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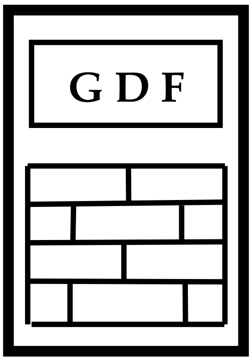

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Structural and relativistic aspects in transforming systems. I.Arrhenius and Universal representations of thermally driven processes.

Summary

Topoenergetic principles are reviewed together with some practical and well known cases of thermally driven processes for which Arrhenius and Universal representations are applied. The results obtained are discussed in view to reveal the general rules in processing experimental data and physical significances of topoenergetic parameters defining the nature and the amplitude of transforming processes.

An overview of topoenergetic principles

Topoenergetic principles were initiated with calorimetric experiments on specimens in which a wide variety of thermally transforming processes occur. The basic idea was that the measuring system including the tested specimen is an energy circuit and can be modeled by elementary components like electric circuits. This basic idea was taken from a working team trying to represent very complicated biological systems by space distribution of such elementary components [1]. Unfortunately their idea led to unsolved problems, so that my solution to remove space and including it in the elementary components has opened a new horizon both on measuring systems and tested specimens. Calorimetric systems were modeled successfully [2] and the immediate observation was about the composite nature of transforming specimens, i.e. they have at least two mutually interacting components: in general an inert one, Cin located in the laboratory time-reference system and a transforming component, Ctr, located in a separate time-reference system. An original kinetic equation was established based on these ideas and considering the transforming heat flow, wtr, obeying Arrhenius law. By submitting a stepwise perturbation on the tested specimen from an initial temperature Tin at which specimen is in equilibrium (no transforming process occurs) to a final temperature T at which the transforming process occurs, the two heat flows (win as immediate and wtr delayed by a specific time, tmax, at which the process has maximum rate value) the following equation exists [3]:

$$\ln (tmax*T) = - E / (R*T) + K \qquad [1],$$

where E is the activation energy, R the gas constant and K a parameter including some constants of the measuring systems and Cin [3,4]. For almost all transforming processes studied by different measuring systems according to these experimental conditions, this equation fairly agree proving the dissipative coupling between Cin and Ctr. The first main result obtained in applying this equation was in observing that E can be positive or negative and this is strongly connected with the relative sign of win and wtr.

However, there are some cases in which this coupling has an inductive element. Amorphous-crystalline coupling in chlorinated polyethylenes (CPE) was studied thoroughly and represents such a case [5-7].

We can observe that the stepwise experiment between the two values of temperature, Tin and T, contains a threshold value To at which transforming process is triggered. For all particular cases studied according to equation (1), another kinetic equation exists:

$$\ln(t\max) = N^* \ln | T - To | + M \qquad (2)$$

where parameters (N, M, To) defines the nature and the amplitude of the transforming process. This kinetic equation was proven to have a Universal character being available for any measuring system and transforming process driven by a general potential, U, in standard experimental conditions imposed by topoenergetic principles [8, 9].

One of the most important experimental conditions for which these topoenergetic equations exist refers to standard volume of tested specimens. So, for a series of tested specimens performing the same transforming process (i.e. the same nature), but having different amplitudes (i.e. different values of Cin and Ctr) further equations exist:

$$\text{Arrhenius representation: } K = n1^*E + m1 \qquad (3)$$

$$\text{Universal representation: } M = n1^*N + m1 \qquad (4).$$

In these conditions, parameters (E, K) and (N, M, Uo) define the behavior of a tested sample with particular values of Cin and Ctr or so called ontogeny of this sample. Parameters (n1, m1) define the group behavior of samples having the same nature of transforming process, but different values of Cin and Ctr, or the phylogeny of these samples. By considering more series of samples related by different other characteristics, it is possible to define higher phylogenies, so that starting from basic experimental data up to highest phylogeny the topoenergetic structure of all these data are structured in pyramidal shape. Universal representation has additional advantages relative to Arrhenius one (see above mentioned citations for more details):

- it can be applied to any transforming process driven by a general potential, U and by considering different values (eigenvalues, θ) of a response function defined in the measuring system; it defines new important characteristics:

- the threshold parameter, Uo;
- the process amplitude, M ~ |ln Ctr|;
- the value of kinetic unit, -M/N ~ |ln ctr|;
- the value of coupling strength between Cin and Ctr, -N^2/M ~ |CS|.

