Preparation and Properties of Magnesium(II) Compounds with Some Bioactive Ligands

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Synthesis, analytical data, IR spectra, as well as antimicrobial activities of seventeen Mg(II) compounds are presented. By means of IR spectral analysis the stereochemistry around Mg(II) atom in the compounds had been studied. Pyridine, nicotinamide, and *N,N*-diethylnicotinamide were coordinated to Mg(II) through the nitrogen atom of their heterocyclic ring. IR data suggest a unidentate coordination of carboxylate ions to Mg(II). The antimicrobial effects have been tested on various strains of bacteria, yeasts, and filamentous fungi. The found antimicrobial effect of the compounds is decreased in the sequence: dermatophytes, phytopathogenic fungi, yeasts, and bacteria. Significant morphological changes of *Botrytis cinerea* were observed by the compounds *VI* and *XVI*. The highest antimicrobial effects were manifested by the compound *V*, especially against dermatophytic fungi *Trichophyton terrestre* and *Microsporum gypseum* (IC_{50} = 623 μg cm^{-3} and 642 μg cm^{-3}, respectively).

It is well known that many metal cations play an active role in a great number of various biological processes [1]. The activity of metallic ions has been examined from various points of view. Generally, toxicity of metals increased with relative atomic mass [2]. In addition to atomic mass, toxicity of metals to various organisms has been shown to be related to the electronegativity of metallic ions and stability of metal chelates [3]. An antimicrobial effect was observed for Mg(II) azidokojate [4]. It is also well known that heterocyclic compounds play a significant role in many biological systems. Especially the six-membered ring system is a component of several vitamins and drugs [5]. From our point of view it was challenging to study the interactions between metal ions and heterocyclic nitrogen compounds that occur in living systems and are used in pharmacy. Thermal properties of the Mg(II) complexes are reported in our previous papers [6—11]. There had been studied the antimicrobial activities of some Cu(II), Ni(II), and Fe(III) compounds [1]. But the reported data on antimicrobial activities and of the IR spectra in the case of Mg(II) compounds are too rare. The present work was aimed to the study of IR spectra, antibacterial and antifungal efficiency of the Mg(II) compounds.

EXPERIMENTAL

The compounds $Mg(ac)_2 \cdot 2H_2O$ (*I*), $Mg(Clac)_2 \cdot 2H_2O$ (*II*), $Mg(Cl_2ac)_2 \cdot H_2O$ (*III*), and $Mg(Cl_3ac)_2 \cdot 3H_2O$ (*IV*) (where ac = CH_3COO^-, Clac = $ClCH_2COO^-$, Cl_2ac = Cl_2CHCOO^-, Cl_3ac = Cl_3CCOO) were prepared by dissolving $Mg(OH)_2$ (1.16 g; 0.02 mol) in 100 cm^3 of the solution of appropriate acetic acid and water (φ_r = 1 2) by gradual stirring. The solutions were reduced in a half volume at room temperature and left to be crystallized. The complexes which formed were filtered off, washed with ether and dried at room temperature.

The compound $Mg(asa)_2(asah)_2$ (*V*), where asa = acetylsalicylate anion and asah = acetylsalicylic acid, was prepared by dissolving asah (9 g; 0.05 mol) in 350 cm^3 of ethanol and gradual adding of $Mg(OH)_2$ (0.725 g; 0.0125 mol). The resulting solution was reduced in a half volume at room temperature and left to be crystallized. The complex which formed, was filtered off, washed with ether and dried at room temperature.

Compounds $Mg(ac)_2(na)_5 \cdot 2H_2O$ (*VI*), $Mg(Clac)_2(na)_6 \cdot 6H_2O$ (*VII*), $Mg(Cl_2ac)_2(na)_6 \cdot 5H_2O$ (*VIII*), and $Mg(Cl_3ac)_2(na)_6 \cdot 2H_2O$ (*IX*) were prepared by dissolving nicotinamide (na) (1.22 g; 0.01

mol) in 100 cm^3 of ethanol and by gradual adding of ethanol solution of appropriate acetato or halogenoacetato Mg(II) complexes in the mole ratio 6 1. The solutions were further treated equally as in compounds *I—IV*.

