as the catalyst.[9]

In attempts to iodinate anthracene (**10a**) with TIG in dioxane, ethanol, or 80% acetic acid in the presence of H_2SO_4 or CF_3COOH, we detected only traces of 9-iodoanthracene (**10b**). In CF_3COOH, iodination of compound **10a** did not take place.

With benzene (**11a**) as the solvent, quite an unexpected result was obtained. At 20 °C with the H_2SO_4 additive, only did the solvent **11a** undergo iodination. Iodobenzene (**11b**), *p*-diiodobenzene (**11c**), and the starting anthracene (**10a**) were detected in the reaction mixture by TLC. When H_2SO_4 was replaced by CF_3COOH and TIG was added in three portions, complete conversion of anthracene **10a** in benzene was reached over a period of 30 min. Monoiodoanthracene **10b** was isolated from the reaction mixture in 25% yield, no iodobenzene was detected by chromatography.

Direct synthesis of 9,10-diiodoanthracene (**10c**) has been mentioned (without indication of the yields or characteristics) in a patent,[16] in which iodination of polycyclic arenes with iodine in the presence of ammonium persulfate was proposed. Our attempts to reproduce these results failed. However, we found that diiodide **10c** can be isolated in 11% yield upon treatment of anthracene with 4 equiv. of TIG in benzene in the presence of CF_3COOH. According to TLC, iodobenzene, anthraquinone, and some unidentified products were also present in the reaction mixture. Somewhat better results in the synthesis of diiodide **10c** were attained in $CHCl_3$ (17%) (see Table 1).

It is noteworthy that benzene **11a** itself can be iodinated rather successfully in dioxane with H_2SO_4 as the additive to give iodobenzene **11b** (56%) (see Table 1); however, further iodination of product **11b** does not occur even when it is kept with 1 equiv. of TIG in dioxane for 0.5 h at 20 °C. However, treatment of solvent **11a** with TIG (30-fold molar excess of benzene with respect to TIG) in the presence of H_2SO_4 gave a solid product containing (TLC) monoiodobenzene (**11b**, traces) and di-, tri-, and tetraiodobenzenes was obtained over the same period of time. From this mixture, 1,4-diiodobenzene (**11c**) (yield 24%) and 1,2,4,5-tetraiodobenzene (**11d**) (yield 3%) were isolated. Under the same conditions but using CF_3COOH as the catalyst, only iodobenzene **11b** was obtained in a yield of 44% (see Table 1). Polyiodination of benzene did not take place in this case.

The efficiency of TIG as a mild iodination reagent is confirmed by the results of experiments on iodination of aromatic amines: aniline (**12a**), diphenylamine (**13a**), acetanilide (**14a**), and *p*-nitroaniline (**15a**) in ethanol (see Table 1). Since aromatic amines are highly prone to oxidation, the addition of TIG in two to four portions proved more efficient (method *B*) (see Table 1). 4-Iodoaniline (**12b**) and 4,4′-diiododiphenylamine (**13b**) were prepared in quite satisfactory yields at 0 °C. Similarly, but at ~20 °C, less active arylamines **14a** and **15a** underwent iodination to give 4-iodoacetanilide (**14b**) and 2-iodo-4-nitroaniline (**15b**), respectively.

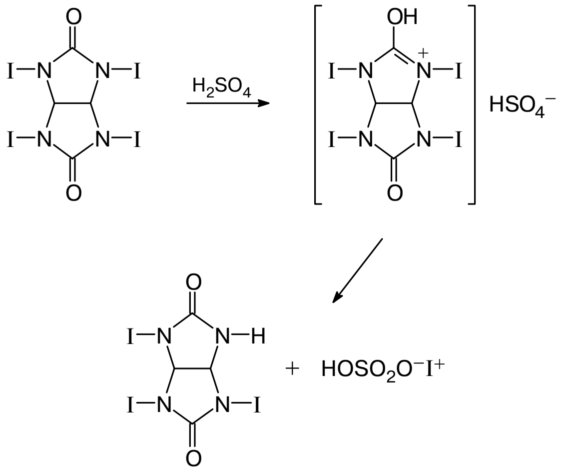
Method *B* is also preferred for iodination of anisole (**16a**) and diphenyl ether (**17a**) in ethanol (see Table 1) and provides the possibility of preparing 4-iodoanisole (**16b**) and 4,4′-diiododiphenyl ether (**17b**). In the case of dibenzofuran (**18a**), method *B* was inefficient. 3,6-Diiododibenzofuran (**18b**) was prepared in a relatively low yield (29%) in acetic acid. By using method *A*, the yield of compound **18b** was increased to 36% (see Table 1).

