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An Unusual Hexafluorosilicato-bridged Chain Compound; Crystal Structure of *catena*-(μ-Hexafluorosilicato)-tetrakis(5-phenylpyrazole)copper(II)

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Abstract

The reaction of a methanolic solution of M(II)-$(BF_4)_2$ salts and 3(5)-phenylpyrazool (pppzH) in glass vessels yields chain compounds of general formula M(II)$(pppzH)_4(SiF_6)$ (M = Co, Ni, Cu), in which the SiF_6^{2-} group is the bridging ligand. The compounds can also be prepared starting from the hydrated hexafluorosilicates. The structure of one of these compounds, *i.e.* *catena*-(μ-hexafluorosilicato)-tetrakis(5-phenylpyrazole)copper(II) $Cu(C_9H_8N_2)_4(SiF_6)$ (I) has been determined from X-ray diffraction data. The crystals are triclinic, space group $P\bar{1}$ lattice constants $a = 5.531(7)$, $b = 12.211(5)$, $c = 12.140(7)$ Å, $\alpha = 121.69(5)$, $\beta = 99.30(4)$, $\gamma = 101.73(3)°$, $Z = 2$. The Cu(II) ions are at special positions 0,0,0 and $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ and are linked by bridging SiF_6^{2-} groups. The copper ions are coordinated by four pyrazole nitrogen atoms and the fluorine atoms of the SiF_6^{2-} group as axial ligands. $Cu-N_{pyr}$ distances are 2.002(5) and 1.992(5) Å for Cu(1) and 2.009(5) and 1.994(5) Å for Cu(2). Cu–F distances are 2.251(3) Å for Cu(1) and 2.245(3) Å for Cu(2). Magnetic susceptibility measurements of **I** and the analog compounds $Ni(C_9H_8N_2)_4(SiF_6)$ (**II**) and $Co(C_9H_8N_2)_4(SiF_6)$ (**III**) show no significant magnetic interaction. The magnetic and spectroscopic properties of the compounds are discussed in relation to their structure.

Introduction

One-dimensional polynuclear compounds of transition metals receive much attention in contemporary literature [1]. The first crystal structure of a chain compound containing coordinated SiF_6^{2-} was reported for *N*-vinylimidazole, abbreviated viz [2]. $Co(viz)_4(SiF_6)$ consists of linear chains of $Co(viz)_4^{2+}$ and SiF_6^{2-} ions, linked by Co–F–Si bonds.

A new type of chain compounds with SiF_6^{2-} as a bridging group is described here. From the reaction in methanol of M(II)$(BF_4)_2 \cdot 6H_2O$ and 3(5)-phenylpyrazool (pppzH) in pyrex glass vessels compounds are formed of general formula M(II)$(pppzH)_4(SiF_6)$ (M = Co, Ni, Cu). The slow release of SiF_6^{2-} into the solution allows these compounds to separate as long needle-shaped crystals. Spectroscopic and magnetic measurements indicated that the structure of these compounds differed considerably from the structure found for $Co(viz)_4(SiF_6)$ [2]. Crystals of the copper compound were analyzed by X-ray diffraction to obtain detailed structural information for this compound.

This paper describes the crystal structure of the copper compound, as well as the synthesis and spectroscopic properties of this and the related Ni(II) and Co(II) compounds. Magnetic susceptibility measurements (2–300 K) were performed to investigate the magnetic exchange in these compounds.

Experimental

Starting Materials

The ligand pppzH was synthesized as described by Elguero and Jacquier [3]. M(II)$(BF_4)_2$ salts were used as commercially available. Satisfactory C, H, N, F and metal analyses were obtained for all compounds studied.

Synthesis and Crystal Growth

$Cu(C_9H_8N_2)_4(SiF_6)$ (I)

A solution of 4 mmol (0.58 g) of pppzH and 1 mmol (0.35 g) $Cu(BF_4)_2 \cdot 6H_2O$ in 50 ml dry methanol was kept at ~60 °C in a pyrex glass vessel. After one to two weeks light-blue needles were formed in the solution. $Ni(C_9H_8N_2)_4(SiF_6)$ (**II**) and $Co(C_9H_8N_2)_4(SiF_6)$ (**III**) were obtained from their tetrafluoroborate salts in the same way.