Compound $Mg(Clac)(OH)(Et_2na)_2 \cdot 2H_2O$ (*X*) was prepared by dissolving $Mg(Clac)_2 \cdot 2H_2O$ (1.24 g; 0.005 mol) in 100 cm^3 of ethanol and by gradual adding of *N,N*-diethylnicotinamide (Et_2na) (1.78 g; 0.1 mol). The resulting solutions were further treated as in previous compounds.

Compounds $Mg(Clac)_2(py)_2 \cdot 2H_2O$ (*XI*), $Mg(Cl_2ac)_2py \cdot H_2O$ (*XII*), and $Mg(Cl_3ac)_2py \cdot H_2O$ (*XIII*) were prepared by gradual adding of pyridine (py) (3.16 g; 0.04 mol) to 150 cm^3 of ethanol solution of appropriate halogenoacetato magnesium complexes in the mole ratio 4 1. The solutions were reduced in a half volume at room temperature and left to be crystallized and then equally treated.

Compound $Mg(SCN)_2 \cdot 5H_2O$ (*XIV*) was prepared by dissolving $MgCl_2 \cdot 6H_2O$ (2.03 g; 0.1 mol) in 100 cm^3 of ethanol and by gradual adding of KSCN (1.94 g; 0.2 mol). Separated KCl was filtered off from the solution. The filtrate was reduced in a half volume at room temperature and then treated as in the previous compounds.

Compounds $Mg(SCN)_2py \cdot 5H_2O$ (*XV*), $Mg(SCN)_2(na)_4 \cdot 3H_2O$ (*XVI*), and $Mg(SCN)_2(Et_2na)_2 \cdot H_2O$ (*XVII*) were prepared by dissolving $MgCl_2 \cdot 6H_2O$ (2.03 g; 0.1 mol) in 100 cm^3 of ethanol and by gradual adding of KSCN (1.94 g; 0.2 mol). Separated KCl was filtered off from the solution and then py (3.16 g; 0.4 mol), na (4.88 g; 0.4 mol) or Et_2na (7.12 g; 0.4 mol) was added, respectively, to filtrate. The resulting solutions were reduced in a half volume at room temperature and left to be crystallized. The isolation of the products was analogous to the previous cases.

The IR spectra were obtained on Philips analytical PU9800 FTIR spectrometer by using Nujol mulls in the range $\tilde{\nu} = 200$—$4000\ cm^{-1}$.

The antimicrobial activity of the magnesium complexes under investigation was evaluated by using G^+ bacterial strain *Bacillus subtilis* CCM 1718 and G^- bacteria *Escherichia coli* CCM 5172 and the yeast *Candida albicans* CCY 29391; the filamentous fungi *Rhizopus oryzae*, *Aspergillus flavus*, *Botrytis cinerea*, *Alternaria alternata*, and *Fusarium nivale* (obtained from the Collection of Microorganisms of the Department of Biochemistry and Microbiology, Faculty of Chemical Technology, Slovak University of Technology) and dermatophyte strains *Microsporum gypseum* and *Trichophyton terrestre* (both isolated from patients).

To test the antimicrobial activity on bacteria and yeasts, 100 cm^3 of appropriate liquid medium (bacteria – Müller—Hinton, yeasts – Sabouraud-glucose) was inoculated with 1 cm^3 of growing overnight culture and distributed in 5 cm^3 aliquots into L-shaped tubes (adapted for direct measurements of absorbance) with 0.05 cm^3 of solution of the tested compounds in dimethyl sulfoxide (DMSO). The cultures of bacteria and yeasts were then incubated under vigorous shaking at 30 °C. Absorbances of duplicate sets of tubes were measured at $\lambda = 650$ nm at intervals.

