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Dielectric relaxation spectroscopy of heteronuclear cobalt(II)–copper(II) complex of 1-phenyl-3-methyl-5-pyrazolone

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Abstract

A bright green $CoCu(PMP)(OH)_2(H_2O)_2$ complex was synthesized. Its structure was elucidated and characterized by different spectroscopic techniques. Both cobalt and copper atoms attain tetrahedral geometry. The complex was investigated by the dielectric relaxation spectroscopy. The dielectric parameters are discussed in terms of temperature and frequency changes. The conductivity of the complex decreased as the temperature increased in the temperature range 30–100 °C, while above this temperature range stepwise increase in the conductivity was observed.
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1. Introduction

Dielectric properties have attracted much attention due to the possibilities for applications to various electronic devices [1]. Owing to this fact, dielectric relaxation spectroscopy (DRS) has become a popular and powerful technique for studying the local and cooperative dynamics of almost any kind of material [2–5]. Pyrazolone compounds are of biological and chemical importance. They are used in medicine principally as hypnotic drugs and produce depressive effect on the central nervous system [6]. In the present work, it is aimed to study the dielectric relaxation properties of the heteronuclear mixed cobalt(II)–copper(II) complex of 1-phenyl-3-methyl-5-pyrazolone (PMP) ligand as a continuation of the work started to study the dielectric properties of some complexes [7,8].
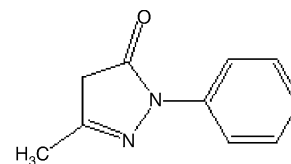
1-phenyl-3-methyl-5- pyrazolone, PMP

2. Experimental

2.1. Synthesis of the CoCu(PMP)(OH)₂(H₂O)₂ complex

To a 0.02 mol of PMP dissolved in 30 ml ethanol, an ammoniacal solution of 0.01 mol of both $CoCl_2$ and 0.01 mol $CuCl_2$ are added. The mixture was refluxed for half an hour. The precipitated complex was separated by filtration after cooling and washed several times by MeOH–H_2O mixture, followed by drying in a vacuum desiccator over anhydrous $CaCl_2$. The melting point of the complex was over 300 °C. The metal ions were determined by the usual

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Table 1
Dielectric properties of CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at 30 °C

