ISSN 1070-3632, Russian Journal of General Chemistry, 2012, Vol. 82, No. 2, pp. 310–316. © Pleiades Publishing, Ltd., 2012.
Original Russian Text © L.G. Golubyatnikova, G.R. Anpilogova, R.A. Khisamutdinov, Yu.I. Murinov, 2012, published in Zhurnal Obshchei Khimii, 2012, Vol. 82, No. 2, pp. 315–321.

Extraction of Hydrochloric and Nitric Acid with 1-{[2-(2,4-Dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]-methyl)-1*H*-1,2,4-triazole and (*RS*)-1-(4-Chlorophenyl)-4,4-dimethyl-3-(1*H*-1,2,4-triazol-1-yl-methyl)-pentan-3-ol

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Received February 21, 2011

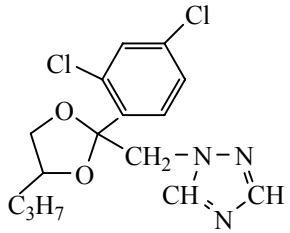
Abstract—Extraction of hydrochloric and nitric acid with 1-{[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]-methyl}-1*H*-1,2,4-triazole (propiconazole) and hydrochloric acid with (*RS*)-1-(4-chlorophenyl)-4,4-dimethyl-3-(1*H*-1,2,4-triazol-1-yl-methyl)-pentan-3-ol (tebuconazole) was studied. It is established that extraction of acids proceeds with the formation of monosolvates as an exothermic process. Effective extraction constants of acids are evaluated. By means of the IR and ^{1}H NMR spectroscopy it was shown that the proton-accepting center of tebuconazole is N^4 atom of the triazole ring.

DOI: 10.1134/S1070363212020223

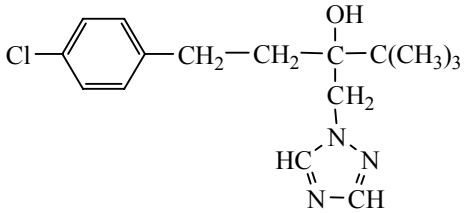
The investigation of extractive properties of system pesticides of the triazole series such as 1-{[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]-methyl)-1*H*-1,2,4-triazole (propiconazole, **L$_1$**) and (*R,S*)-1-(4-chlorophenyl)-4,4-dimethyl-3-(1*H*-1,2,4-triazol-1-yl-methyl)-pentan-3-ol (tebuconazole, **L$_2$**) widely used in agriculture showed that these reagents are effective and selective extragents of the platinum group metals, gold(III), copper(II), and gallium(III) for isolation them from the hydrochloric acid solutions [1–5]. Propiconazole also selectively and effectively extracts palladium(II) from the nitric and nitro-nitrate solutions modeling the composition of raffinates of the PUREX-process [6].

Weakly basic and complex-forming properties of reagents are caused by the presence of triazole ring in their structure.

For establishing the mechanism of extraction of metals data on the behavior of reagents under the extraction conditions are required. They are also useful for the choice of washing solution for the extract of the target metal and re-extractants. But extraction of mineral acids with the derivatives of 1,2,4-triazole is poorly studied. Khisamutdinov et al. [1] by means of the ^{1}H and ^{13}C NMR spectroscopy have shown that in the course of interaction of propiconazole with hydrochloric and nitric acids the reagent is protonated by N^4 atom of the triazole ring.



L$_1$



L$_2$

The aim of this work is the investigation of extraction of hydrochloric acid with propiconazole $\mathbf{L_1}$ and tebuconazole $\mathbf{L_2}$ (toluene with 15 vol % of *n*-decanol as a diluent) and of nitric acid with propiconazole (toluene as a diluent). The objects of these studies were chosen among the previously studied extraction systems possessing practical interest for the isolation and separation of metals.