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Synthesis from $M(II)(SiF_6)\cdot 6H_2O$

A warm solution of 1 mmol $M(II)(SiF_6)\cdot H_2O$ in 25 ml dry methanol is added slowly to a warm solution of 4 mmol (0.58 g) of pppzH in 30 ml dry methanol. The compounds precipitate instantaneously as powders.

Physical Measurements and Analyses

Infrared spectra were recorded on a Perkin-Elmer Model 580B IR spectrophotometer in the region 4000–180 cm^{-1} as KBr pellets. Ligand field spectra were recorded in the region 28 000–4000 cm^{-1} on a Perkin-Elmer 330 UV–Vis spectrophotometer, using the diffuse-reflectance technique with MgO as a reference. Magnetic susceptibility data were obtained on powdered samples in the temperature region 2–300 K from a Faraday balance [4]. The data were corrected for diamagnetism using Pascals constants.

X-ray Data Collection

A single crystal of **I** was sealed in a capillary and mounted on an Enraf-Nonius CAD-4 four circle diffractometer. Mo Kα radiation ($\lambda = 0.71073$ Å), monochromated by graphite, was used to determine the unit-cell parameters and the space group, as well as to measure the reflection intensities. Diffraction and crystal data are summarized in Table 1.

TABLE 1. Crystal and Diffraction Data for $Cu(pzzH)_4(SiF_6)$ (I)

Crystal system	triclinic
Space group	$P\bar{1}$
a (Å)	15.531(7)
b (Å)	12.211(5)
c (Å)	12.140(7)
α (°)	121.69(5)
β (°)	99.30(4)
γ (°)	101.73(3)
Z	2
D (calc.) (g cm^{-3})	1.43
D (obs.) (g cm^{-3})	1.41
θ range (°)	2–21
Range of indices	
h	−15 to 15
k	−12 to 12
l	0 to 12
F(000)	802.85
No. independent reflections	4407
No. significant reflections ($I > 2\sigma(I)$)	2568
Final R	3.22
Final R_w	3.82

Structure Determination and Refinement of $Cu(C_9H_8N_2)_4SiF_6$

The data were corrected for Lorentz and polarization effects [5]. All calculations were carried out on the Leiden University computer (IBM 3083) using a set of computer programs written or modified by E. W. Rutten-Keulenmans and R. A. G. de Graaff. The function minimized during least-squares refinement was $\Sigma w(|F_o| - |F_c|)^2$ with $w = 1/\sigma^2 F$. Reliability indices referred to in Table 1 are $R = \Sigma(|F_o| - |F_c|)/F_o$ and $R_w = [\Sigma w(|F_o| - |F_c|)^2/\Sigma w|F_o|^2]^{1/2}$. The positions of the copper and silicon atoms and of the coordinating fluorine atoms were determined from an E^2_{obs} Patterson synthesis. The positions of the other non-hydrogen atoms (except one) were located by using the program AUTOFOUR [6]. After several iterations of refinement a difference Fourier yielded the positions of the last carbon atom. The position of all hydrogen atoms could be determined from difference Fourier maps. The refinement of the hydrogen atoms and the ligand ring was unsatisfactory, this is probably due to thermal movements of the phenyl groups. The positions of the hydrogen atoms were calculated and they were kept at a fixed distance from their parent atom during further refinement. This resulted in a more satisfactory refinement of the structure. The hydrogen atoms were refined with one overall isotropic temperature parameter. Disorder was found in the positions of non-coordinating fluorine atoms (F(3), F(4), F(5) and F(6)) of the hexafluorosilicato group. Four additional peaks were found, positioned between the fluorine atoms at angles close to 45° (labelled F(33), F(44), F(55) and F(66)). The occupation ratio was refined as one parameter for all eight fluorine atoms. The occupation ratios were found to have relative values close to 2:1.

Further refinement gave the R values stated in Table 1. The positional parameters and the isotropic temperature factors of all non-hydrogen atoms are listed in Table 2. The corresponding labelling of the atoms is given in the ORTEP [7] drawing in Fig. 1. See also 'Supplementary Material'.