Frequency	Potential	θ	C	D	ε	ε'	ε''	σ	$\log \sigma$	$\log \varepsilon'$	$\log \varepsilon''$
100	7.15E+06	83.3	0.00E+00	0	0.00	0.00	0.00	1.68E−05	−4.78		
200	7.18E+06	83.7	0.00E+00	0	0.00	0.00	0.00	1.78E−05	−4.75		
300	7.17E+06	84.2	0.00E+00	0	0.00	0.00	0.00	1.93E−05	−4.71		
400	7.17E+06	84.4	2.66E−10	0.47	420.77	418.76	41.06	2.00E−05	−4.70	2.62	1.61
500	6.87E+06	84.6	2.11E−10	0.05	333.77	332.29	31.41	2.17E−05	−4.66	2.52	1.50
600	6.87E+06	82.7	1.81E−10	0.043	286.16	283.84	36.36	1.60E−05	−4.79	2.45	1.56
700	6.87E+06	82.7	1.66E−10	0.057	262.43	260.30	33.35	1.60E−05	−4.79	2.42	1.52
800	6.87E+06	79.2	1.28E−10	0.066	202.63	199.04	37.97	1.09E−05	−4.96	2.30	1.58
900	6.87E+06	80.2	1.15E−10	0.043	181.28	178.63	30.6	1.20E−05	−4.92	2.25	1.49
1000	6.59E+06	81.1	1.14E−10	0.061	179.54	177.38	27.78	1.37E−05	−4.86	2.25	1.44
2000	6.33E+06	80.5	5.71E−11	0.075	90.32	89.09	14.91	1.34E−05	−4.87	1.95	1.17
3000	5.27E+06	79.1	3.86E−11	0.098	61.06	59.96	11.55	1.40E−05	−4.85	1.78	1.06
4000	7.95E+06	78.9	2.97E−11	0.131	46.98	46.10	9.04	1.47E−05	−4.83	1.66	0.96
5000	4.62E+06	78.9	2.41E−11	0.145	38.12	37.41	7.34	1.57E−05	−4.80	1.57	0.87
6000	4.17E+06	79.2	2.11E−11	0.122	33.38	32.79	6.25	1.79E−05	−4.75	1.52	0.80
7000	3.86E+06	79.0	1.87E−11	0.128	29.58	29.04	5.64	1.90E−05	−4.72	1.46	0.72
8000	3.52E+06	78.6	1.68E−11	0.167	26.57	26.05	5.25	2.01E−05	−4.70	1.42	0.75
9000	3.23E+06	78.5	1.58E−11	0.170	24.99	24.49	4.98	2.17E−05	−4.66	1.39	0.70
10000	2.99E+06	78.1	1.45E−11	0.179	22.94	22.44	4.73	2.27E−05	−4.64	1.35	0.67
20000	1.78E+06	75.4	8.60E−12	0.221	13.60	13.16	3.43	3.12E−05	−4.51	1.12	0.54
30000	1.28E+06	73.6	7.40E−12	0.243	11.71	11.23	3.30	3.88E−05	−4.41	1.05	0.52
40000	1.01E+06	72.5	7.00E−12	0.26	11.07	10.56	3.33	4.16E−05	−4.34	1.02	0.52
50000	8.40E+05	71.9	6.60E−12	0.269	10.44	9.92	3.24	5.37E−05	−4.27	1.00	0.51
60000	7.16E+05	71.4	6.50E−12	0.271	10.28	9.74	3.28	6.13E−05	−4.21	0.99	0.52
70000	6.17E+05	71.4	6.40E−12	0.275	10.12	9.60	3.23	7.12E−05	−4.15	0.98	0.51
80000	5.41E+05	71.8	6.40E−12	0.262	10.12	9.62	3.16	8.29E−05	−4.08	0.98	0.50
90000	4.74E+05	73.0	6.60E−12	0.239	10.44	9.98	3.05	1.01E−05	−4.00	1.00	0.48
1000000	4.14E+05	75.2	7.10E−12	0.225	11.23	10.86	2.87	1.32E−05	−3.88	1.04	0.46

complexmetric titration methods [9]. Electronic absorption spectra were measured using a PYE-Unicam spectrophotometer model 1750 covering the wavelengths from 190 to 900 nm. Infrared spectra were recorded using a Perkin-Elmer spectrophotometer model 1430 covering the frequency range 200–4000 cm^{-1}. The analytical data of the bright green CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex was: Co (16.1%, found 16.2%); Cu (17.3%, found 17.3%); C (32.7%, found 32.8%); H (4.4%, found 4.6%) and N (7.6%, found 7.7%).

The CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex was pressed into disks of 10 mm diameter and 1–2 mm thickness at a pressure of 9.8×10^8 Pa. Silver paste was painted on the major faces of each tested piece as electrodes. The measurements of the dielectric constant and the dielectric loss were performed in the frequency range 10^2–10^5 Hz and temperature range 30–140 °C. Tables 1 and 2 represent the obtained data at 30 and 140 °C, respectively.

3. Results and discussions

The structure of the CoCu(PMP(OH)$_2$(H$_2$O)$_2$ complex is critically discussed. The fundamental IR bands of the mixed metal complex gave the following points:

1- The broad band at 3442 and 3444 cm^{-1} in the ligand and its mixed complex, respectively, are due to hydrogen bond interaction of the ligand and some water molecules in the complex [10].

2- The ligand ν_{NH} band at 3130 cm^{-1} disappeared completely on complexation to assign M−N bonding [10].

3- The ligand carbonyl vibrations at 1729, 1716 and 1696 cm^{-1} are nearly not affected on complexation.

4- The $\nu_{C=N}$ and $\nu_{C=C}$ overlapped in the free ligand at 1603 and 1521 cm^{-1}, are slightly affected on complexation with the presence of splitted bands in the range 1643–1616 cm^{-1}.