In Fig. 1 the isotherms of extraction of acids are presented. The concentration of acid (*Y*) extracted with the reagent **L** was calculated from the difference in concentration of acid in extract and in the diluent in the absence of extractant. The isotherms have the *S*-shape form typical for the isolation of strong mineral acids with such weakly basic extractants as *N*-octyl-*N*-benzylaniline [7] and neutral extractants such as sulfoxides [8, 9], *N,N*-dialkylamides [10], and organophosphorus compounds [11]. Such form of isotherms is characteristic of the extraction of electrolyte dissociated in water phase with the formation of non-dissociated extracted substance [7, 12]. As known, the extracted monosolvate substances of nitric acid with dialkylsulfoxides and tributylphosphate are not dissociated in the extract [13]. Low value of equivalent electroconductivity of the tebuconazole hydrochloride solution in acetone (11.1 Sm cm^2/g-eq.) indicates a low dissociation of salt even in more polar solvent than the mixture of toluene with *n*-decanol.

It follows from Fig. 1 that the extractability of less strong nitric acid is higher than the extractability of hydrochloric acid which is caused in particular by lower hydration energy of nitrate ions as compared to chloride ones [14]. The reagents under study have very low basicity (pK_a 1.09 for propiconazole [15]). As known, the decrease in the basicity of an extractant shifts the area of effective extraction of strong mineral acids to the side of their higher concentrations in the water phase [7, 16]. The moderately and strongly basic aliphatic amines effectively extract mineral acids according to the neutralization mechanism from the low acidic solutions (at the acidity of water phase 0.1–0.5 M amines are completely protonated in the organic phase) [16, 17], but the extraction of nitric acid with propiconazole from the solution with the concentration of acid 0.5 M is insignificant (ratio of molar concentration of the extracted acid to the total starting molar concentration of reagent in the organic phase [HA]:[$\mathbf{L}_0$] = 0.025). The noticeable extraction of hydrochloric acid with tebuconazole and propiconazole occurs at the acidity of the water phase no less than

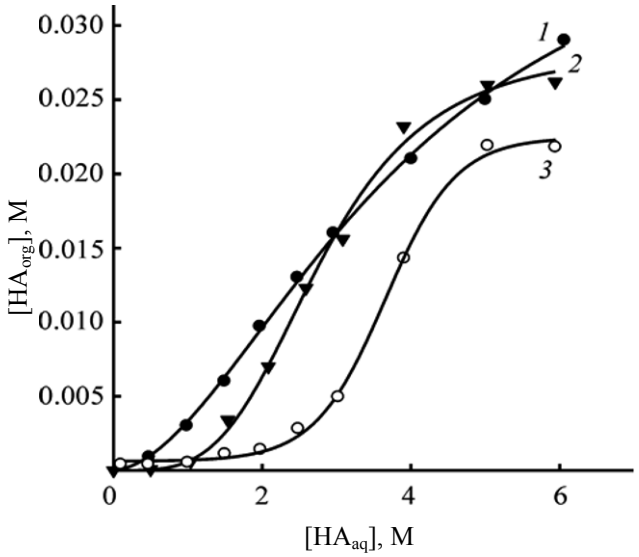


Fig. 1. Isotherms of extraction of (*1*) nitric acid with propiconazole, (*2*) hydrochloric acid with tebukobazol, and (*3*) propiconazole. [$\mathbf{L}_0$] 0.02 M.

1 and 2 M respectively ([HA]:[$\mathbf{L}_0$] ~ 0.03). Under the identical conditions the isotherm of extraction of HCl with propiconazole is shifted to more acidic area as compared to tebuconazole. It permits a suggestion that the acidity of tebuconazole is higher than of propyconazol. The extraction of acids with reagents increases with the acidity of the water phase. [HA]:[$\mathbf{L}_0$] ~ 1 is achieved at the equilibrium concentration of acid in water phase ~4 M of HNO_3 for propiconazole and ~3.9 and ~5 M of HCl for tebuconazole and propiconazole respectively. Under further increase of acidity a superstoichiometric extraction of HNO_3 with propiconazole and HCl with tebuconazole is observed. For the extraction of HCl with propiconazole in the range 5–6 M [HA]:[$\mathbf{L}_0$] ratio is 1.1 (Fig. 1).