Some more advantages will be revealed in the next analysis of particular cases.

Review of some particular cases

The main advantage of thermally driven processes is that it is possible to imagine calorimetric model evidencing the exact nature and structural origin of win and wtr. This was the reason to introduce the notion of process polarity directly connected with the endothermal or exothermal sign of these energy flows [10].

Table 1 gathers a series of thermally driven processes thoroughly studied according to topoenergetic principles. Only several cases are reviewed in the following with the aim to evidence and to fix topoenergetic principles.

Crystallization from melt

Crystallization from melt of different polyethylenes (PE) was the first transforming process studied according to these new principles [3-7]. Additionally, some of experimental data from literature are processed according to the above Arrhenius and Universal representations [8, 11].

In Figures 1 and 2 show Arrhenus and Universal representations of experimental data obtained in differential calorimetric system for the three high density PE (HDPE) (Hizex, Mitsui-Co, Japan) studied in the series of studies on amorphous-crystalline coupling [3].The PE powders are analyzed as such (dry, D) and in mixture with silicon oil (wet, W).It is interesting to observe that the process amplitude separates in the same order the samples and silicon oil increases process amplitude in both representations.

In Figures 3-8 the crystallization from melt of a series of HDPE fractions by using a dilatometric measuring system is presented for which tmax corresponds to half time of conversion function.

Crystallization from melt results with different polarity signs in the two representations due by the only fact that wtr and win are exothermal relative to the other series of transforming processes where these are endothermal and polarity results to be positive in both representations (Table 1).Crystallization from melt can be considered from structural point of view as a polymerization process. However, the proper polymerization-curing processes occur by rising stepwise temperature perturbation where win = ENDO and wtr = EXO [23]. We may observe first that:

crystallization from melt: To = melting point (Tm); (5)

polymerization-curing: To = freezing/glass transition point of kinetic entity (Tg).

On the other hand, melting-crystallization processes are reversible, while polymerization processes are not. Unfortunately, there are not available similar experimental data on melting processes yet in view to complete this topoenergetic view. However, approximate experiments were made with small increasing temperature steps (successive steps of 5 ^{0}C) up to and over Tm by differential calorimetry [24]. Figures 9 and 10.present three samples originating from the same LPE-1 (D means dry powder like in Figures 1 and 2). D-CPE*-1 (chlorinated LPE-1 in aqueous suspension with approx. 40% wt Cl) is the genuine sample tested (so called at first calorimetric round), while D-CPE-1 is the same test specimen tested second time (at second round).For each tested sample three eigenvalues are considered proportional with heat capacity, denoted as Ca, Ch and Cah.It results that Universal representation appears to be more sensitive than Arrhenius one to structural and kinetic differences between samples. On the other hand, in this measuring system partial annealing of samples involves some additional processes based on defect precipitation and lamellar thickening and/or breaking (see bellow).
The difference between crystallization from melt and melting processes arises in the fact that the stepwise temperature perturbation is applied in opposite directions and this appears like the time “flows” in opposite senses, so that:

crystallization from melt, To = Tm , (win, wtr) = EXO, win*wtr > 0 (6)
melting of crystalline phase, To = Tm, (win, wtr) = ENDO, win*wtr > 0.

Another important fact is that amorphous and crystalline phases coexist separately even in molten state (see the Hosemann’s team studies on paracrystalline structures [25]).