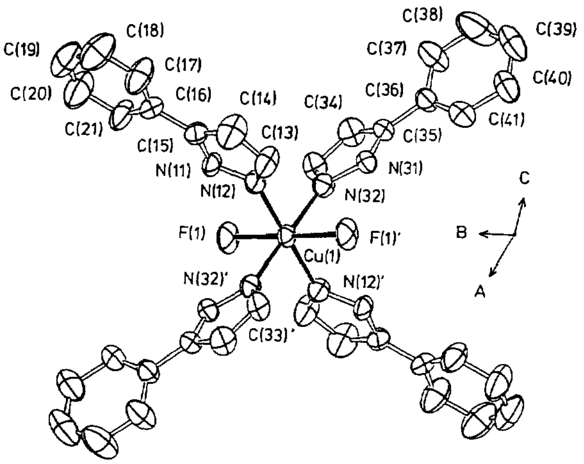

Fig. 1. ORTEP drawing and atomic labelling system for $Cu(pzzH)_4(SiF_6)$ (I), Cu(1).

TABLE 2. Positional Parameters $\times 10^4$ ($\times 10^3$ for F(3)–F(66)) and Isotropic B Values $(8/3 \times \pi^2 \bar{U}) \times 10$ for the Non-hydrogen Atoms of Cu(pppzH)$_4$(SiF$_6$) (I)^a

Atom	*x/a*	*y/b*	*z/c*	B_{iso}
Cu(1)*	0	0	0	27.4(4)
Cu(2)*	5000	5000	5000	29.1(4)
Si	2522(1)	2291(2)	2607(2)	32.0(6)
F(1)	1468(2)	1233(3)	1436(4)	45.1(12)
F(2)	3572(2)	3425(3)	3732(4)	49.3(13)
F(3)	288(1)	237(2)	140(1)	62(5)
F(4)	287(1)	98(1)	218(2)	80(7)
F(5)	219(1)	225(2)	378(1)	80(7)
F(6)	221(1)	364(1)	298(2)	61(6)
F(33)	242(3)	325(3)	198(4)	78(13)
F(44)	303(1)	151(4)	153(3)	76(13)
F(55)	256(2)	138(3)	316(4)	65(13)
F(66)	197(1)	312(3)	366(2)	69(11)
N(11)	179(3)	2878(5)	2153(5)	32(2)
N(12)	−392(3)	1613(5)	1086(5)	33(2)
C(13)	−1202(5)	1747(1)	812(7)	49(3)
C(14)	−1145(5)	3097(8)	1706(8)	61(3)
C(15)	−251(4)	3812(6)	2573(6)	36(2)
N(31)	−1183(3)	−1472(5)	878(5)	32(2)
N(32)	−313(3)	−680(5)	1111(5)	32(2)
C(33)	214(5)	−516(6)	2189(7)	42(2)
C(34)	−290(5)	−1184(7)	2635(7)	44(2)
C(35)	−1188(4)	−1793(6)	1782(7)	33(2)
N(51)	6173(3)	4169(5)	6440(5)	34(2)
N(52)	5304(3)	3993(5)	5789(5)	33(2)
C(53)	4759(5)	2921(7)	5677(7)	43(2)
C(54)	5259(5)	2428(7)	6249(7)	44(3)
C(55)	6173(5)	3253(7)	6738(6)	36(2)
N(71)	4930(3)	2992(5)	2136(5)	34(2)
N(72)	5472(3)	3901(5)	3462(5)	34(2)
C(73)	6315(4)	3925(7)	3427(8)	47(3)
C(74)	6327(5)	3039(7)	2133(8)	53(3)
C(75)	5419(4)	2430(6)	1290(7)	37(2)
C(16)	206(5)	5264(6)	3719(7)	37(2)
C(17)	−296(6)	6121(8)	4079(8)	60(3)
C(18)	98(7)	7479(9)	5140(8)	71(4)
C(19)	1014(7)	7968(8)	5849(8)	68(3)
C(20)	1530(6)	7141(8)	5547(8)	69(3)
C(21)	1126(5)	5779(7)	4463(8)	56(3)
C(36)	−2027(5)	−2621(6)	1759(8)	39(2)
C(37)	−2059(5)	−2525(8)	2933(8)	58(3)
C(38)	−2832(7)	−3312(10)	2931(10)	81(4)
C(39)	−3565(6)	−4184(9)	1795(11)	74(4)
C(40)	−3560(5)	−4307(8)	605(9)	70(3)
C(41)	−2790(5)	−3516(7)	601(8)	51(3)
C(56)	7031(5)	3200(7)	7390(7)	44(3)
C(57)	6995(7)	2109(9)	7456(10)	98(4)
C(58)	7766(9)	2038(12)	8084(13)	125(6)
C(59)	8608(8)	3030(12)	8608(11)	99(5)
C(60)	8661(6)	4104(10)	8535(9)	80(4)
C(61)	7883(5)	4199(9)	7937(8)	60(3)
C(76)	5006(5)	1455(7)	−188(7)	40(3)
C(77)	5477(6)	670(8)	−939(8)	51(3)
C(78)	5098(7)	−227(9)	−2331(10)	64(4)
C(79)	4242(7)	−367(8)	−2997(8)	71(3)
C(80)	3776(6)	423(9)	−2254(8)	73(3)
C(81)	4153(6)	1329(8)	−843(8)	63(3)