By means of the shift of the extraction equilibrium it was established that in the range of equilibrium concentrations of HNO_3 in water phase 0.48–1.98 M and the extraction with propiconazole and in the range of equilibrium concentrations of HCl 1.54–3.08 M (extraction with propiconazole) and 1–3.03 M (extraction with tebuconazole) the value of the solvate number is equal to 1 (Fig. 2). Hence, in these ranges of acidity of water phase and the starting concentrations of extractants the extraction of acids proceeds with the preferred formation of monosolvates and can be described by the following equation.

$$\mathbf{L}_{org} + H^+_{aq} + A^-_{aq} \rightleftarrows \mathbf{L}\cdot HA_{org}. \quad (1)$$

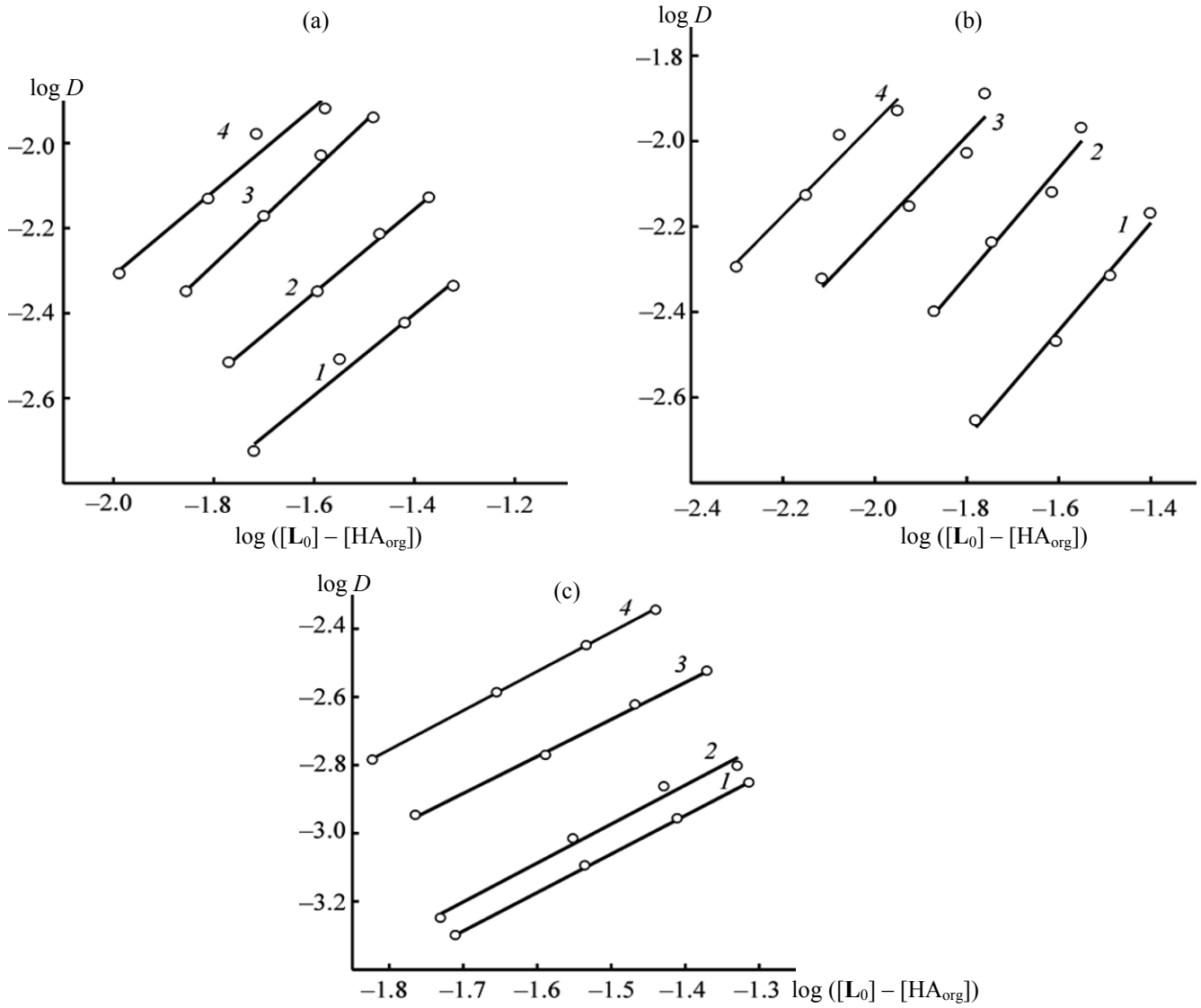


Fig. 2. Dependence of distribution coefficient of the acid D on the concentration of free extractant [$\mathbf{L}_0$]–[HA] and the equilibrium concentration of acid. (a) Propiconazole (toluene).(*1*) 0.48 M HNO_3, tan α 0.93, correlation coefficient r^2 0.981, (*2*) 0.99 M HNO_3, tan α 0.98, r^2 0.999, (*3*) 1.50 M HNO_3, tan α 1.12, r^2 0.993, (*4*) 1.98 M HNO_3, tan α 0.99, r^2 0.964. (b) Tebuconazole (toluene with 15 vol % of *n*-decanol). (*1*) 1.54 M HCl, tan α 1.26, r^2 0.989, (*2*) 