The effects on filamentous fungi were tested during static culturing. Therefore 0.06 cm^3 of the tested compound in DMSO was added to Petri dishes (diameter 60 mm) immediately before pouring 6 cm^3 of malt extract agar (filamentous fungi) or Sabouraud-glucose agar (dermatophytes) to obtain desired concentrations of inhibitors. The solidified plates were then inoculated in the centre with 0.005 cm^3 of the spore suspension (spore density $10^5\ cm^{-3}$) of the filamentous fungi from 21 days old strains in 0.1 vol. % aqueous Tween 80. Duplicate sets of agar plates were incubated at 25 °C and the diameters of growing colonies were measured at intervals (96 h, 144 h, 196 h, 360 h, and 384 h in the case of *M. gypseum* and *T. terrestre*, 72 h, 96 h, 120 h, 144 h, and 168 h in the case of *A. flavus*, *B. cinerea*, *A. alternata*, and *F. nivale*, and 24 h and 48 h in the case of *R. oryzae*).

Chromatographically pure compounds were dissolved in DMSO. Its final concentration never exceeded 1 vol. % either in control or treated samples. This concentration of DMSO did not affect the growth of tested microorganisms. The compounds under investigation were tested at concentrations ranging from 100 to 1000 $\mu g\ cm^{-3}$. The antimicrobial effect was characterized by IC_{50} values (concentration of a compound which in comparison to the control inhibits microbial growth by 50 %) and MIC values (minimal inhibitory concentration of a compound which inhibits microbial growth by 100 %). The IC_{50} and MIC values could be read from the toxicity curves.

MIC experiments on subculture dishes were used to assess the minimal microbicidal concentration (MMC) values. The subcultures were prepared separately in the Petri dishes containing competent agar medium for dermatophyte strains and incubated at 25 °C for 96 h. The MMC value was taken as the lowest concentration which showed no visible growth of microbial colonies in the subculture dishes.

The optical microscopic measurements: In addition to measurements of diameter of colonies, morphology of hyphae was checked microscopically by optical microscope (Zeiss, Jena). Microphotography was performed *in situ* after staining with 0.5 vol. % methyl blue in lactophenol.

The content of carbon, hydrogen, and nitrogen was determined by elemental analysis equipment and the content of magnesium was established by complexiometric titration.

Table 1. Analytical Data of the Complexes *I*—*XVII*

Compound	w_i(calc.)/%				w_i(found)/%			
	C		H		N		Mg	
I	26.96	26.92	5.61	5.58			13.66	13.61
II	19.41	19.17	3.23	3.10			9.83	9.84
III	16.10	16.53	1.34	1.63			8.15	8.14
IV	11.91	11.99	1.48	1.67			6.03	6.04
V	58.22	58.95	4.04	4.08			3.27	3.27
VI	51.77	51.93	5.08	5.13	17.77	17.72	3.05	3.03
VII	45.67	45.74	4.95	4.95	15.98	15.94	2.31	2.34
VIII	43.58	43.40	4.36	4.46	15.25	15.18	2.21	2.23
IX	42.96	42.95	3.58	3.66	15.04	15.05	2.20	2.26
X	50.09	50.04	6.64	6.64	10.61	10.54	4.61	4.63
XI	41.45	41.55	4.44	4.47	6.91	6.85	5.93	5.99
XII	28.63	28.16	2.38	2.37	3.71	3.74	6.44	6.45
XIII	20.13	19.96	3.17	3.15	2.61	2.52	4.53	4.52
XIV	10.43	10.33	4.34	4.38	12.17	12.15	10.43	10.92
XV	27.15	26.62	4.92	4.04	13.58	13.62	7.86	7.87
XVI	45.75	46.19	4.40	4.44	20.53	19.83	3.52	3.48
XVII	51.36	51.43	5.84	5.86	16.34	16.20	4.67	4.75