^{a}e.s.d.s in the last significant digits are given in parentheses. Special positions are marked with an asterisk.

Results and Discussion

Description of the Structure of Cu(pppzH)$_4$(SiF$_6$) (I)

The structure and atomic labelling of **I** is depicted in Figs. 1 and 2; relevant interatomic distances and angles are given in Tables 3 and 4. The unit cell of **I** is shown in Fig. 3. The copper ions are located at

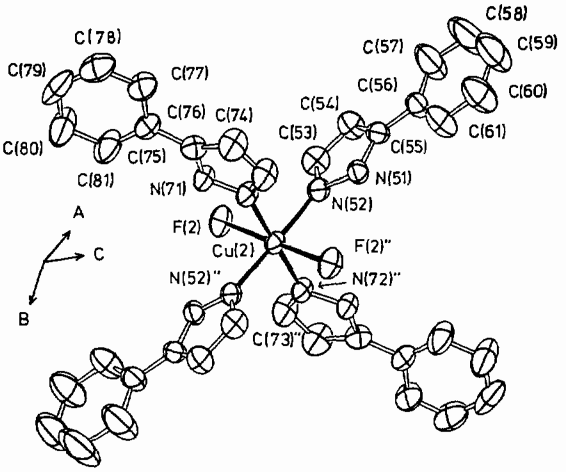

Fig. 2. ORTEP drawing and atomic labelling system for Cu(pppzH)$_4$(SiF$_6$) (I), Cu(2).

TABLE 3. Relevant Interatomic Distances (Å) in Cu(pppzH)$_4$(SiF$_6$) (I)^a

Cu(1)–N(12)	2.002(5)	Cu(2)–N(52)	1.994(2)
Cu(1)–N(32)	1.992(5)	Cu(2)–N(72)	2.009(5)
Cu(1)–F(1)	2.251(3)	Cu(2)–F(2)	2.245(3)
Si–F(1)	1.681(3)	Si–F(2)	1.678(3)
N(11)–N(12)	1.349(6)	N(31)–N(32)	1.378(6)
N(11)–C(15)	1.347(7)	N(31)–C(35)	1.347(7)
N(12)–C(13)	1.314(7)	N(32)–C(33)	1.317(7)
C(13)–C(14)	1.390(9)	C(33)–C(34)	1.383(8)
C(14)–C(15)	1.367(8)	C(34)–C(35)	1.368(8)
C(15)–C(16)	1.470(8)	C(35)–C(36)	1.464(8)
N(51)–N(52)	1.361(6)	N(71)–N(72)	1.355(6)
N(51)–C(55)	1.344(7)	N(71)–C(75)	1.351(7)
N(52)–C(53)	1.333(7)	N(72)–C(73)	1.310(7)
C(53)–C(54)	1.379(8)	C(73)–C(74)	1.369(8)
C(54)–C(55)	1.379(8)	C(74)–C(75)	1.377(8)
C(55)–C(56)	1.468(9)	C(75)–C(76)	1.457(8)

^{a}e.s.d.s in the last significant digits are given in parentheses.