5- The ν_{C-N} and ν_{N-N} bands at 1348, 1305 and 1245 cm^{-1} in the ligand are slightly affected with different degrees by complexation.

6- The ligand methyl group vibration bands at 1456 and 1391 cm^{-1} are splitted into three bands at 1449, 1411 and 1389 cm^{-1} on complexation with different intensities. These shifts proposed some sort of M−C bonding [10].

7- A remarkable feature is given, where the ν_{CH} of the free ligand vanished on complexation, to point, again the probable M−C bonding [10].

8- Some metal sensitive bands are observed at (ν_{M-O} at 590 and 508 cm^{-1} and ν_{M-N} at 432 and 408 cm^{-1}) [10].

The Nujol mull electronic absorption spectra of the brown mixed complex gave five bands at 245, 297, 333, 360 and

Table 2
Dielectric properties of CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at 140 °C

Frequency	Potential	θ	C	D	ε	ε'	ε''	σ	$\log \sigma$	$\log \varepsilon'$	$\log \varepsilon''$
100	1.81E+06	72.1	0.00E+00	0.000	0.00	0.00	0.00	2.52E−05	−4.60	–	–
200	1.81E+06	71.0	0.00E+00	0.000	0.00	0.00	0.00	2.38E−05	−4.62	–	–
300	1.54E+06	71.0	0.00E+00	0.000	0.00	0.00	0.00	2.80E−05	−4.55	–	–
400	1.45E+06	71.4	9.00E−12	0.891	14.24	13.49	4.54	3.02E−05	−4.52	1.13	0.66
500	1.41E+06	71.1	8.10E−12	0.830	12.81	12.12	4.15	3.07E−05	−4.51	1.08	0.62
600	1.39E+06	71.2	7.30E−12	0.769	11.55	10.93	3.72	3.13E−05	−4.50	1.04	0.57
700	1.38E+06	71.1	7.00E−12	0.716	11.07	10.48	3.59	3.13E−05	−4.50	1.02	0.55
800	1.38E+06	70.2	6.60E−12	0.674	10.44	9.82	3.54	3.00E−05	−4.52	0.99	0.55
900	1.31E+06	70.1	6.30E−12	0.646	9.97	9.37	3.39	3.14E−05	−4.50	0.97	0.53
1000	1.31E+06	69.9	6.10E−12	0.624	9.95	9.06	3.32	3.11E−05	−4.51	0.96	0.52
2000	1.28E+06	69.3	4.90E−12	0.547	7.75	7.25	2.74	3.10E−05	−4.51	0.86	0.44
3000	1.23E+06	68.9	4.50E−12	0.522	7.12	6.64	2.56	3.17E−05	−4.50	0.82	0.41
4000	1.19E+06	68.2	6.10E−12	0.441	9.65	8.96	3.58	3.16E−05	−4.50	0.95	0.55
5000	1.15E+06	68.7	5.60E−12	0.414	8.86	8.25	3.22	3.36E−05	−4.47	0.92	0.51
6000	1.13E+06	69.3	4.80E−12	0.397	7.59	7.10	2.68	3.52E−05	−4.45	0.85	0.43
7000	1.09E+06	70.1	4.50E−12	0.377	7.12	6.69	2.42	3.79E−05	−4.42	0.83	0.38
8000	1.04E+06	70.1	4.40E−12	0.375	6.96	6.54	2.37	3.95E−05	−4.40	0.82	0.37
9000	1.00E+06	71.1	4.10E−12	0.369	6.49	6.14	2.10	4.31E−05	−4.37	0.79	0.32
10000	9.56E+05	71.3	3.90E−12	0.354	6.17	5.84	1.98	4.57E−05	−4.34	0.77	0.30
20000	6.90E+05	73.8	3.20E−12	0.319	5.06	4.86	1.41	7.28E−05	−4.14	0.69	0.15
30000	5.12e+05	74.1	2.90E−12	0.299	4.59	4.41	1.26	9.81E−05	−4.01	0.64	0.10
40000	4.12E+05	74.8	2.80E−12	0.284	4.43	4.27	1.16	1.27E−04	−3.90	0.63	0.06
50000	3.54E+05	75.3	2.70E−12	0.271	4.27	4.13	1.08	1.56E−04	−3.81	0.62	0.03
60000	3.03E+05	76.1	2.70E−12	0.252	7.27	4.15	1.03	1.92E−04	−3.72	0.62	0.01
70000	2.69E+05	76.9	2.70E−12	0.237	4.27	4.16	0.97	2.30E−04	−3.64	0.62	−0.01
80000	2.40E+05	78.2	2.70E−12	0.212	4.27	4.18	0.87	2.85E−04	−3.54	0.62	−0.06
90000	2.11E+05	80.1	2.80E−12	0.176	4.43	4.36	0.76	3.86E−04	−3.41	0.64	−0.12
1000000	1.88E+05	83.0	3.00E−12	0.122	4.75	4.71	0.58	6.11E−04	−3.21	0.67	−0.24