Study of the mechanism of iodination by TIG was beyond the scope of this work; nevertheless, we tried to answer a key question significant for the understanding of the reaction mechanism, namely, whether iodination occurs upon direct transfer of iodine from the TIG molecule to the substrate, or TIG acts only as the source for the formation of some electrophilic iodine species in the solution. The latter type of transformation takes place, for example, in the case of *N*-iodosuccinimide (rather close analog of TIG); in trifluoromethanesulfonic acid, this gives the solvated iodonium cation $CF_3SO_2O^-I^+$, which acts directly as the iodinating agent.[17]

To solve this problem tentatively, we studied the ^{13}C NMR spectra of TIG and glycoluril in D_2SO_4. It was found that as soon as 2—3 min after dissolution of TIG in concentrated D_2SO_4 at 20 °C the spectrum exhibited signals for the CO and CH groups at δ 165.9 and 71.5, which coincide completely with the corresponding signals in the spectrum of glycoluril in D_2SO_4. Dehalogenation of TIG occured rapidly, and no intermediates containing one to three iodine atoms could be detected in the spectra. However, in the ^{13}C NMR spectrum of a solution of TIG in D_2SO_4, no signal at δ 163.3 corresponding to the carbonyl group of protonated urea can be found even two days after dissolution; this suggests that glycoluril itself does not decompose under these conditions.

Thus, TIG, like *N*-iodosuccinimide,[17] functions as a source of I^+ in the reaction mixture. Presumably, elimination of iodine is a stepwise process including protonation in each of the four steps. One step is shown in Scheme 2.

Scheme 2



It should be noted that in the absence of an acid, a solution of TIG in, for example, DMSO-d_6 remains unchanged for two days at 25 °C.

The form of existence of iodine-containing electrophilic species in solutions of TIG in sulfuric and trifluoroacetic acids can be judged resorting to published data.[17,18] In all probability, in sulfuric acid, they are solvated forms $HOSO_2O^-I^+$ and/or $(I^{+-}O)_2SO_2$, and in trifluoroacetic acid, this is $CF_3COO^-I^+$.

It is also noteworthy that, unlike iodination with the TIG—H_2SO_4 system where the maximum conversion of the substrate requires 2 equiv. of TIG for the introduction of one iodine atom,[1] iodination in organic solvents takes normally one equivalent of the reagent with respect to the substrate (0.25 moles of TIG per mole of the substrate) irrespective of the nature of the acid.

Thus, TIG is one of few iodinating reagents able to iodinate under mild conditions a variety of aromatic compounds ranging from highly deactivated to activated and polycyclic ones. However, the iodination conditions (the solvent, the type of acid catalyst, the order of addition of the reactants) are specific for each type of aromatic substrates.

Experimental

The reactions were monitored and the purity of compounds was checked by TLC on Silufol UV-254 plates in hexane (or in benzene, for compounds **12a,b—15a,b**); substances were visualized in the UV light. ^{13}C NMR spectra were recorded on a Tesla BS-587 spectrometer using D_2O as the external standard. Tetraiodoglycoluril was prepared by a previously published procedure.[19]