2.09 M HCl, tan α 1.26, r^2 0.973, (*3*) 2.60 M HCl, tan α 1.12, r^2 0.945, (*4*) 3.08 M HCl, tan α 1.09, r^2 0.950. (c) Propiconazole (toluene with 15 vol % of *n*-decanol). (*1*) 1.00 M HCl, tan α 1.12, r^2 1.00, (*2*) 1.98 M HCl, tan α 1.14, r^2 0.990, (*3*) 2.49 M HCl, tan α 1.08, r^2 0.998, (*4*) 3.03 M HCl, tan α 1.15, r^2 1.00.

The concentrational (Fig. 3) and effective (Table 1) extraction constants of acids were calculated according to Eqs. (2) and (3) respectively.

$$\tilde{K} = [\mathbf{L}\cdot HA_{org}]/([\mathbf{L}_{org}]\cdot[H^+_{aq}]\cdot[A^-_{aq}]) = [HA_{org}]/\{([\mathbf{L}_0] - [HA_{org}])\cdot[HA_{aq}]^2\}, \quad (2)$$

$$\bar{K} = \tilde{K}\cdot\gamma_{\pm}^{-2}, \quad (3)$$

where [HA] = [$\mathbf{L}\cdot HA_{org}$], M, is the equilibrium concentration of acid extracted with the reagent; [$\mathbf{L}_{org}$] = [$\mathbf{L}_0$] – [HA] (M) is the equilibrium concentration of free extractant in the organic phase; $X = [H^+_{aq}] = [A^-_{aq}]$ (M) is the equilibrium concentration of acid in water phase; ($\gamma_{\pm}$) molar average ionic activity coefficient of acid in water phase calculated from the reference data on molal average ionic activity coefficients [18] according to the known equation [19].