650 nm to assign four-coordinated structure of both cobalt and copper. From the previous data gathered with its elemental analysis the structure of the complex is suggested as follows:

$$H_2O \rightarrow M \cdots \text{(structure)}$$

M= Co^{2+}, or Cu^{2+}
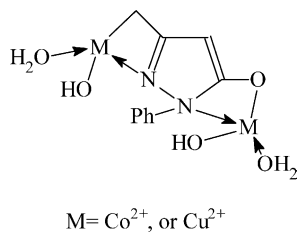
3.1. Dielectric properties

The mode of studies is based on changing the frequency from 0.1 to 100 kHz and recording the potential difference, *V*, the phase angle, θ, the dielectric loss, *D* and the capacitance, *C*, in the temperature range 30–140 °C. However, the ac conductivity is frequency independent in the range (10^2–10^4 Hz), and their values are similar to those of the dc conductivity. Such results are in accordance with the obtained data based on the following equation [5]: $\sigma_{(\omega)} = \sigma_{dc} + \text{const}\,\omega^n$, where $\omega = 2\pi f$ and *n* is a constant depends on the nature of the material. Figs. 1 and 2 represent, respectively, the potential difference and conductivity values versus the frequency of the complex at different temperatures. However, at high

frequencies (10^4–10^5 Hz), the conductivity increases with increasing the frequency. On plotting the ac conductivity, frequency relation in logarithmic representation, it is found that the conductivity decreases as the temperature increases in the temperature range 30–100 °C. Further increase of the temperature, up to 140 °C, stepwise increase in the conductivity occurs. The *n* values of each frequency region are collected in Table 3. The intercept of the straight line gave the corresponding dc conductivity values.

The $\log[\sigma_{ac}-T]$ 1000/*T* relation at different frequencies can be used to calculate the ΔE values based on the following
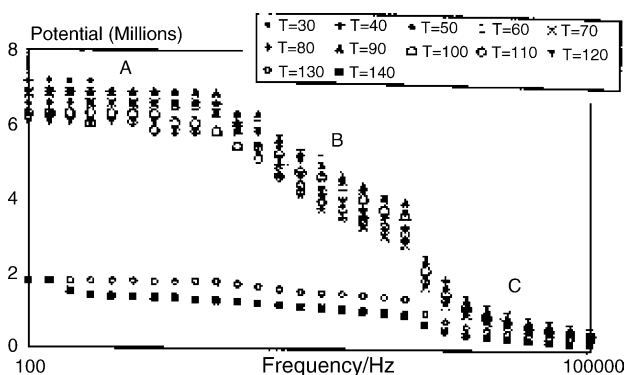