Iodination of compounds 1a—9a, 11a, and 18a (method *A*). TIG (1.62 g, 2.5 mmol) was added to a solution of aromatic substrate (10 mmol) in 15 mL of the corresponding solvent, and 3 mL of concentrated H_2SO_4 was added with cooling. The mixture was stirred at 20 °C for the period of time indicated in Table 1. In the synthesis of di- or triiodo-substituted products (**1c—3c, 4c,d**, **5b—7b**, **9b**, and **18b**), the amount of the substrate taken was decreased two- or three-fold. The reaction mixture was diluted with 100 mL of water. Liquid products **2b**, **3b**, and **11b** were extracted with chloroform, the extracts were dried with $CaCl_2$ and concentrated, and the residue was distilled. Solid compounds were filtered off, dried, and, if required, crystallized from the appropriate solvent. Product **8b** was extracted with hexane, the extract was dried with $CaCl_2$, filtered through a layer of Al_2O_3, and distilled *in vacuo*. To isolate diiodopyrene **9b**, the reaction mixture was chromatographed on a short column with Al_2O_3 using hexane as the eluent. Three recrystallizations from chlorobenzene gave individual 1,6-diiodopyrene **9b**.

9-Iodoanthracene (10b) (method *B*). CF_3COOH (3 mL) was added to a solution of compound **10a** (0.89 g, 5 mmol) in 20 mL of benzene. At 20 °C, TIG (1.62 g, 2.5 mmol) was added in three portions over a period of 2—3 min, and the mixture was stirred for 30 min at 20 °C. A 3% solution of Na_2SO_3 (50 mL) was added to the reaction mixture, and the glycoluril that precipitated (0.35 g) was filtered off. The benzene layer was separated and dried with $CaCl_2$, the solvent was evaporated *in vacuo*, and the residue was chromatographed on a short column with Al_2O_3 using hexane as the eluent. After removal of hexane *in vacuo*, iodide **10a** was crystallized from a methanol—benzene mixture (2 : 1).

9,10-Diiodoanthracene (10c) (method *B*). CF_3COOH (3 mL) was added to a solution of compound **10a** (0.45 g, 2.5 mmol) in 20 mL of $CHCl_3$. At 20 °C, TIG (1.62 g, 2.5 mmol) was added in four portions over a period of 30 min. Chloroform (10 mL) and a 3% solution of Na_2SO_3 (50 mL) were added to the reaction mixture. The precipitate of glycoluril (0.34 g) was filtered off. The chloroform layer was separated, dried with $CaCl_2$, the solvent was evaporated *in vacuo*, and the residue was chromatographed on a short column with Al_2O_3 using hexane as the eluent.

Reaction of benzene (11a) with TIG. TIG (1.62 g, 2.5 mmol) and H_2SO_4 (4 mL) were added to 27 mL of benzene (**11a**), the mixture was stirred for 30 min at 20 °C and washed with water, the benzene layer was separated and dried with $CaCl_2$. Evaporation of benzene *in vacuo* gave 1.61 g of a solid. This product was extracted into 5 mL of boiling EtOH. Cooling of the extract gave 1,4-diiodobenzene (**11c**), yield 0.79 g (24%), m.p. 127—128 °C (*cf.* Ref. 4: m.p. 130—131 °C). The residue insoluble in boiling EtOH was 1,2,4,5-tetraiodobenzene (**11d**), yield 0.17 g (3%), m.p. 250—252 °C (*cf.* Ref. 16: m.p. 254 °C). When CF_3COOH was used, iodobenzene (**11b**) was obtained as the only product.

Iodination of amines (12a—15a) and phenol ethers (16a, 17a) (method *B*). Sulfuric acid (1.5 mL) was added to a solution of substrate **13a—17a** (5 mmol) (or 10 mmol of aniline **12a**) in 15 mL of EtOH, and TIG (1.62 g, 2.5 mmol) was added in three portions over a period of 2—3 min. Iodination of compounds **12a** and **13a** was carried out at 0 °C; other compounds were iodinated at 20 °C. After completion of the reaction, a 5% solution of NaOH (30 mL) was added, and the products were filtered off, washed with water, dried, and crystallized. To isolate compounds **16b** and **17b**, the reaction mixture was diluted with 50 mL of 3% Na_2SO_3 and the products were filtered off, dried, and crystallized.