In Table 1 the confidence ranges of effective extraction constants calculated from the Student

t-distribution for *P* 0.95 are presented. It is known that the increase in the polarity of diluent at the addition of ethanol modifier increases the degree of isolation of strong mineral acids with amines due to the non-specific solvation of anions [14]. Nevertheless, despite the more polar diluent the extraction constant of HCl with propiconazole is ten times smaller than the extraction constant of HNO_3. Under equal conditions the constant of extraction of hydrogen chloride with tebuconazole is higher than with propiconazole, which is caused evidently by the higher basicity of tebuconazole. The constant of extraction of mineral acids with substituted 1*H*-1,2,4-triazoles is significantly lower than the constants of extraction of acids with aliphatic amines according to neutralization mechanism (for tertiary amines $\tilde{K}$ 10^4–10^5 [17]) corresponding to very low basicity of the reagents. The comparison of the extraction ability of propiconazole with the series of known neutral extractants for the isolation of nitric acid ac-cording to the mechanism (1) showed that the constant of extraction of HNO_3 with propiconazole is somewhat lower than the constant of extraction of this acid with the solution of 0.7 M of dihexyl sulfoxide in *o*-xylene ($\tilde{K}$ = 1 in the range X = 0.50–3.41 M [20]) and 0.05 M of trioctylphosphine oxide in benzene ($\tilde{K}$ = 1.28, X = 0.09–0.66 M [21]). It is comparable with the constant of extraction of acid with the solutions of 0.5 M of dialkyl phosphonates in CCl_4 ($\tilde{K}$ = 0.10–0.51, X = 0.01– 4.40 M [20]) and to some extent exceeds the constants of extraction of acid with 0.5 M solutions of trialkyl phosphates in CCl_4 ($\tilde{K}$ = 0.10–0.18, X = 0.93–4.53 M [20]). Constants of extraction of nitric acid with alcohols and ethers [20, 22] are by two orders of magnitude less than the extraction constant for propyconazole which confirms that the protonation of reagent takes place by the nitrogen atom of the triazole ring.

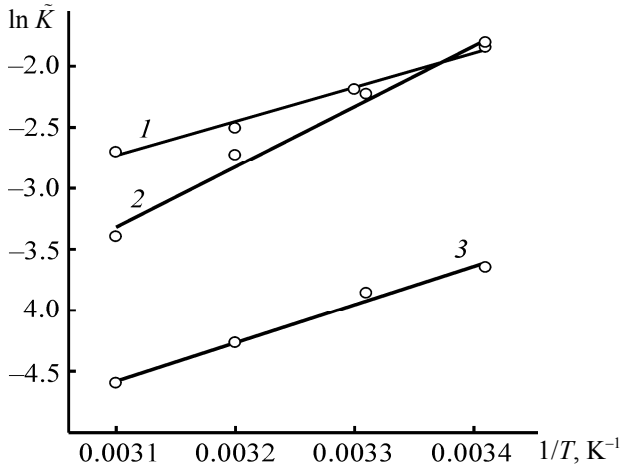
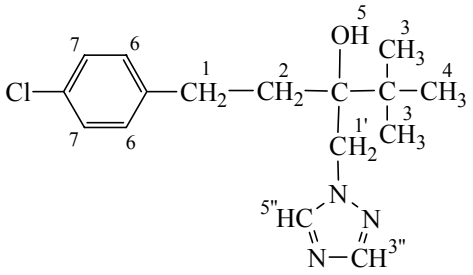


Fig. 3. Dependence of concentrational extraction constant on temperature. Starting concentrations of acid and extractant: (*1*) 1 M HNO_3, 0.02 M of propiconazole (toluene), r^2 0.988; (*2*) 2.09 M HCl, 0.03 M of tebuconazole (toluene with 15 vol % of *n*-decanol), r^2 0.985. (*3*) 2.49 M HCl, 0.02 M of propiconazole (toluene with 15 vol% of *n*-decanol), r^2 0.989.

