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Topoenergetic aspects of water structuring as revealed by ac electric conductivity

Summary

Accurate ac measurements of R and C by using a RLC bridge (Agilent U1732B) were performed in two cities located at long distance each other on successively diluted solutions of ionic salts starting from the same stock saturated solutions and using local tap water as solvent. The results obtained by Universal representation of R and C as function of frequency as driving potential are discussed and compared with the results obtained in other two ones for more restricted conditions.

Introduction

Topoenergetic aspects of water structuring by ionic salts was thoroughly studied in a series of previous works by using calorimetry [1-5], densimetry, thermo-mechanic analysis [5], ac electric conductivity [6] and vapor-liquid equilibria [7,8]. The main conclusions were that (i) cations can perform specific hydrates as supramolecular structures; (ii) the following series of cations in decreasing order of water structuring potential was established in the molecular series of solutes MeSO4 [1, 5]:

$$\text{Me: Cu} > \text{Na2} > \text{Fe} > \text{Ni} > \text{Mn} > \text{Mg} \qquad (1);$$

(iii) the following series of cations and anions in decreasing order of water structuring potential was established for the series of solutes XY [7,8]:

$$\begin{array}{l} \text{X: Li} > \text{Mg} > \text{Na} > \text{K} \qquad (2); \\ \text{Y: F} > \text{I} > \text{Br} > \text{Cl} \end{array}$$

(iv) in any saturated aqueous solution so-called “crystalline” water remains non-attacked by solute, so that even pure water has a composite structure mainly consisting in an amorphous and a crystalline phase.

To suggestively evidence these conclusions let consider the solubility of some ionic salts in different hydrated structures [9]. Figure 1 shows the parameters (E, K) obtained by Arrhenius representation [10] of

θ = saturation solute concentration expressed in
grams of anhydrous salt/grams of water.

If there would not be any structural difference between water in hydrated salts and water solvent (amorphous phase), all species of LiBr and LiI solutes would be represented by a singular point, respectively. But, each species in a series shows different solubility depending on hydration water content and structure.

Experimental

Stock solutions were kept at room temperature for at least 3 years and the clear solutions were used after mixing before.

Measurements were performed in Sydney (Australia) and Bucharest (Romania) (October 2010) in conditions of repeatability, excepting the water as solvent and location. Local tap water was initially boiled, cooled at room temperature (20.5 ± max 0.5 ^{0}C) and immediately used.

The following solutes are used: $CuSO4.5H2O$; $Na2SO4$; $NaHCO3$; $K2Cr2O7$.

Conductivity cell was home made with the dimensions of 2x5x10 mm. The portable LRC bridge Agilent U1732B was used for R and C measurements at 100, 120 Hz, 1, 10 kHz with the same voltage amplitude.

Measurements were performed at room temperature on 20 ml solution starting with the stock solution considered with the concentration 1 (in relative units) and successively diluted by extracting 5 ml and adding 5 ml of solvent by using a syringe also for mixing (without air addition), so that the dilution ratio is 0.75.Measurements on pure solvents were performed separately.

Results and discussion

Dependences of R (in Ohm) and C (in nF) on solute concentration (x=[solute] in relative units) are trivial, namely they have a decreasing and increasing pure sigmoidal shape, respectively. The best sigmoidal representation for C is:

$$S(C, x) = (a+b*x)/(c+x), \qquad (3),$$

where b is the saturation value for C.

First of all it is important to note that R*C represents the relaxation or decay time of dipoles created by ac excitation. This time depends on ac frequency, f, more exactly it decreases with f, or the proper dipole frequency fp = 1/(R*C) increases with f of applied ac. The ratio f/fp = f*(R*C) explains better the relationship between applied perturbation and solution reaction.

Figures 2 - 5 show this value as function of x for $CuSO4$ and $Na2SO4$ in the two cities. To note the following observations:

- this ratio is always < 1, i.e. the dipole reaction is faster than applied perturbation;

- its dependence on x has also a sigmoidal shape with value almost zero for $x=0$ (dipoles are small and faster) and increases to a saturation value for $x=1$;
- fp at any f and for all solutes has decreasing sigmoidal shape vs x showing also that at $x=0$ dipoles are small and faster and increase with x;
- the following decreasing order results for fp at $f=100$ Hz and $x=1$:

$$\text{fp:}\quad \text{CuSO}_4(\text{Sydney}>\text{Bucharest}) > \text{Na}_2\text{SO}_4(\text{Sydney}>\text{Bucharest}) \qquad (4)$$

showing the decreasing order for dipole size (electric mass) at $f=100$ Hz and $x=1$;

- both measurement series show specific transitions in dipole structure for $CuSO_4$ at approximately $x=1$ for 100 and 120 Hz;
- dependence on f shows a different dispersions both for the two solutes and cities.

In view to evaluate in more detail these kinetic and structural aspects, Universal representation of R, C as eigenvalue = θ and f as driven potential will be considered, namely:

$$U(\theta, f):\quad \ln(\theta) = N*\ln(f-fo) + M \qquad (5)$$

where parameters (N, M, fo) define the nature and the amplitude of electric conductivity process. It has to note that for both eigenvalues:

$$N,\ n1 < 0,\ N*n1>0, \text{ process polarity P is negative,} \qquad (6)$$
$$M \sim \ln Ctr,\ -M/N \sim \ln ctr,\ -N^2/M \sim CS$$

where Ctr is the value of transforming component in the tested specimen, ctr is the value of kinetic entity and CS is the coupling strength between inert (Cin) and Ctr (see more details in [10] and citations therein).

Figures 6-13 show these ontogenic parameters as function of x for $CuSO_4$ and Na_2SO_4 in both series of measurements.