TABLE 4. Relevant Interatomic Angles (°) in $Cu(pppzH)_4(SiF_6)$ (I)[a]

F(1)–Cu(1)–N(12)	90.2(2)	F(2)–Cu(2)–N(52)	91.2(2)
F(1)–Cu(1)–N(32)	91.8(2)	F(2)–Cu(2)–N(72)	89.4(2)
N(12)–Cu(1)–N(32)	90.8(2)	N(52)–Cu(1)–N(72)	89.7(2)
F(1)–Si–F(2)	175.5(2)	Cu(1)–F(1)–Si	173.8(2)
Cu(2)–F(2)–Si	171.4(2)		
N(11)–N(12)–C(13)	105.0(5)	N(51)–N(52)–C(53)	104.0(5)
N(12)–C(13)–C(14)	110.4(6)	N(52)–C(53)–C(54)	111.9(6)
C(13)–C(14)–C(15)	106.9(6)	C(53)–C(54)–C(55)	105.6(6)
C(14)–C(15)–N(11)	104.6(5)	C(54)–C(55)–N(51)	106.1(5)
C(15)–N(11)–N(12)	113.0(5)	C(55)–N(51)–N(52)	112.4(5)
N(11)–C(15)–C(16)	124.4(6)	N(51)–C(55)–C(56)	122.6(6)
C(14)–C(15)–C(16)	131.0(6)	C(54)–C(55)–C(56)	131.2(7)
N(31)–N(32)–C(33)	103.9(5)	N(71)–N(72)–C(73)	104.1(5)
N(32)–C(33)–C(34)	111.6(6)	N(72)–C(73)–C(74)	112.3(6)
C(33)–C(34)–C(35)	106.8(6)	C(73)–C(74)–C(75)	106.3(6)
C(34)–C(35)–N(31)	105.3(5)	C(74)–C(75)–N(71)	104.6(6)
C(35)–N(31)–N(32)	112.4(5)	C(75)–N(71)–N(72)	112.7(5)
N(31)–C(35)–C(36)	123.1(6)	N(71)–C(75)–C(76)	123.7(6)
C(34)–C(35)–C(36)	131.6(7)	C(74)–C(75)–C(76)	131.6(7)

[a] e.s.d.s in the last significant digits are given in parentheses.

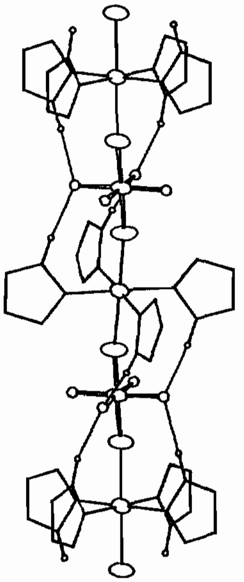

Fig. 3. ORTEP drawing of the unit cell for $Cu(pppzH)_4(SiF_6)$ (I).

inversion centers. Both copper ions are coordinated by two fluorine atoms from the SiF_6 group (F(1) and F(2)) and four pyrazole nitrogen atoms, forming a distorted D_{4h} coordination. The pyrazole nitrogen atoms are in a slightly distorted square, Cu–N distances are 2.002(5) and 1.992(5) Å for Cu(1) and 1.994(2) and 2.009(5) Å for Cu(2), normal values for Cu(II)–N_{pyr} distances [8]. The fluorine atoms are at axial positions at distances of 2.251(3) Å for Cu(1) and 2.245(3) Å for Cu(2), they are relatively long compared to Cu^{2+}–F^- distances (1.90–2.26 Å) found in F^--bridged structures [8], but still rather short for tetragonal Cu(II) compounds. The angle between a line through the coordinating fluorine atoms and the plane of the nitrogen atoms is 88.2(1)° for Cu(1) and 88.7(1)° for Cu(2). The bridging hexafluorosilicato groups are not exactly collinear with the copper chain. A line through the coordinating F(1) and F(2) atoms makes an angle of 3.21(8)° with the line through the copper ions. Hydrogen atoms of all pyrazole nitrogen atoms form hydrogen bridges to non-coordinating fluorine atoms. The disorder in SiF_6^{2-} is probably caused by the tendency of pyrazole to form such hydrogen bridges. The N_{pyr}–F distances are between 2.81–3.01 Å and are comparable to hydrogen bridge contacts found in azole compounds with fluoride or tetrafluoroborate anions [8, 9]. Relevant distances and angles for the hydrogen bridges are given in Table 5.