Table 2. Infrared Spectral Data of Complexes *I*—*V*, $\tilde{\nu}$/cm^{-1}

Assignment	*I*	*II*	*III*	*IV*	*V*
$\nu_{as}(COO^-)$	1649	1644	1678	1674	1754, 1604
$\nu_s(COO^-)$	1448	1408	1414	1464	1460, 1419
Δ_{COO^-}	201	236	264	210	294, 185
ν(C—C)	918	949	924	949	916
ν(CH)	2849	2924	2924	2921	2926
ν(OH)	3351	3374	3414	3451	3489
δ(HOH)	1595	1578	1643	1613	1605
$\rho(H_2O)$	625, 658	696, 779	677, 781	687, 745	666, 704, 756
	689, 947	945, 901	822	843	787, 804, 970
ν(Mg—O)	324, 327	309, 405	303, 334	293, 395	326, 370
$\pi(CO_2)$	542	538	541	540	539

as = antisymmetric, s = symmetric.

RESULTS AND DISCUSSION

The analytical data of the compounds *I*—*XVII*, reported in Table 1, show a good agreement between the experimental and calculated data.

The most important infrared spectral data are reported in Tables 2—4. The absorption bands which occur in the range $\tilde{\nu}$ = 3125—3651 cm^{-1} (ν_s(OH) and ν_{as}(OH)) and 1578—1637 cm^{-1} (δ(HOH bending)) show the presence of water of crystallization [12] and the absorption bands which occur in the range 650—1000 cm^{-1} (rocking and wagging stretching) confirm the presence of water as coordinated in the complexes [13]. The presence of water as water of crystallization and as coordinated water in the compounds is further confirmed by the thermal decomposition data [7—11]. The carboxylate ions can coordinate to metal ions in a number of ways such as unidentate, bidentate (chelating) or bridging [13] and there is an evidence of that fact in the IR spectrum. The analysis of COO^- group bands frequencies allowed the determination of the parameter $\Delta_{COO} = \tilde{\nu}(\nu_{as}(COO^-)) - \tilde{\nu}(\nu_s(COO^-))$. The magnitude of Δ_{COO} has been used by *Nakamoto* [13] as a criterion of the way of carboxylate binding to metal ions. The calculated ones from the examined spectra values of Δ_{COO} are in the range 201—294 cm^{-1}. These values and the three bands (COO^- deformation) at 720—920 cm^{-1} and a strong band $\pi(CO)_2$ near to 540 cm^{-1} [13] in the case of complexes *I*—*XVII* are in good agreement with the literature data for unidentately bonded acetates structures.

The absorption bands which occur in the range 206—250 cm^{-1} (ν(Mg—N)) confirm the coordination of pyridine and its derivatives through their nitrogen atom of the heterocyclic ring to the Mg ion [14]. The absorption bands which occur in the range 605—628 cm^{-1} and 407—426 cm^{-1} are due to the ring deformation (in-plane and out-of-plane) of the pyridine ring. These bands shifted to higher value upon complex formation. The absorption bands which occur in the range 293—410 cm^{-1} (ν(Mg—O)) confirm the coordination of oxygen to the Mg ion.

Table 3. Infrared Spectral Data of Complexes *VI*—*XIII*, $\tilde{\nu}/\mathrm{cm}^{-1}$

Assignment	*VI*	*VII*	*VIII*	*IX*	*X*	*XI*	*XII*	*XIII*
ν(C—H)$_{ac}$	2849	2923	2913	2847	2851	2924	2845	2847
ν(CO)	1680	1673	1679	1686				
ν(ring)	1593	1593	1593	1593	1590	1580	1574	1576
	1574		1559	1574				
ν(C—H)$_{ac}$	918	936	936	934	918	926	933	926
ν(C—H)$_{py}$	830	833	830	841	824	848	828	839
γ(CCC)	646	640	639	655	652			
	625	613	617	625				
δ(py)	605	613	613	625	625	602	605	609
	412	407	420	412	410	407	426	426
ν(Mg—N)	206	216	206	210	204	218	250	214
	239		235	220	212	256		219
$\nu_{as}(COO^-)$	1680	1773	1669	1638	1717	1696	1695	1720
$\nu_s(COO^-)$	1423	1500	1464	1421	1462	1462	1446	1446
Δ_{COO^-}	257	273	205	217	225	234	249	274
ν(OH)	3620	3651		3372	3231	3125	3274	3360
δ(HOH)			1633	1637		1609	1609	1604
$\rho(H_2O)$	664	669	639	675	824	700	629	679
	702	709	970	698	877	925	677	693
	935	833	675	933	947	935	702	700
	968	836		964		952		925
ν(Mg—O)	392	407	304	345	410	359, 385	378	376
$\pi(CO_2)$	542	539	540	537	543	541	536	544