It is to observe the followings:

- all ontogenic parameters show sigmoidal dependence on x;
- all parameters show maximum dispersion at approximately $x=2$ and tend to unique values for $x=0$ and $x=1$ (excepting ln ctr (C));
- ctr (C) represents the dipole size (electric mass) which stores electric energy and increases with x, so that (Figure 11):

$$\text{Na}_2\text{SO}_4(\text{Sydney}>\text{Bucharest}) > \text{CuSO}_4(\text{Bucharest}>\text{Sydney}) \qquad (7);$$

- the overall process amplitude (Ctr = sum of all ctr) shows a reverse order (Figure 10) and CS the same order (Figure 12).

Pyramidal structure of ontogenic and phylogenic parameters is given in Table 1. All uncertainties mentioned in Tables and Figures are standard deviations with confidence level of 68.3%. It results differences in (n1, m1) parameters corresponding to measurements in the two cities especially for $CuSO_4$.
The following linear relationship was observed:

$$L(b, \ln(f)): \quad b = n1 * \ln(f) + m1 \qquad (8).$$

Pyramidal structure of these representations is given in Table 2. It is also important to note the following observations:

- $n1<0$ for all measurements is consistent with the initially mentioned fact that saturation values of C decrease with f;
- important differences between solutes and cities result, for the overall process amplitude (Ctr), so that:

$$|n1|, m1: \quad CuSO_4(Sydney>Bucharest) > Na_2SO_4(Bucharest>Sydney) \quad (9)$$

which is exactly as the above mentioned observation for Universal representation (Figure 10). The all story is graphically shown in Figure 14. Here m1 represents Ctr (not in logarithmic scale as in the previous Universal representation).

Concluding remarks

1. R and C values of successively diluted solutions are measured in the same standard experimental conditions and represent the dissipative and capacitive elements in energy circuits of the tested specimens, respectively;
2. Process of dipole creation as the result of applied ac voltage can be described by Universal representation in view to reveal its nature and amplitude for all tested solutions;
3. All tested aqueous solutions show the same nature of this process, so that a unique pyramidal structure results;
4. However, important structural differences in process amplitude have resulted both between solutes and locations of measurements (including the local water as the solvent);
5. Negative values of threshold fo for both $\theta = R, C$, show the existence of dipoles next to the electrodes (double layers) without and before ac excitation.

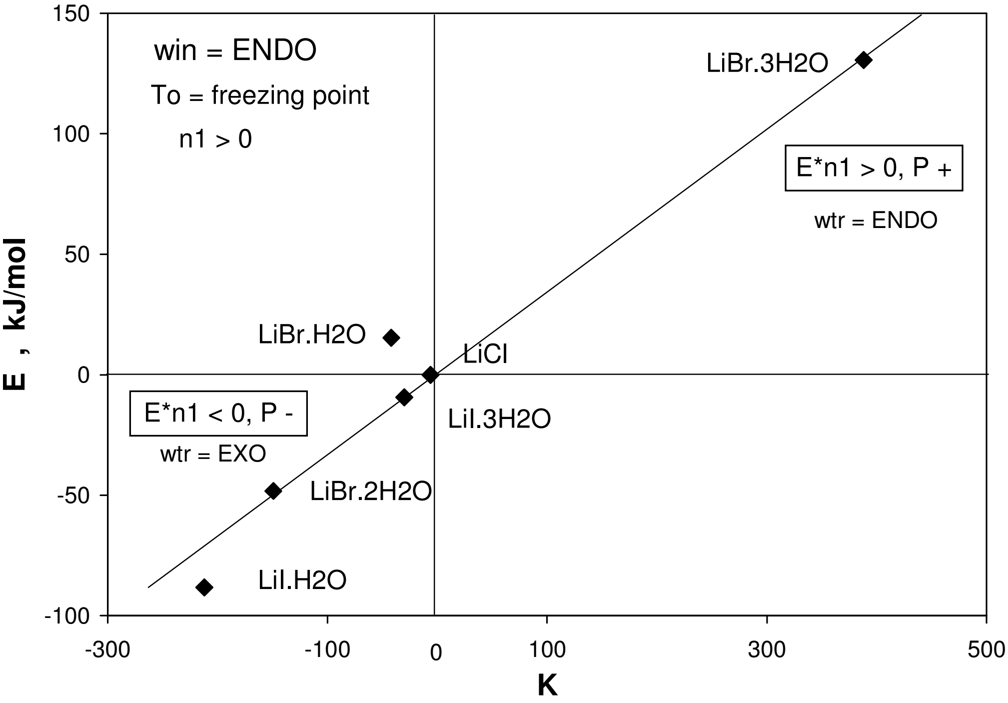


Figure 1.