Table 1. Effective extraction constants of acids

[$\mathbf{L}_0$], M	Extraction of HNO_3 with propiconazole (toluene)			Extraction of HCl with propiconazole (toluene with 15 vol % of *n*-decanol)			Extraction of HCl with tebuconazole (toluene with 15 vol % of *n*-decanol)		
	[HA_{aq}], M	[HA_{org}], M	$\bar{K}$	[HA_{aq}], M	[HA_{org}], M	$\bar{K}$	[HA_{aq}], M	[HA_{org}], M	$\bar{K}$
0.02	0.48	0.0009	0.39	1.00	0.0005	0.038	2.08	0.0069	0.11
	0.99	0.0030	0.33	1.50	0.0011	0.033	2.59	0.0114	0.13
	1.49	0.0060	0.31				3.08	0.0152	0.16
	1.97	0.0097	0.34						
0.03	0.99	0.0044	0.32	1.00	0.0008	0.040	2.08	0.0105	0.11
	1.49	0.0100	0.36	1.50	0.0013	0.026	2.58	0.0174	0.13
	1.96	0.0145	0.34				3.07	0.0224	0.15
0.04	0.48	0.0018	0.39	1.00	0.0010	0.038	1.53	0.0072	0.11
	0.98	0.0060	0.33	1.50	0.0018	0.027	2.07	0.0156	0.13
	1.50	0.0140	0.38	1.98	0.0030	0.019	2.57	0.0234	0.14
0.05	0.48	0.0020	0.34	1.50	0.0023	0.027	1.53	0.0101	0.12
	1.48	0.0170	0.37	2.48	0.0070	0.018	2.07	0.0220	0.16
	1.96	0.0230	0.32						
			0.35±0.02			0.028±0.006			0.13±0.01

Table 2. Characteristic of the ^{1}H NMR spectra of tebuconazole and tebuconazole hydrochloride



Compound	Parameter	Chemical shift values δ and alterations in the signals of protons Δδ, ppm									
		atom	1	2	3, 4	5	6	7	1'	3"	5"
L	δ	H_a H_b	2.44 m 1.67 m	1.77 m	1.00 s	3.28 s	6.93	7.15	4.32 s	7.95 s	8.19 s
L$_2$·HCl	δ	H_a H_b	2.50 1.70	1.60	1.06	3.00–4.00 br.s	6.99	7.19	4.49	8.20	9.24
	Δδ	H_a H_b	0.06 0.03	–0.17	0.06	0.22	0.06	0.04	0.17	0.25	1.05

The superstoichiometric extraction of acids from the solutions with high acidity (Fig. 1) may be caused by the addition of the acid molecule to monosolvate due to the dipole-dipole interaction [14]. The addition of the second acid molecule can also take place at the ether (for propiconazole) or the alcohol group of reagent (for tebuconazole) because the extraction of acids with the aliphatic alcohols and ethers increases with the acidity of water phase [20, 22].

It is established that the concentrational extraction constants of acids decrease with the increase in temperature in the range 20–50°C (Fig. 3). Hence, the process of extraction is exothermic. From the slope of the log $\tilde{K} = f(1/T)$ in keeping with the integral form of the van't-Hoff equation (4):

$$\ln \tilde{K} = -\Delta H/RT + \text{const}, \quad (4)$$

where ΔH value was calculated. It was equal to –23.8 kJ mol^{-1} for the extraction of HNO_3 with propiconazole, and –26.1 and –41.4 kJ mol^{-1} for the extraction of HCl with propiconazole and tebuconazole respectively. For establishing proton-accepting center of tebuconazole the IR and ^{1}H NMR spectra of its hydrochloride were analyzed. The high-frequency shift by 23 cm^{-1} of the absorption band of the stretching-bending vibrations of the triazole ring ν_{ring} (present in the IR spectrum of tebuconazole at 1500 cm^{-1}) in the spectrum of salt and also the presence of broad intense absorption band in the range 2500–2800 cm^{-1} with the maxima at 2687 and 2650 cm^{-1} which can be attributed to $\nu(N–H^+)$ of the protonated amino group of the type $HC=NH^+$ [23,24] suggest that the protonation of reagent at the pyridine-type nitrogen atom of the triazole ring takes place [24].