Table 4. Infrared Spectral Data of na and Complexes *XIV*—*XVII*, $\tilde{\nu}/\mathrm{cm}^{-1}$

Assignment	na	*XIV*	*XV*	*XVI*	*XVII*
$\nu_{as}(NH_2)$ or (NR_2)	3357			3360	3370
$\nu_s(NH_2)$ or (NR_2)	3150			3163	3173
ν(CO)	1678			1674	
$\delta(NH_2)$ or (NR_2)	1617			1617	1618
ν(ring)	1593		1593	1593	1597
	1576		1572	1574	1579
δ(py)			628, 424	605,412	407
ν(C—H)$_{ring}$			846	830	820
γ(CCC)	644		654	644	659
	621		628	625	638
ν(Mg—N)			247	210, 216, 237	204, 214
ν(CN)		2125, 2080	2114, 2070	2089, 2043	2087, 2035
ν(CS)		704, 764, 804	700	830, 704	820, 801, 708
δ(NCS)		437	424	480, 413	480, 407
ν(OH)		3303, 3239	3372, 3220	3478, 3303	3568
				3254	
δ(HOH)		1635	1603, 1620	1605, 1617	1618
$\rho(H_2O)$		617, 891, 939	675, 758, 785	644, 748, 773	659, 756, 779
			847, 882, 949	891, 941	880, 947, 980
ν(Mg—O)		335	336	393	316, 347, 381

as = antisymmetric, s = symmetric and R = CH_3CH_2.

The SCN group may coordinate to a metal through the nitrogen or the sulfur atom or both (M—NCS—M′). Several empirical criteria have been developed to determine the bonding type of the NCS group in metal complexes [13]. The absorption bands which occur in the range 2070—2125 cm^{-1}(ν(CN)) and near to 420 cm^{-1} (δ(NCS)) confirm the coordination of SCN group to the Mg(II) metal through the sulfur atom in complexes *XIV* and *XV*. The absorption bands which occur in the range 2035—2089 cm^{-1}(ν(CN)), 704—830 cm^{-1} (ν(CS) stretchings), and near to 480 cm^{-1}(for N-bonded) and 420 cm^{-1}(for S-bonded) confirm the coordination of the SCN group to Mg(II) through sulfur and nitrogen in complexes *XVI* and *XVII*.

All tested compounds were inactive against bacteria. There are five compounds, *i.e.* *V*, *VI*, *VIII*, *IX*, and *XVI* which showed some effects against yeasts

Table 5. Antimicrobial Activity of Mg(II) Compounds Characterized by the Numerical Values of $IC_{50}/(\mu g\ cm^{-3})$

Compound	1	2	3	4	5	6	7
V	1000	>1000	>1000	900[b]	642[a]	623[a]	>1000
VI	>1000	500[b]	730[b]	>1000	1000	300[b]	1000
VIII	>1000	>1000	>1000	>1000	1000	630[b]	>1000
IX	>1000	>1000	>1000	>1000	1000	540[b]	>1000
XVI	>1000	1000	>1000	>1000	1000	875[b]	>1000

1 – *R. oryzae*, 2 – *B. cinerea*, 3 – *F. nivale*, 4 – *A. alternata*, 5 – *M. gypseum*, 6 – *T. terrestre*, 7 – *C. albicans*. *a*) MIC, MMC = 700 $\mu g\ cm^{-3}$; *b*) MIC, MMC > 1000 $\mu g\ cm^{-3}$.