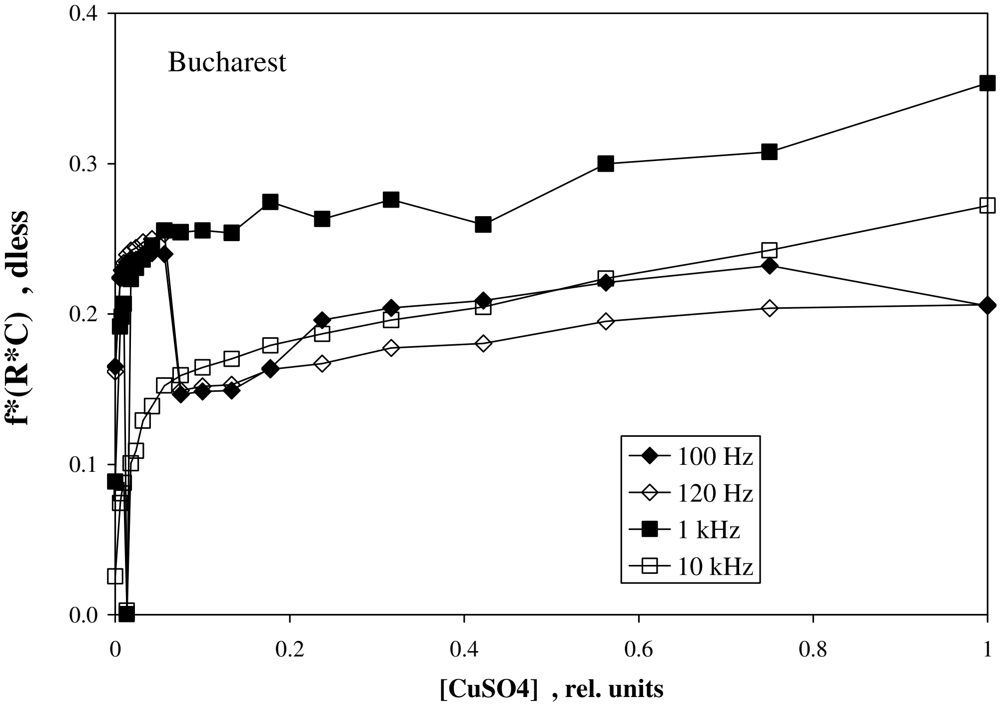
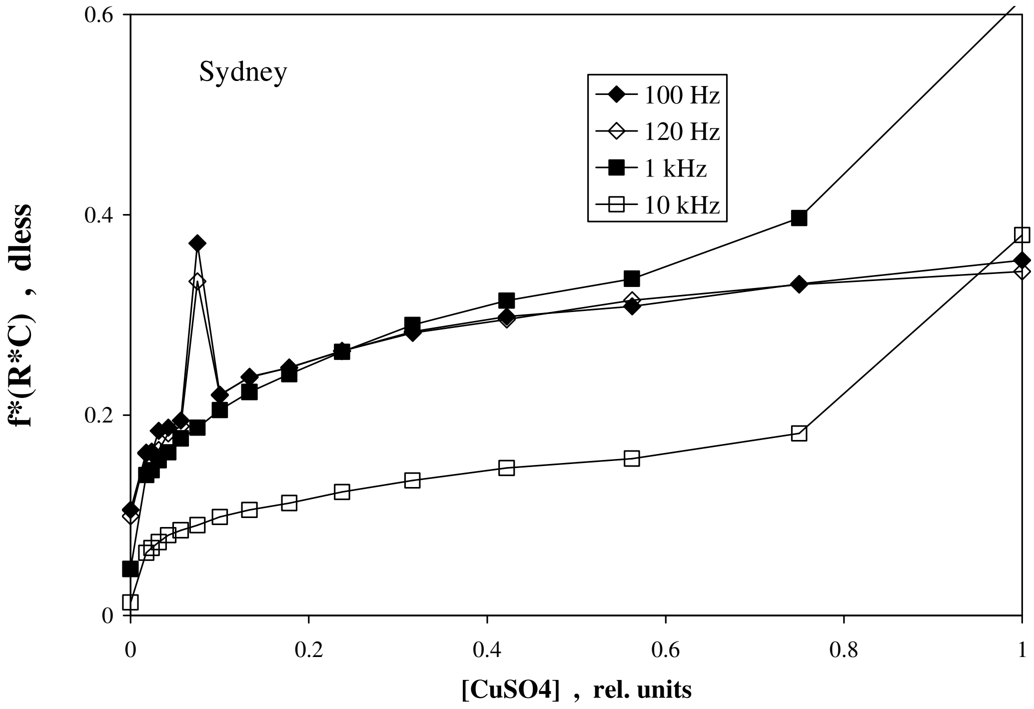


Figure 2.



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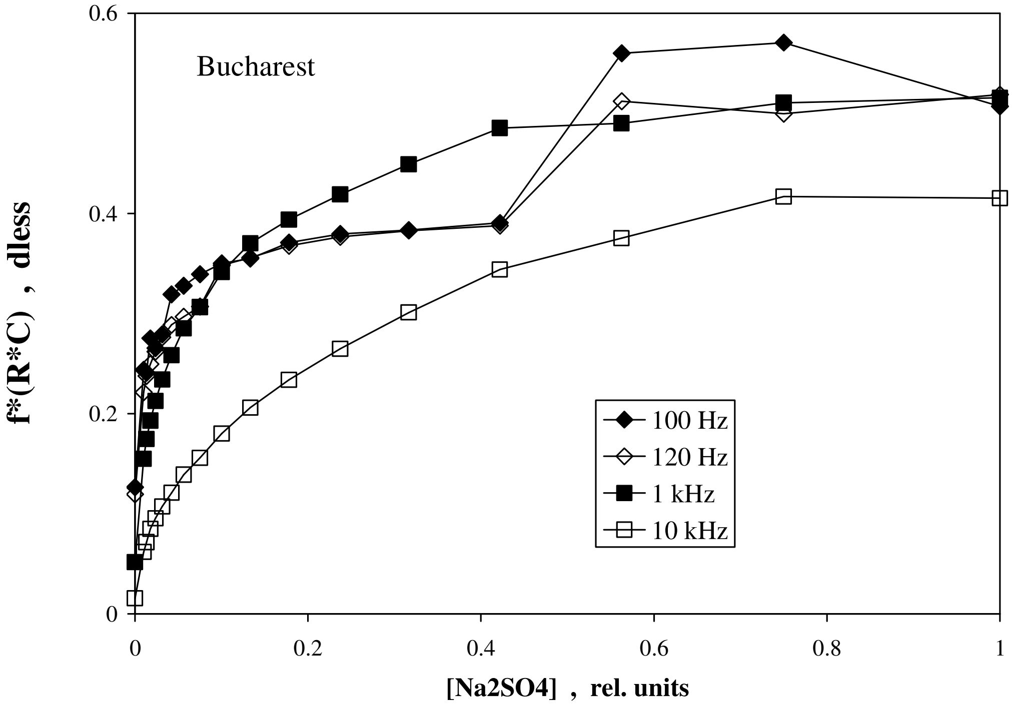