Fig. 1. Potential difference vs. frequency for CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different temperatures (°C).
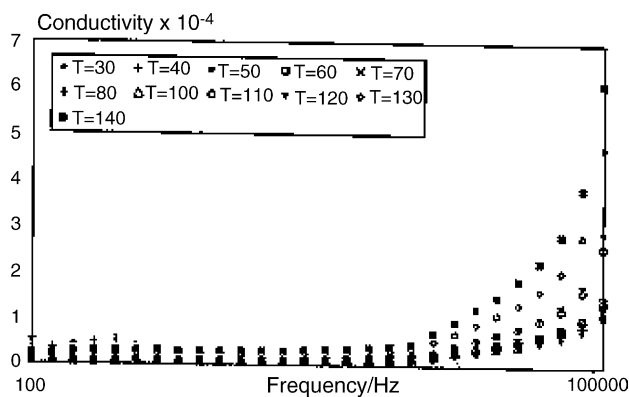
Fig. 2. Conductivity vs. frequency for CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different temperatures.

Table 3
n and σ_{dc} values for the CoCu(PMP)·4H$_2$O complex

Temperature (°C)	n^a	n^b	n^c	$\sigma_{dc}{}^a$	$\sigma_{dc}{}^b$	$\sigma_{dc}{}^c$
30	0.175	0.422	0.866	−5.29	−6.70	−8.97
50	∼zero	0.554	0.981	−5.32	−7.57	−9.50
80	∼zero	0.612	1.210	−5.24	−8.92	−10.80
120	∼zero	0.729	1.268	−5.16	−8.31	−10.93

[a] In the frequency range, 0.1–0.9 kHz.
[b] In the frequency range, 1–10 kHz.
[c] In the frequency range, 10–100 kHz.

equation [5], Table 4.

$$\sigma_{ac} = \frac{\sigma_o}{T e^{-\Delta E_a / KT}}$$

$$\log[\sigma T] = \log \sigma_o - \Delta E_a / KT$$

The permitivity value ε, depends on the changeable external factors (the frequency of the voltage applied to the dielectric and the temperature). It was reported that the permitivity of non-polar dielectrics does not depend on frequency when it changes within the very broad limits [11]. Also, for cases with dipole polarization, when the frequency of alternating voltage increases, the value of ε for a polar dielectric at first remains invariable, and then begins to decrease by increasing the frequency. At very high frequencies, ε reaches the values typical to non-polar dielectrics (Fig. 3). A remarkable feature is given where at low temperature (till 40 °C), the complex acts as a polar dielectric, whereas at a high temperatures (>40 °C), it is of non-polar behavior. However, the temperature does not affect the process of electronic polarization in non-polar dielectrics. The ε values versus temperature relationship at selective frequencies (4×10^2, 9×10^2, 7×10^3,

Table 4
Values of ΔE for the CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different frequencies

Frequency (kHz)	ΔE (eV)
0.1	0.059
1.0	0.049
10.0	0.109
100.0	0.351
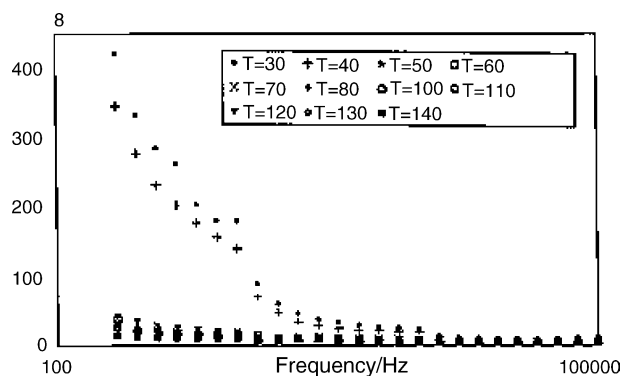
Fig. 3. Permitivity vs. frequency for CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different temperatures.

2×10^4 and 7×10^4 Hz) gave that at very high frequency, the complex is of non-polar dielectrics property. The course of change of ε as a function of temperature, when the latter changes within broad limit (assuming that the matter does not undergo chemical transformation) [11], at a medium frequency (7×10^3 Hz), gave that the complex at is also a non-polar dielectric. The sharp change in ε can be attributed to the probable transition of the material from one phase to another. However, in some cases, the value of ε may diminish when the temperature rises, particularly in substances, where ionic displacement intensifies the internal field and thereby the electronic polarization. This case is assigned at low frequencies (4×10^2 and 9×10^2 Hz).