Defect precipitation in CPE samples

Annealing of genuine CPE samples at temperatures close and bellow to Tm appears in isothermal calorimetric system only by win (called as “IN VIVO” measuring system) [5, 7]. However, these annealed samples show dramatic changes in crystalline structure in differential scanning calorimeter (DSC called as “IN VITRO” measuring system). More exact, two simultaneous processes occur by coherent precipitation of chlorinated defects: (i) the segregation of crystalline lamellae and (ii) the formation of inter-lamellar structure. The melting endotherm splits in two peaks, Tm1 and Tm2 > Tm1, corresponding to the melting of inter-lamellar order and of lamellar fragments, respectively [5, 7]. Amplitudes of the two processes IN VIVO measuring system are in reverse relationship, i.e. are simultaneous, equal and of opposite signs (Table 2):

$$wtr(Tm1)(EXO) = - wtr(Tm2)(ENDO) \qquad (7).$$

so that they annihilate each other. They can be measured only IN VITRO measuring system by different eigenvalues (Table 2, [7]).

From structural point of view the thoroughly studies established that these two transforming components are connected by an inductive element [5, 6].

Similar process with wtr(Tm1)(EXO) appears in amorphous materials and revealed also by IN VITRO measuring systems (DSC, impact test, dilatometry, etc.) [18].

Adsorption of gas component on stationary phase

Gas chromatography was compared with differential thermal analysis, so that the retention time, tR, corresponds with tmax. The process of retention of a gas component is wtr(EXO) while win(ENDO) represents its heating from room temperature to the column one.

Figures 11 – 18 show the Arrhenius and Universal representations on gas chromatography of several n-alkanes and n-alcohols. The experimental conditions are commonly used as routine procedure in any gas chromatography laboratory: 140-190 ^{0}C, chromosorb S as stationary phase, nitrogen as carrier gas and the same conditions for all components [17].

Thermally flow of Newtonian liquids

Viscosity represents an important open problem of actual science and technology because measuring systems used for viscosity are far from its basic definition.

In view to evidence topoenergetic aspects of thermally flow of Newtonian liquids as the simplest flow process, we can imagine a calorimetric cell with a frozen liquid at Tin on the surface which is placed a metallic ball. This cell is transferred in an isothermal differential calorimeter at a temperature T at which the liquid melts and the metallic ball begins to fall and helps us to evaluate the liquid viscosity. It simply results that win(ENDO) and wtr(EXO) ~ dynamic viscosity (DV).

Figures 19 – 21 show the Arrhenius and Universal representations for a series of standard liquids thoroughly studied in the German Institute of Metrology (Physikalisch-Technische Bundesanstalt) [20].

In conclusion, Tables 3 and 4 give the general relationships between the ontogenic and first phylogenic parameters in the Arrhenius and Universal representations. Universal representation was successfully applied to many other transforming processes driven by a wide variety of potentials and measuring systems.

Table 1. Arrhenius and Universal representations of some thermally driven transforming processes.

transforming process	measuring system, eigenvalue (θ)	win	wtr	ARRHENIUS					UNIVERSAL				
				win*wtr	E	n1	E*n1	P	N	n1	N*n1	M	P
PE melt crystallization [3, 11]	differential calorimetry, dilatometry , θ = tmax	EXO	EXO	+	+ ln Ctr	+	+	+	–	–	+	ln Ctr	–
PE melting [24]	differential calorimetry, θ = heat capacity	ENDO	ENDO										
diffusion in solid metals DIFFUTOR® [12, 13]	θ = diffusion coefficient	ENDO	ENDO	+	+ ln Ctr	+	+	+	+	–	–	-ln Ctr	+
Vapor-Liquid Equilibria [14] VAPORSAT® [15]	θ = equilibrium pressure												
annealing of PES fibers [16]	dilatometry under constant stress, θ = relative shrinkage												
adsorption of gas component on stationary phase [17]	gas chromatography θ = retention time, tR	ENDO	EXO	–	– -ln Ctr	+	–	–	–	–	+	ln Ctr	–
structuring in amorphous polymers [18]	impact resistance, shrinkage θ = tmax												
defect precipitation by annealing in crystalline phase (chlorinated PE) [7]	differential calorimetry θ = splitting coefficient of melting endotherm												
quenching of atomic & molecular excited species in rarefied gases [19]	luminescence decay of activated species, θ = 1/(deactivation rate)												
flow of Newtonian liquids [20]	dynamic viscosity, θ =DV												
electric conductance of NTC thermistors [21]	θ = electric resistance												
oxidation of amorphous phase in PE [4, 22]	differential calorimetry θ = tmax												
curing-polymerization (epoxy+amine [23])													

Table 2. Representations of the two concurrent processes occurred in CPE samples by annealing and revealed by differential scanning calorimetry as after effects (so called “IN VITRO” measuring system [5,7]).