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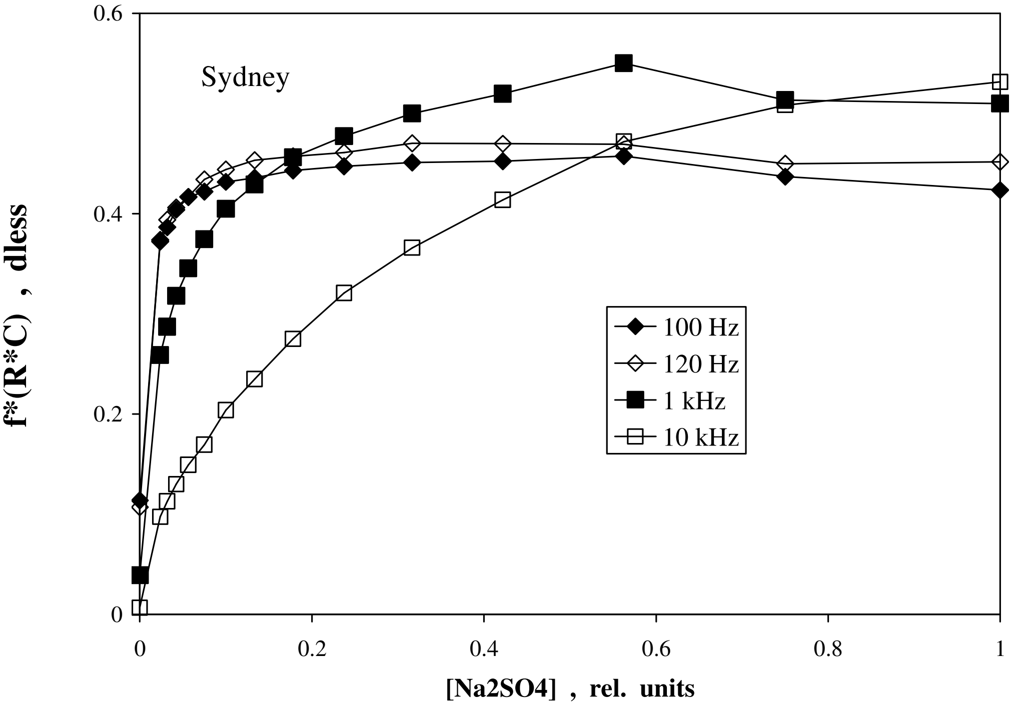
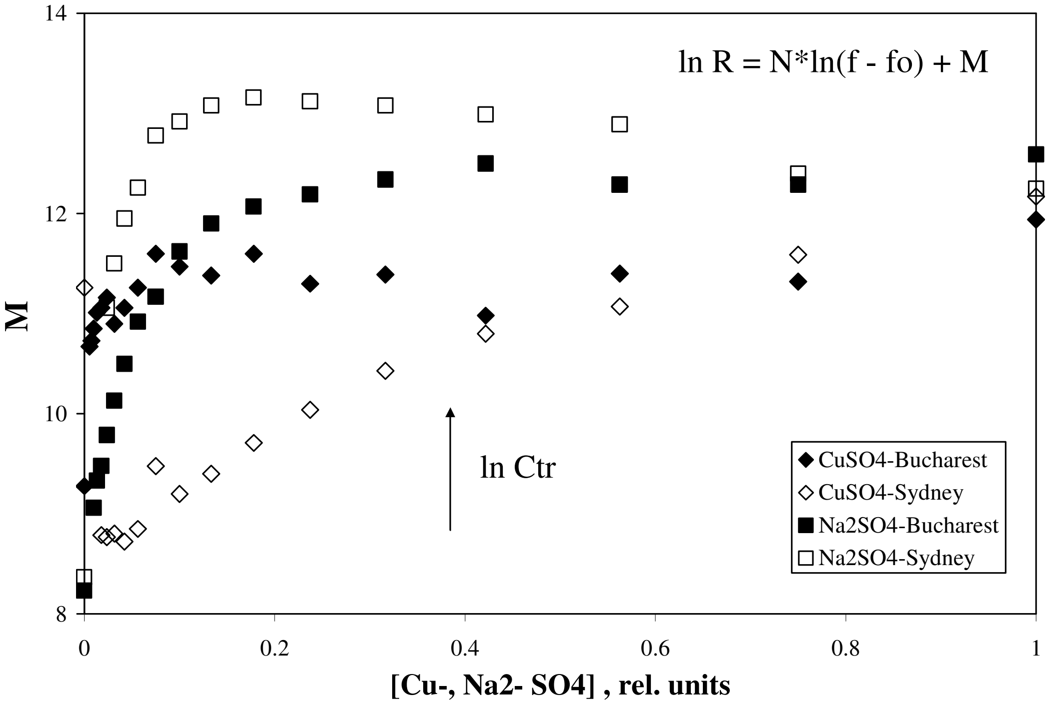


Figure 5.