The locus of ε' (real value of permitivity) and ε'' (imaginary value of permitivity) for various frequencies in the rectangular axes of coordinates ε' and ε'' should form a semi-circle for polar dielectrics, with the center lying axis with coordinates $(\varepsilon'_{st} \pm \varepsilon'_{opt})/2.0$ [11].

The ε_{st} is the static permitivity at direct voltage and ε'_{opt} is the optical permitivity for high frequencies approaching the frequencies of light oscillations [11]. This is in harmony with the well-known Cole–Cole diagrams for CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different temperatures (30–140 °C). At low temperatures (30–40 °C) this diagram is almost applied where ε'_{st} and ε'_{opt} amount 650 and 587.5 at 30 °C and amount 441.6 and 420.8 at 40 °C, respectively. However, at higher temperature (>40 °C), where the complex is of non-polar property, the Cole–Cole diagram is not obeyed. The logarithmic value of ε was plotted against log ω at different temperatures according to the relations:

$$\varepsilon \alpha \omega^{-n}$$

$$\log \varepsilon = \text{const} - n(\log \omega)$$

where n is a constant depends on the nature of the material. Best-fit straight lines are obtained at the different temperatures selected for the studies the n values are computed and given in Table 5.

In general, the ε' values due to the intrinsic properties of the complex decreased with increasing the temperature

Table 5
n Values for the $CoCu(PMP)(OH)_2(H_2O)_2$ complex at different temperatures

Temperature (°C)	Frequency (n) (kHz)		
	0.1–0.9	1–10	10–100
30	0.921	0.91	0.26
50	0.51	0.41	0.11
80	0.32	0.41	0.11
120	0.78	0.63	0.11

Table 6
n Values for the $CoCu(PMP)(OH)_2(H_2O)_2$ complex at different temperatures and frequencies

Temperature (°C)	Frequency (n) (kHz)		
	0.1–0.9	1–10	10–100
30	0.44	0.70	0.09
50	0.71	0.50	0.12
120	0.54	0.50	0.33

and frequency. When the logarithmic value of the dielectric, ε''_{opt} due to the extrinsic properties [12] is plotted against $\log \omega$ based on the following equations [12] we can obtain the slope of this equation, n, Table 6.

$$\varepsilon'' = \frac{\sigma_{ac}}{\varepsilon_o \omega^n}$$

$$\log \varepsilon'' = \frac{\log \sigma_{ac}}{\varepsilon_o - n \log \omega}$$

It seems that the ε'' values decreased with increasing the frequency and temperature.

ε_o (the dielectric constant in vacuum) can be calculated at any given ac conductivity using the intercept ($\log \sigma_{ac}/\varepsilon_o$). An example is given in Table 7 that collects the ε_o values at different temperatures.

The dependence of capacitance on frequency is determined based on the following equation [11]:

$$C = C_g + \frac{s\tau}{\varpi^2 \tau^2 + 1}$$

where C is the capacitance of the given dielectric, C_g the geometrical capacitance, s the conduction corresponding to absorption current and τ is the relaxation time.

Thus, the geometrical capacitance corresponds to the capacitance at a very high frequency when the effect of the reactive component of absorption current does not practically manifest itself at all [11]. At low temperature (30–40 °C), the capacitance decreases as the frequency increases. At high temperature (>40 °C), the capacitance is independent of the

Table 7
ε_o Values for $CoCu(PMP)(OH)_2(H_2O)_2$ complex at different temperatures