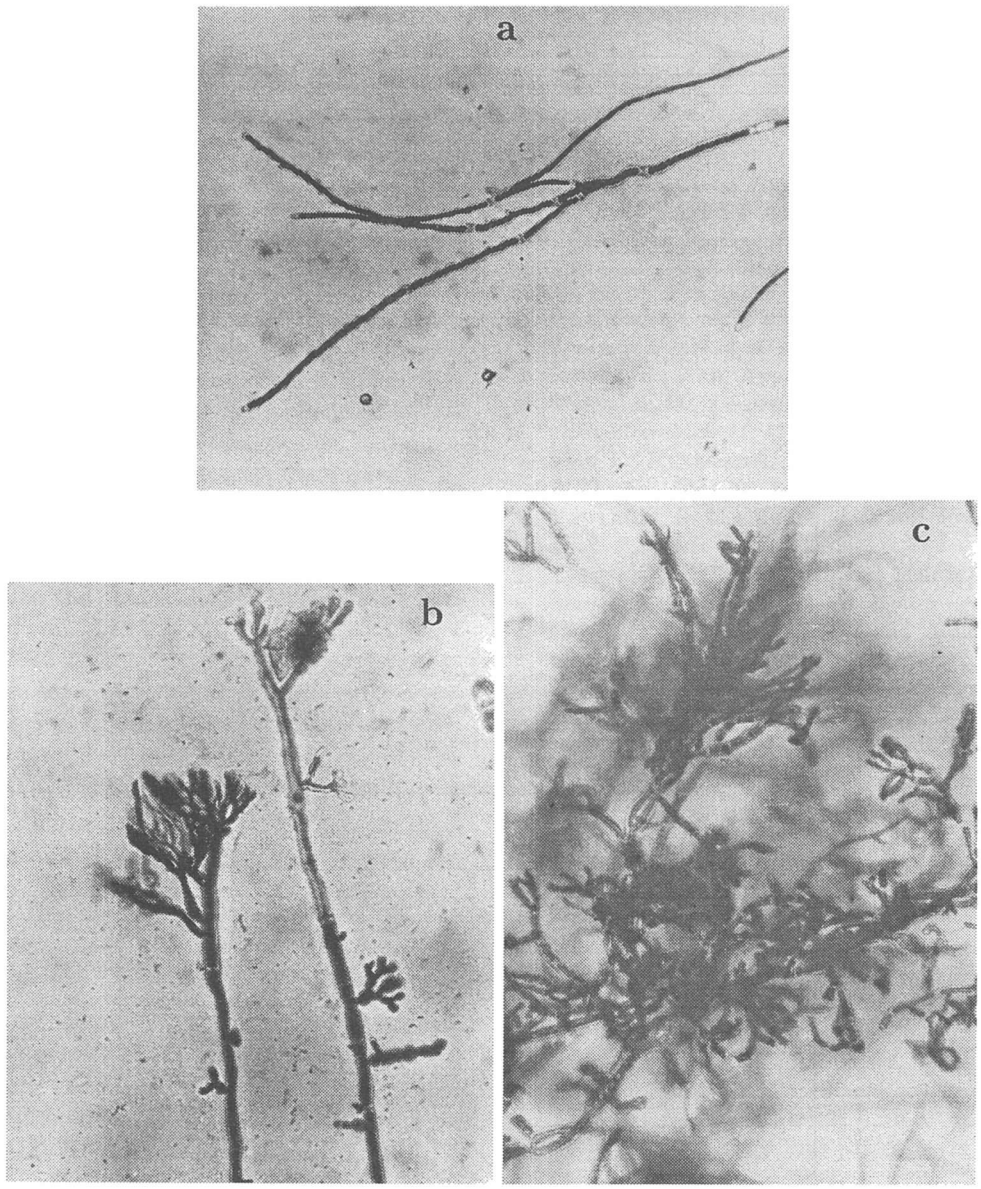

Fig. 1. Changes of morphology of *B. cinerea* hyphae induced by compounds *VI* and *XVI*. *B. cinerea* was cultivated for 4 days in malt agar containing: a) w(DMSO) = 1 %, b) ρ (*VI*) = 1000 $\mu g\ cm^{-3}$ or c) ρ (*XVI*) = 1000 $\mu g\ cm^{-3}$. Magnification about 350 ×.