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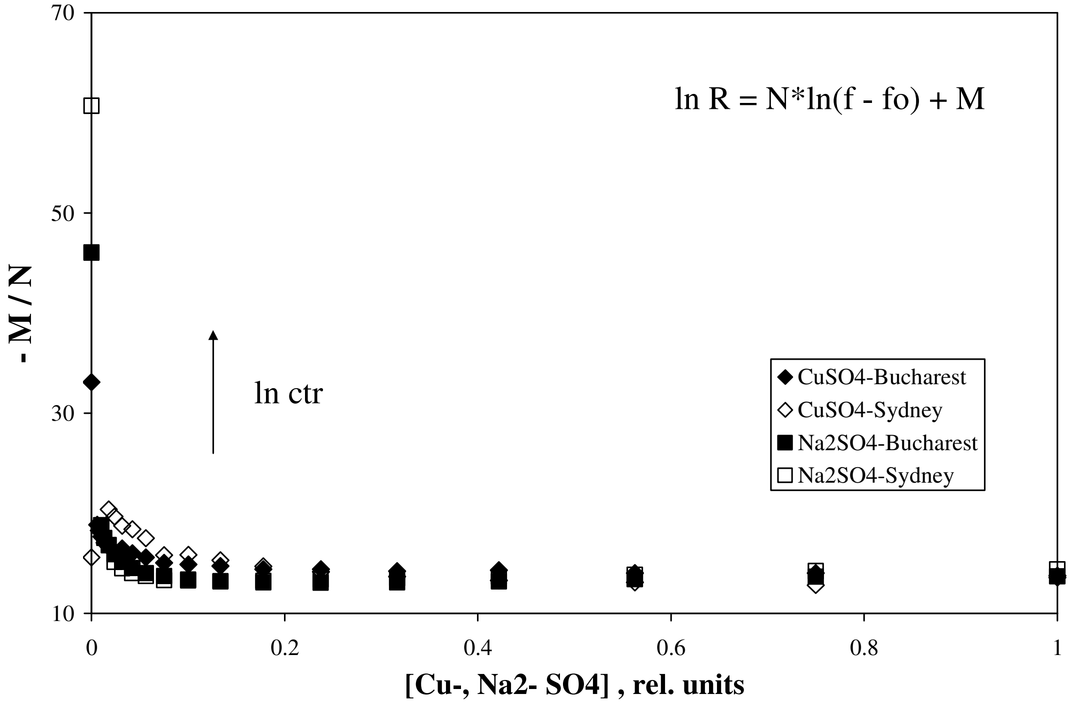

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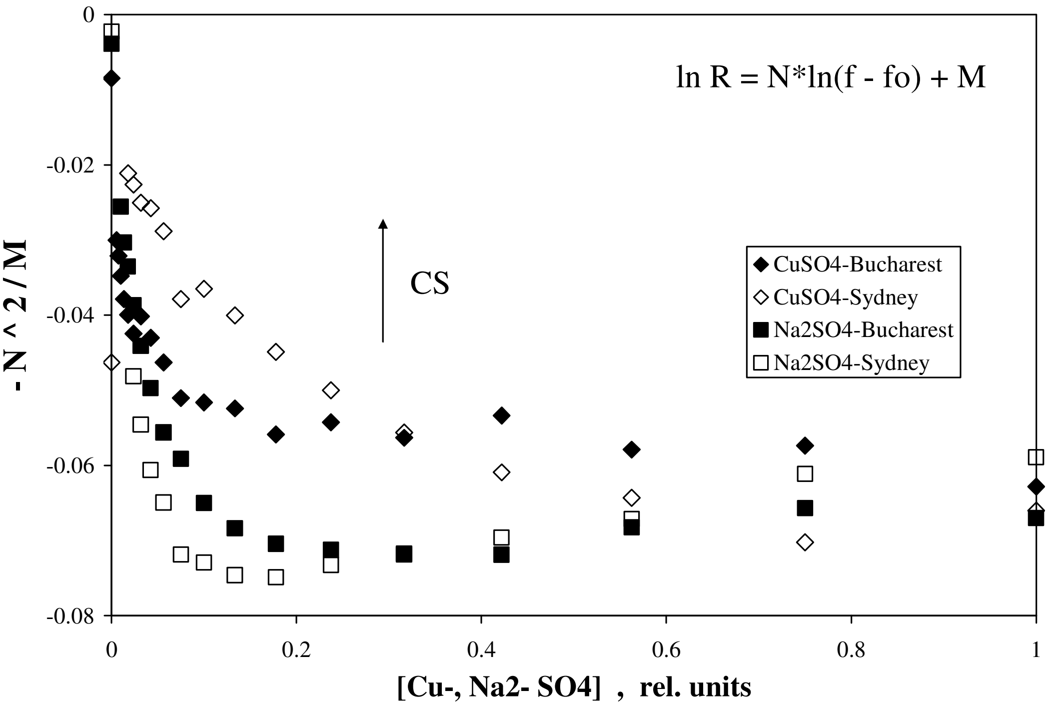

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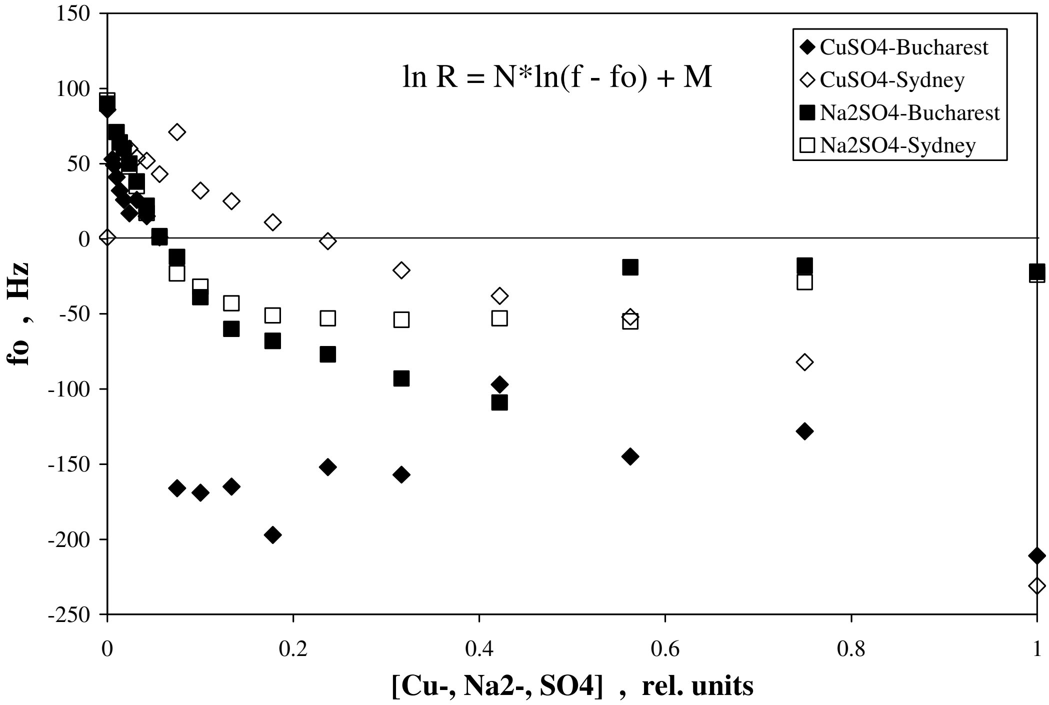