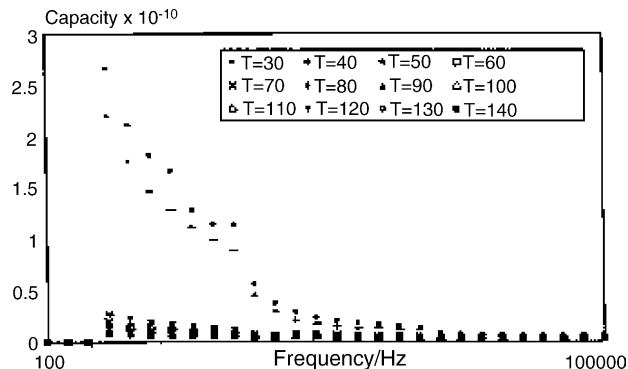| Temperature (°C)	Frequency (Hz)		
	10^2–10^3	10^3–10^4	10^4–10^5
30	2.8×10^{-8}	3.4×10^{-9}	3.9×10^{-6}
50	1.9×10^{-8}	2.1×10^{-8}	1.4×10^{-6}
120	4.6×10^{-9}	3.4×10^{-8}	6.9×10^{-7}

Fig. 4. Capacity vs. frequency for $CoCu(PMP)(OH)_2(H_2O)_2$ complex at different temperatures.

frequency (Fig. 4). Table 8 collects the values of C_∞, C_o and $s\tau$ at the temperatures 30 and 40 °C.

The dielectric loss, D values depend on various external factors. The behavior of dielectric loss in the low frequency is connected with the character of the phase transition in some cases [11]. The nucleation of a new phase and the further growth of the nuclei are regarded as the main source of the peak value for D at the phase transition temperature. The dependence of D on frequency of an electrical insulating material observed different natures of dependence in the range of the frequency change, either increase or decrease at a higher frequency, or transition through the maximum. At a temperature <50 °C, the dielectric loss increases by increasing the frequency of the applied voltage. However, at 50 and 60 °C, the dielectric loss passes through a maximum at around 2500 Hz. Further increase of temperature turns the complex into non-polar dielectric as shown from the decrease of the dielectric loss by increasing the frequency. The $\log D$ related to $\log \omega$ at different temperatures (Fig. 5) based on the following equations:

$$D = \left(\frac{1}{RC\omega} \right)^x$$

$$\log D = \frac{x \log 1}{RC - x \log \omega}$$

At temperature (30–40 °C), the relation gave a best-fit line with the same slope in all frequencies studied. By increasing the temperature (>40 °C), a simultaneous increase and a decrease in the $\log D$–$\log \omega$ relationship is observed as a result of phase transitions in the $CoCu(PMP)(OH)_2(H_2O)_2$ complex.

In polar substances, the dependence of D on temperature at different frequencies gave that the ordinates of the plot are

Table 8
C_∞ and C_o values of $CoCu(PMP)(OH)_2(H_2O)_2$ complex at different temperatures

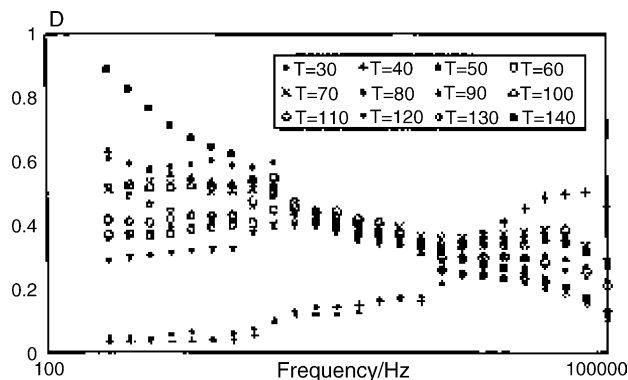| Temperature (°C)	$C_\infty \times 10^{-12}$	$C_o \times 10^{-11}$	$s\tau \times 10^{-11}$
30	6.5	26.60	25.95
40	4.4	21.90	21.46

Fig. 5. *D* vs. frequency for CoCu(PMP)(OH)$_2$(H$_2$O)$_2$ complex at different temperatures.

the sum of the ordinates of two components: (A) produced by purely dipole losses and (B) caused by the losses due to electrical conduction. On increasing the frequency, the data are characterized to the purely dipole losses.

If the dielectric is a mixture of two or more polar substances, the dependence of *D* on temperature may have two or more maximums produced by the effect of the separate components of the mixture. In some cases, the curve of a chemically individual dielectric may have maximum. This is due to the different character of the losses (dipole and structural, etc. . . .).

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