In Table 2 ^{1}H NMR spectra of tebuconazole and its protonated form are presented. In the spectrum of protonated form of reagent all the signals are broadened because of the exchange processes, but the location of signals alters insignificantly (0.1–0.3 ppm) as compared to the spectra of reagent save the proton signal of the $H^{5"}$. The change in the chemical shift of the $H^{5"}$ proton signal results from the redistribution of electronic density in the delocalized $H^{5"}C=N–H^+$. It is as large as 1.05 ppm and indicates the protonation of N^4 atom of the triazole ring of reagent.

Hence, our studies showed that by the extractability with respect to strong mineral acids the substituted 1*H*-1,2,4-triazoles are close to dialkylsulfoxides, trioctylphosphine oxide, dialkyl phosphonates, and trialkyl phosphates.

EXPERIMENTAL

1-{[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]methyl}-1*H*-1,2,4-trizole and (*RS*)-1-(4-chlorophenyl)-4,4-dimethyl-3-(1*H*-1,2,4-triazol-1-yl-methyl)-

pentan-3-ol with the main substance content not less than 98% were used.

The purity and individuality of compounds were established by elemental analysis, potentiometric titration in the anhydrous acetic acid, and also by the ^{1}H and ^{13}C NMR spectroscopy

Melting points were measured on a Boetius heating block. The specific electroconductivity of 0.001 M solution of tebuconazole hydrochloride in acetone of "extra pure" grade was evaluated on an OK 102.1 conductometer (Hungary) with an OP-907/3 electrode. IR spectra of tebuconazole and its hydrochloride were obtained on a Specord M80 spectrometer in Nujol in the range 4000-400 cm^{-1}. ^{1}H NMR spectra were taken on a Bruker AM-300 spectrometer (300 MHz) in chloroform-d_6.

Propiconazole is a light yellow amorphous substance, mp 58°C. Tebuconazole is a white crystalline substance, mp 103–104°C. Solutions of organic reagents were prepared from accurately weighed samples. Toluene of the "pure for analysis" grade was used as a diluent, and *n*-decanol of "pure for analysis" grade was a modifier.

Water solutions of acids were prepared by dilution of the concentrated "chemically pure" hydrochloric acid and of the "pure for analysis" nitric acid. Concentrations of acids in water solutions were evaluated by titration, and in the extracts, by the two-phase titration [25] in the presence of bromocresol green indicator. The standard 0.1 M KOH solution was used as the titrant. The extraction of acids was carried out in the separating funnels at 20.5±0.5°C and the ratio of organic and water phase 1:1 vol/vol under the intense stirring. It was established preliminary that the time of contact necessary for the achievement of equilibrium is no more than 5 min. The extraction of acids was investigated at the time of contact between phases equal to 10 min. The time of phase separation in the extraction systems was no more than 20 s. For the investigation of the effect of temperature on the extraction of acids the separation funnels with temperature control were used. The temperature was maintained by means of thermostate with the accuracy ±0.2°C. Solutions of extractants with the concentration 0.02–0.05 M were used because at the contact of more concentrated solutions (≥ 0.1 M) with the solutions of acids of the concentration above 3–4 M precipitates of salts of reagents were formed.

Tebuconazole hydrochloride formed as a voluminous precipitate during the contact of 0.65 M tebuconazole solution in chloroform of "pure for analysis" grade with 6 M HCl was filtered off, washed with hexane of the "pure" grade, and dried in air until the constant mass. The elemental analysis data showed that the substance isolated corresponds to the compound $\mathbf{L_2}$·HCl (found, %: C 55.35, H 6.86, N 11.95, Cl 20.62; calculated for $C_{16}H_{23}Cl_2N_3O$, %: C 55.82, H 6.75, N 12.21, Cl 20.59). According to the IR and ^{1}H NMR data it does not contain water, mp 126–128°C. The salt obtained is well soluble in acetone and ethanol, moderately soluble in *N*-decanol, and poorly soluble in toluene.

ACKNOWLEDGMENTS

The work was carried out with the financial support of the OHNM-5 Program of the Department of Chemistry and the Materials sciences of the Russian Academy of Sciences.

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