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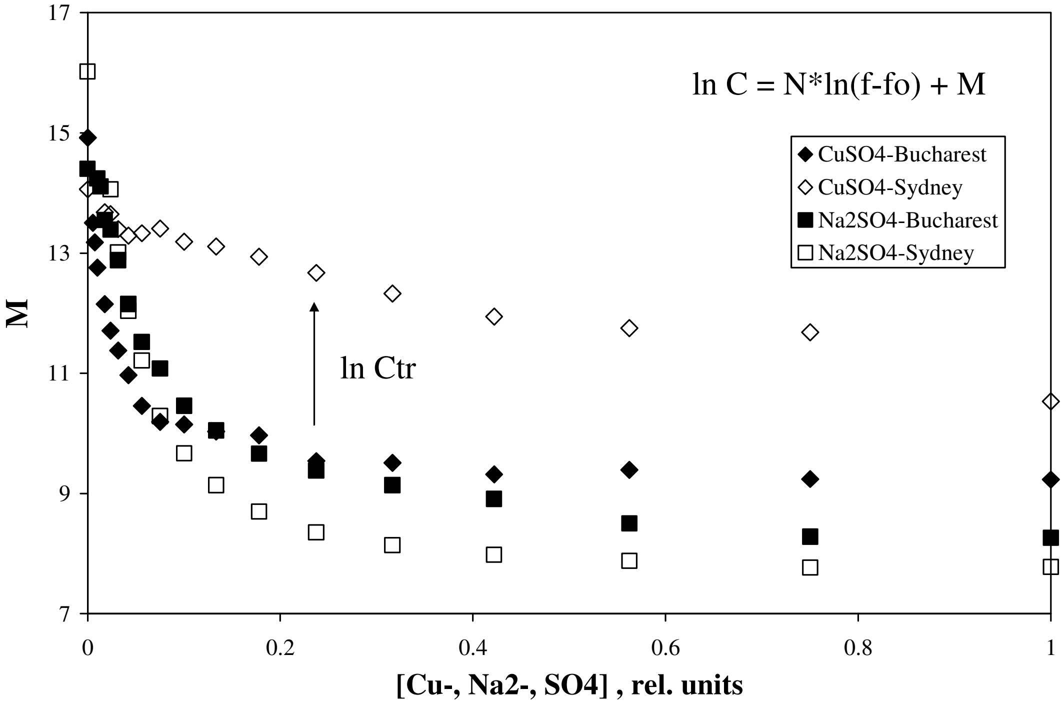


Figure 10.

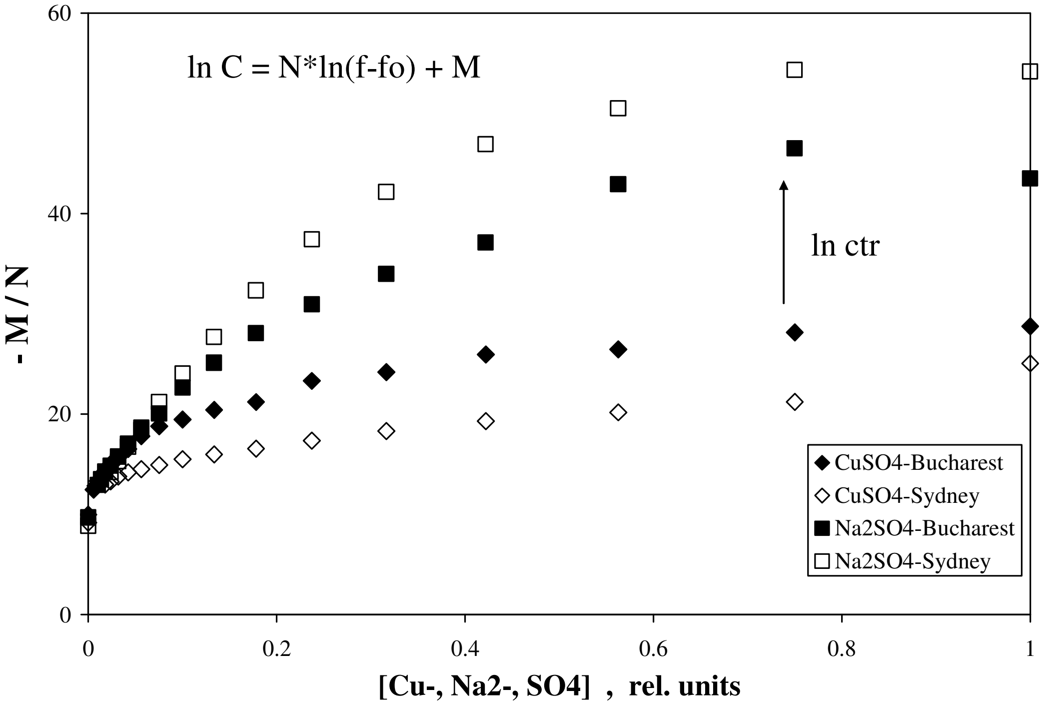
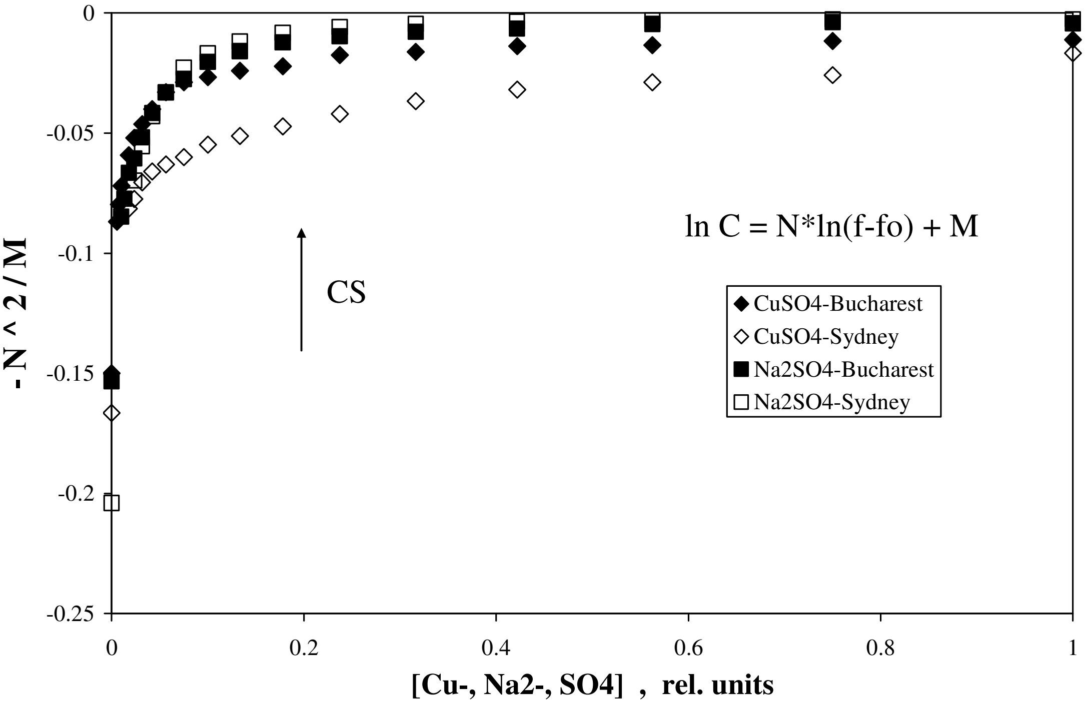