Spectroscopic Measurements

The ligand vibrations in the infrared spectra of the compound studied are nearly identical with respect to their positions and relative intensities. For all three compounds a sharp band between 3305 and 3260 cm^{-1} with a shoulder at about 3360 cm^{-1} is found which is assigned as the N–H stretching vibration. This indicates that the N–H bonds and thus the F...H bonds are of nearly equal strength, which can also be seen from Table 5. For uncoordinated SiF_6^{2-} two IR active vibrations are expected [10] at 740 (ν_3) and 480 cm^{-1} (ν_4). On coordination the symmetry of SiF_6^{2-} is lowered from O_h to D_{4h} which results in splitting of the two bands [10]. By the lowering of the symmetry the two Raman active bands, ν_1 and ν_2 (663 and 470 cm^{-1}) may become visible in the IR spectra. The free ppzH has absorptions at 440,

TABLE 5. Relevant Interatomic Distances (Å) and Angles (°) for the Hydrogen Bridges in $Cu(ppzH)_4(SiF_6)$ (I)[a]

N–H	0.975		
N(11)–F(66)	2.93(3)	N(11)–H(11)–F(66)	158(1)
H(11)–F(66)	2.00(3)		
N(11)–F(6)	2.94(1)	N(11)–H(11)–F(6)	162.1(7)
H(11)–F(6)	2.00(1)		
N(31)′–F(3)	3.01(1)	N(31)′–H(31)′–F(3)	179.0(6)
H(31)′–F(3)	2.04(1)		
N(31)′–F(33)	2.94(2)	N(31)′–H(31)′–F(33)	142(1)
H(31)′–F(33)	2.11(3)		
N(51)″–F(33)	2.81(2)	N(51)″–H(51)″–F(33)	147(2)
H(51)″–F(33)	1.94(2)		
N(51)″–F(6)	2.94(1)	N(51)″–H(51)″–F(6)	173.8(6)
H(51)″–F(6)	1.97(1)		
N(71)–F(3)	2.98(1)	N(71)–H(71)–F(3)	173.7(3)
H(71)–F(3)	2.01(1)		
N(71)–F(44)	2.88(2)	N(71)–H(71)–F(44)	143(2)
H(71)–F(44)	2.04(3)		

[a] e.s.d.s in the last significant digits are given in parentheses. Primed atoms are generated by $-x, -y, -z$; double primed atoms are generated by $1-x, 1-y, 1-z$.

TABLE 6. Relevant IR and LF Data of **I**, **II** and **III**

Compound	IR (cm^{-1})		
	N–H	SiF_6	
		ν_1	$\nu_4(\nu_2)$
$Cu(C_9H_8N_2)_4(SiF_6)$ (**I**)	3255[a]	690(b)	468 493
$Ni(C_9H_8N_2)_4(SiF_6)$ (**II**)	3290[a]	680 640	465 505
$Co(C_9H_8N_2)_4(SiF_6)$ (**III**)	3300[a]	680 640	465 505

	LF (cm^{-1})						
	CT	^2E_g	$^2B_{2g}$				$\leftarrow {}^2B_{1g}$
I	29.670	17.000	13.480				
	$^3T_{1g}(P)$	$^3T_{1g}(F)$	$^3T_{2g}$				$\leftarrow {}^3A_{2g}$
II	28.090	17.070	13.830sh	11.990	8.020		
	CT	$^4A_{2g}$	^4E_g		$^4B_{1g}$	^4E_g	$\leftarrow {}^4A_{2g}$
III	27.780	24.800sh	20.410	19.340	15.500sh	9.760	

[a] Shoulder at about 3360 cm^{-1}.