C. albicans, phytopathogenic filamentous fungi *B. cinerea*, *A. alternata*, and *F. nivale*, and dermatophytic fungi *M. gypseum* and *T. terrestre*. The IC_{50} and MIC values of these compounds are summarized in Table 5. The other tested compounds were inactive against tested microorganisms (IC_{50} > 1000 μg

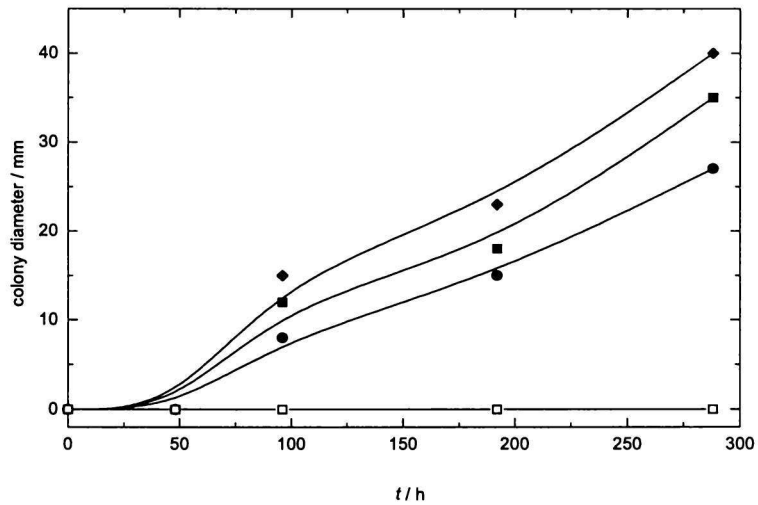

Fig. 2. Colony growth of *Trichophyton terrestre* induced by compound *V*. Final concentration numerical values of compound $\rho/(\mu g\ cm^{-3})$: □ 700, ● 600, ■ 500, ♦ control.

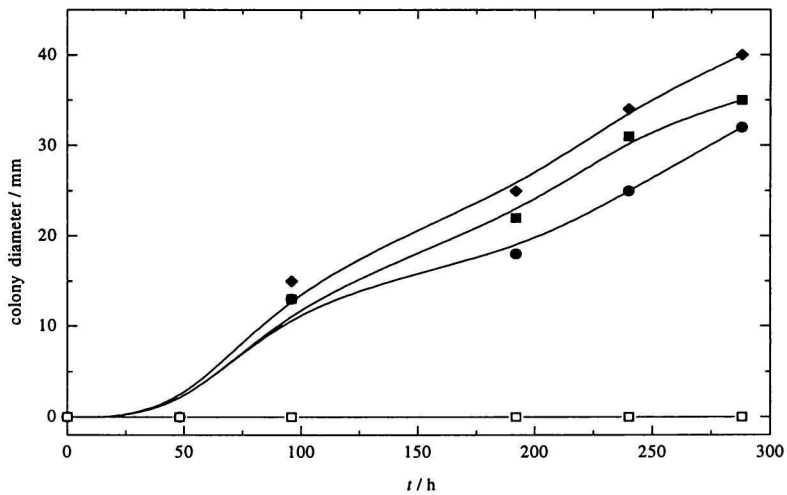

Fig. 3. Colony growth of *Microsporum gypseum* induced by compound *V*. Final concentration numerical values of compound $\rho/(\mu g\ cm^{-3})$: □ 700, ● 600, ■ 500, ♦ control.