Figure 11.



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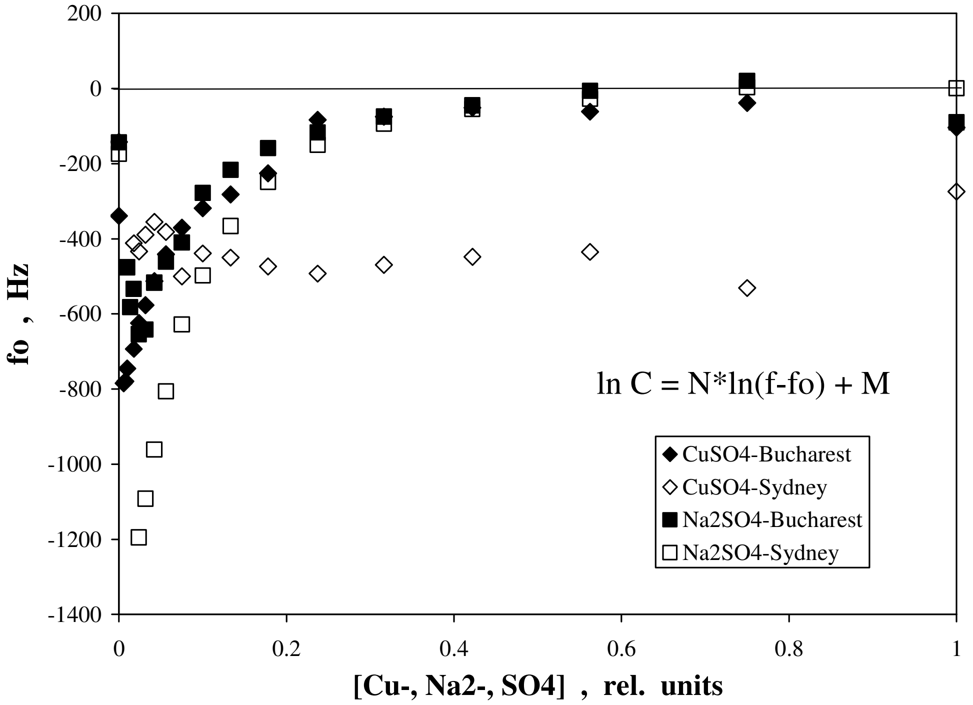

Figure 13.