500 and 750 cm^{-1}. Therefore the exact position of the anion vibrations are difficult to determine. For **II** and **III** two bands are found at 465 and 505 cm^{-1}, which can be assigned to a splitting of the ν_4, or to the ν_2 stretching vibration and is probably a combination of those bands. Assignment of the ν_3 is not possible, because of the ligand absorptions in this region. Additional vibrations at 680 and 640 cm^{-1} can be assigned to the ν_1 stretching vibration. For **I** the splitting is smaller, vibrations are found at 468 and 493 (ν_2, ν_4) and at 690(b) (ν_1) cm^{-1}. This indicates that the symmetry of the SiF_6^{2-} group is considerably more distorted in the Ni(II) and Co(II) compounds **II** and **III** as compared to the Cu(II) compound **I**. Some relevant IR vibrations are given in Table 6. The ligand-field spectra confirm six-fold coordination of the metal in the Co(II) and Ni(II) compounds. The spectrum of **I** shows a considerable tetragonal distortion in this copper compound. The spectra for the nickel **II** and cobalt **III** compounds indicate that in these compounds the coordination sphere must be less distorted, indicating that the SiF_6^{2-} group is coordinated stronger in these compounds. As stronger coordination will result in a larger deviation from O_h

symmetry for the hexafluorosilicato group the results from the LF spectra are in accordance with the results from the IR spectra. The transitions in the ligand-field spectra are given in Table 6, together with a tentative assignment.

The compounds prepared from hydrated hexafluorosilicates have identical IR and LF spectra as the compounds **I**, **II** and **III** but powder diffraction shows the presence of some impurities.

Magnetic Susceptibility

The magnetic susceptibility measurements for **I**, **II** and **III** for these three compounds show no maximum down to 2 K. This indicates that the magnetic interaction in these compounds is very small. The structure of **I** shows that the copper ion is coordinated by four pyrazole nitrogen atoms in a square and two weaker coordinating axial ligands. This coordination results in a magnetic orbital on the copper ions consisting mainly of $d_{x^2-y^2}$. The magnetic orbitals are thus in parallel plains and no significant interaction can be expected along the chains. As all hydrogen bridge contacts for **I** are within one chain no intra-chain interaction resulting from these contacts can be expected. For Ni(II) compounds in a tetragonal coordination, the zero-field splitting is significant [11]. The magnetic data for **II** were analysed according to a model for mononuclear Ni(II), including zero-field splitting. The parameters obtained are given in Table 7. For the Co(II) compound **III** the magnetic susceptibility at lower temperatures is influenced by single-ion properties, resulting in a decrease in the value of $\chi \times T$ with decreasing temperatures. For the copper compound **I** the value of $\chi \times T$ decreases slightly upon cooling, this indicates that a small antiferromagnetic interaction is present in this compound. In Table 7 some magnetic parameters are given.

TABLE 7. Magnetic Parameters of **I**, **II** and **III**

Compound	μ (BM)		
	5 K	80 K	300 K
$Cu(C_9H_8N_2)_4(SiF_6)$ (**I**)	2.00	1.99	2.14
$Ni(C_9H_8N_2)_4(SiF_6)$ (**II**)	2.54	3.13	3.20
$Co(C_9H_8N_2)_4(SiF_6)$ (**III**)	4.43	5.13	5.30
$Ni(C_9H_8N_2)_4(SiF_6)$ (**II**)	$D = -10$ cm^{-1} $g = 2.27$		

Concluding Remarks

The compounds described in this paper are chain compounds with SiF_6^{2-} as a bridging anion. The magnetic exchange in these compounds is very small. For the Ni(II) and Co(II) compounds, the single-ion properties are more pronounced then the magnetic exchange.

Supplementary Material

The following tables are available from the authors on request: Table SI, the anisotropic temperature factors for the non-hydrogen atoms; Table SII, the positional and isotropic temperature factors of the hydrogen atoms; Tables SIII and SIV, additional interatomic distances and angles; as well as a list of observed and calculated structure factors.

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