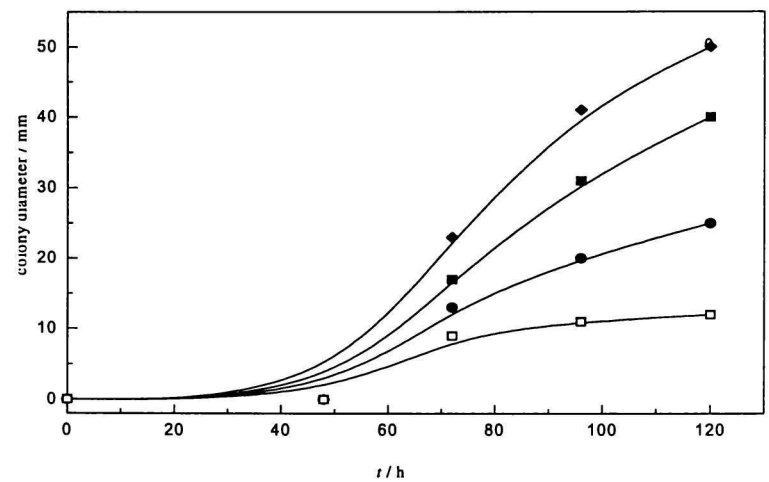

Fig. 4. Colony growth of *Botrytis cinerea* induced by compound *VI*. Final concentration numerical values of compound $\rho/(\mu g\ cm^{-3})$: □ 1000, ● 500, ■ 100, ♦ control.

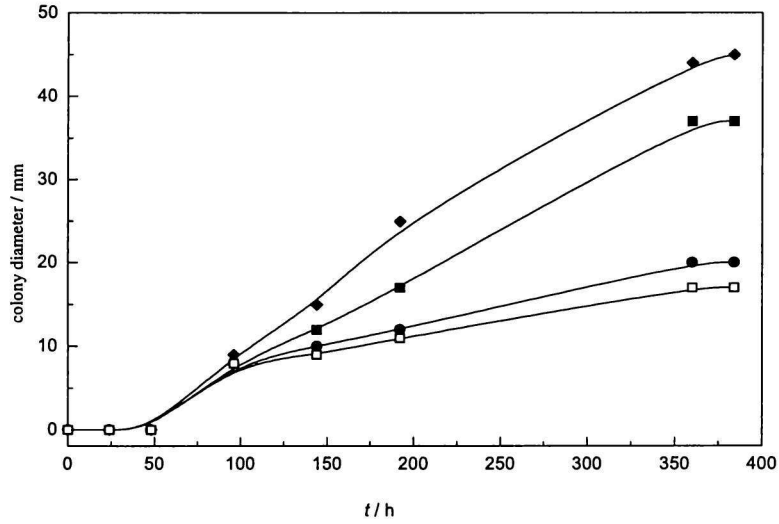

Fig. 5. Colony growth of *Trichophyton terrestre* induced by compound *VI*. Final concentration numerical values of compound $\rho/(\mu g\ cm^{-3})$: □ 1000, ● 500, ■ 100, ♦ control.

cm^{-3}). The compounds *VI* and *XVI* induced morphological changes in growing hyphae of *B. cinerea* (Fig. 1) and *F. nivale* at concentration which partially inhibited the growth. The highest antimicrobial activity was manifested by the compounds *V* and *VI*. The most sensitive fungi to the compound *V* were *T. terrestre* (IC_{50} = 623 $\mu g\ cm^{-3}$) and *M. gypseum* (IC_{50} = 642 $\mu g\ cm^{-3}$). The values of MIC and MMC were 700 $\mu g\ cm^{-3}$ for both dermatophytic fungi. The most sensitive fungi to the compound *VI* were *T. terrestre* (IC_{50} = 300 $\mu g\ cm^{-3}$) and *B. cinerea* (IC_{50} = 500 $\mu g\ cm^{-3}$). Figs. 2—5 illustrate the growth inhibition of *T. terrestre*, *M. gypseum*, and *B. cinerea* by the compounds *V* and *VI*. Antimicrobial effect of the compounds is decreased in the sequence: dermatophytes, phytopathogenic fungi, yeasts, and bacteria.

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