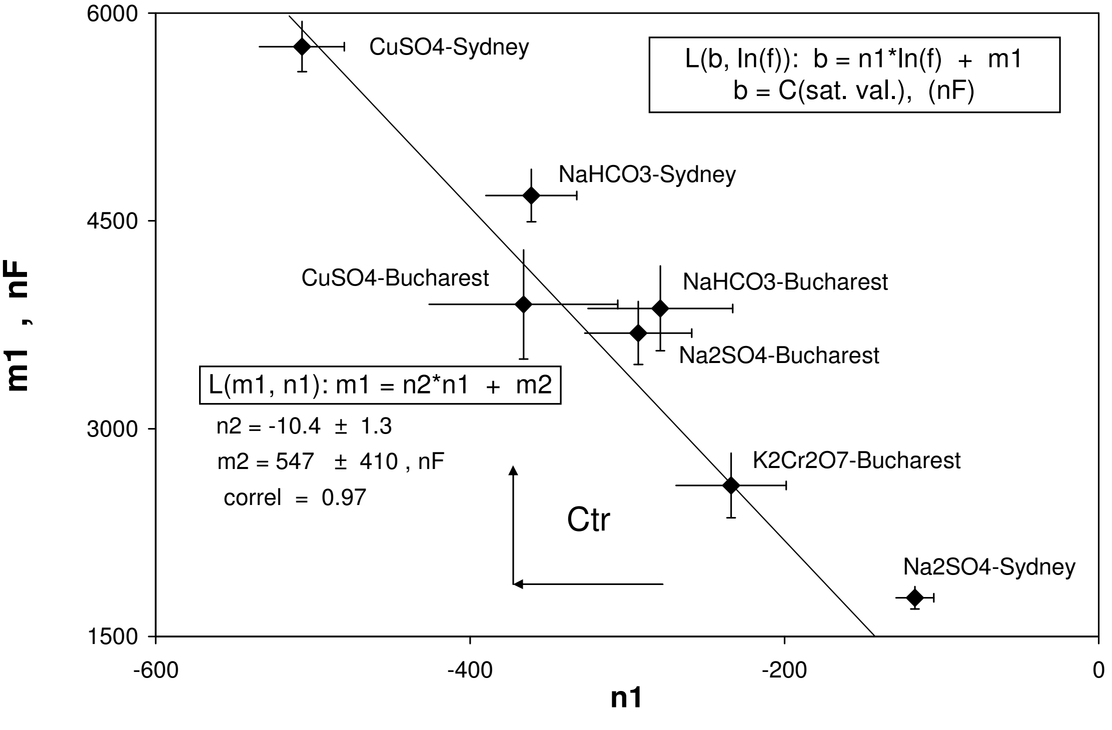

Figure 14.

Table 1. Pyramidal structure of R(f) and C(f) BED in Universal representation.

L(m1, n1)	n2	m2	u(n2)	u(m2)	correl
	1.13	13.5	0.19	1	0.86

L(m1, n1)	n2	m2	u(n2)	u(m2)	correl	n2	m2	u(n2)	u(m2)	correl
	0.929	11.99	0.15	0.88	0.94	0.966	13.04	0.33	1.7	0.8

L(M, N)	n1	m1	u(n1)	u(m1)	correl	n1	m1	u(n1)	u(m1)	correl
CuSO4-Bucharest	-3.98	8.321	0.27	0.2	0.96	-5.3	7.482	0.13	0.1	0.995
CuSo4-Sydney	-6.73	5.557	0.56	0.37	0.96	-3.2	10.14	0.44	0.25	0.89
Na2SO4-Bucharest	-6.33	6.237	0.41	0.32	0.97	-5.64	7.739	0.36	0.25	0.97
Na2SO4-Sydney	-5.75	7.328	0.29	0.26	0.98	-5.55	7.363	0.39	0.26	0.97
NaHCO3-Bucharest	-5.77	6.44	0.25	0.16	0.98	-5.17	8.658	0.37	0.41	0.96
NaHCO3-Sydney	-6.02	6.153	0.47	0.34	0.96	-4.9	9.096	0.59	0.6	0.91
K2Cr2O7-Bucharest	-4.76	7.351	0.39	0.21	0.94	-4.48	7.697	0.22	0.17	0.98
Universal representation	U(R, f): lnR = N*ln(f-fo) + M					U(C, f): lnC = N*ln(f-fo) + M				
Basic Experimental Data (BED)	θ = R (Ohm), U = f (Hz)					θ = C (nF), U = f (Hz)				

Table 2. Pyramidal structure in sigmoidal and semilogaritmic representations for $\theta = C$ (nF); $U = f$ (Hz).

L(m1, n1): m1 = n2*n1 + m2

n2	m2(nF)	u(n2)	u(m2)
-10.3	569	1.3	410

L(b, ln(f)): b = n1*ln (f) + m1

		n1	m1(nF)	u(n1)	u(m1)	m1/b(100Hz)
CuSO4	Bucharest	-366	3896	60	395	1.62
	Sydney	-507	5758	27	182	1.67
NaHCO3	Bucharest	-279	3868	46	306	1.48
	Sydney	-361	4683	29	190	1.55
Na2SO4	Bucharest	-293	3690	34	227	1.54
	Sydney	-117	1779	12	81	1.42
K2Cr2O7	Bucharest	-234	2590	35	233	1.63

S(C, x): C = (a+b*x)/(c+x) , x = [solute]

solute	ln(f), Hz	a	b (nF)	c
CuSO4	4.60517	61.21	2406	0.1638
Bucharest	4.787492	53.08	2046	0.1338
	6.907755	14.09	1168	0.11
	9.21034	3.611	631	0.228
CuSO4	4.60517	9.826	3440	0.0397
Sydney	4.787492	9.824	3379	0.0442
	6.907755	4.539	2135	0.0958
	9.21034	7.195	1150	0.519
NaHCO3	4.60517	38.19	2610	0.0617
Bucharest	4.787492	58.95	2608	0.0774
	6.907755	11.13	1732	0.0985
	9.21034	0.695	1395	0.846
NaHCO3	4.60517	0.6969	3027	0.0219
Sydney	4.787492	1.31	3011	0.0276
	6.907755	2.499	2062	0.0808
	9.21034	-9.516	1416	0.6119
Na2SO4	4.60517	6.0056	2389	0.01142
Bucharest	4.787492	6.4753	2321	0.01366
	6.907755	0.9284	1515	0.02094
	9.21034	-2.6737	1071	0.1173
Na2SO4	4.60517	2.0515	1254	0.005715
Sydney	4.787492	1.8986	1230	0.006147
	6.907755	0.2666	913	0.0108
	9.21034	-1.768	725	0.0867
K2Cr2O7	4.60517	21.07	1592	0.0542
Bucharest	4.787492	12.651	1436	0.0403
	6.907755	4.573	813	0.0768
	9.21034	0.648	483	0.312

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1998	2	1	Editorial: socio-psychological implications in creation and utilization of a databank (Ioan-Bradu Iamandescu); Behavior in vapor-liquid equilibria (VLE): I. Structural aspects; Behavior in vapor-liquid equilibria: II. Several structures in databanks; Symposium on VDC-4 held on 30 October 1997 at Lubrifin-SA, Brasov (Romania).	F
1998	2	2	Practical course of metrology (Romanian)	AFI
1998	2	3	DIFFUTOR-01: Thermally driven diffusion in pure metals	AFI
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