This work was supported by the Russian Foundation for Basic Research (Project No. 00-03-32812a).

References

1. V. K. Chaikovski, V. D. Filimonov, A. Y. Yagovkin, and T. S. Kharlova, *Tetrahedron Lett.*, 2000, **41**, 9101.
2. V. K. Chaikovski, T. S. Kharlova, V. D. Filimonov, and T. A. Saryucheva, *Synthesis*, 1999, 748.
3. V. K. Chaikovski, T. S. Kharlova, and V. D. Filimonov, *Izv. Akad. Nauk, Ser. Khim.*, 1999, 1303 [*Russ. Chem. Bull.*, 1999, **48**, 1291 (Engl. Transl.)].
4. *Sintezy iodistykh organicheskikh soedinenii* [*Syntheses of Organic Iodine Compounds*], Eds. L. M. Yagupol´skii, A. N. Novikov, and E. B. Merkushev, Izd-vo Tomsk Gos. Un-ta, Tomsk, 1976, 25 (in Russian).
5. *Dictonary of Organic Compounds*, Eds. I. Heibron and H. M. Bunbury, London, 1946, **1**, **2**.
6. E. B. Merkushev, *Preparativnye sintezy iodaromaticheskikh soedinenii* [*Preparative Syntheses of Iodoaromatic Compounds*], Izd-vo Tomsk. Gos. Un-ta, Tomsk, 1985, 72 (in Russian).
7. E. B. Merkushev and N. D. Yudina, *Zh. Org. Khimii*, 1981, **17**, 2598 [*J. Org. Chem. USSR*, 1981, **17** (Engl. Transl.)].
8. V. K. Chaikovski, and A. N. Novikov, *Zh. Org. Khim.*, 1984, **20**, 1482 [*J. Org. Chem. USSR*, 1984, **20** (Engl. Transl.)].
9. M. Kodomari, N. Amanokura, K. Takeuchi, and S. Yoshitomi, *Bull. Chem. Soc. Jpn.*, 1992, **65**, 306.
10. F. B. Duerr, Y.-S. Chang, and W. A. Czarnik, *J. Org. Chem.*, 1988, **53**, 2120.
11. M. I. Bardamova, Z. P. Trotsenko, and I. L. Kotlyarevskii, *Izv. Akad. Nauk SSSR, Ser. Khim.*, 1982, 1184 [*Bull. Acad. Sci. USSR, Div. Chem. Sci.*, 1982, **31** (Engl. Transl.)].
12. D. E. Turner, R. F. O´Malley, J. Sardella, L. S. Barinelli, and P. Kaul, *J. Org. Chem.*, 1994, **59**, 7335.
13. V. K. Chaikovski, V. T. Novikov, A. N. Novikov, and F. Sh. Garifullina, *Zh. Obshch. Khim.*, 1985, **55**, 893 [*J. Gen. Chem. USSR*, 1985, **55** (Engl. Transl.)].
14. E. Klar. *Politsiklicheskie uglevodorody* [*Polycyclic Hydrocarbons*], Khimiya, Moscow, 1971, **1**, 290 (in Russian).
15. H. Suzuki, A. Kondo, M. Inouye, and T. Ogawa, *Synthesis*, 1986, 121.
16. I. M. Trevor, I. P. Szabo, and R. T. Sam, US Pat., 4240987.
17. G. A. Olah, Qi. W. Wang, G. Sandford, and G. K. S. Prakash, *J. Org. Chem.*, 1993, **58**, 3194.
18. P. Lulinski and L. Skulski, *Bull. Chem. Soc. Jpn.*, 2000, **73**, 951.
19. A. Yu. Yagovkin, A. A. Bakibaev, and E. L. Bystritskii, *Khimiya Geterotsikl. soedinenii*, 1995, 1695 [*Chem. Heterocycl. Compd.*, 1995 (Engl. Transl.)].

Received November 4, 2000
in revised form May 10, 2001