IN VITRO endotherms	process significance	eigenvalue*) θ	win	wtr	win*wtr	ARRHENIUS				UNIVERSAL				
						E	n1	E*n1	P	N	n1	N*n1	M	P
Tm2	order-disorder (melting endotherm) of lamellar fragments remained after coherent precipitation of defects	α = h2/(h1+h2)	ENDO	ENDO	+	+ ln Ctr	+	+	+	+	–	–	-ln Ctr	+
Tm1	order-disorder (melting endotherm) of interlamellar order created by coherent precipitation of defects	1- α = h1/(h1+h2)	ENDO	EXO	–	– -ln Ctr	+	–	–	–	–	+	ln Ctr	–

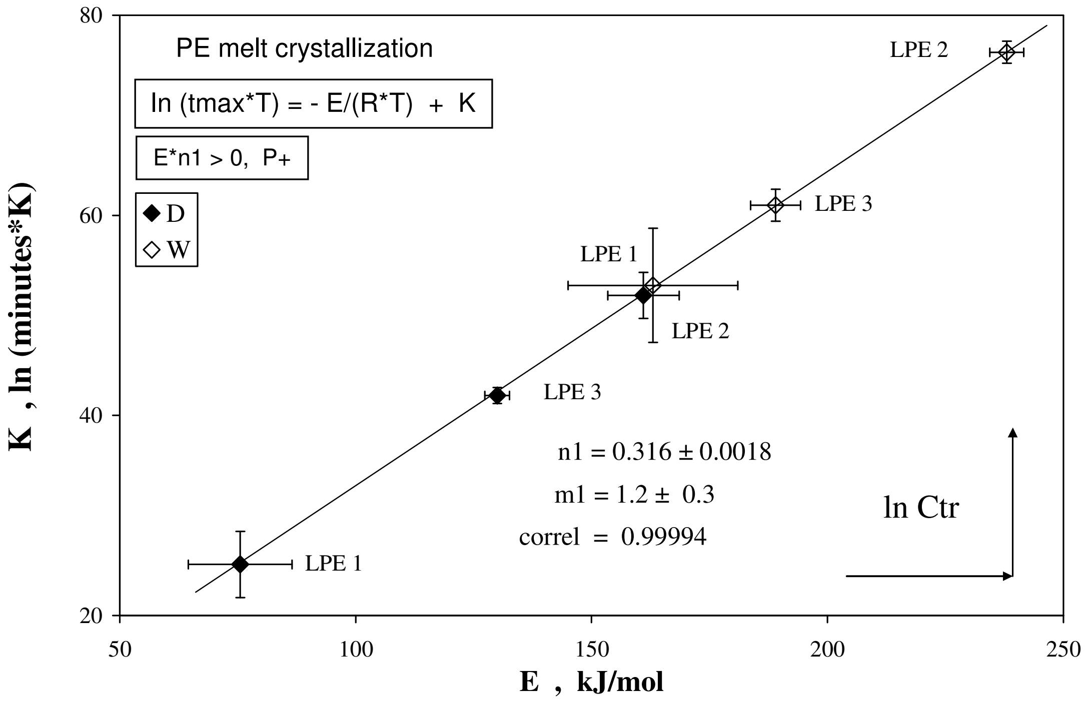
*) h1, h2 are the peak heights of Tm1 and Tm2, respectively.

Table 3. ARRHENIUS representation (K = n1*E + m1).

win*wtr	E*n1	P	E
+	+	+	ln Ctr
–	–	–	- ln Ctr

Table 4. UNIVERSAL representation (M = n1*N + M).

N*n1	N	P	M	-M/N	-N^2/M
+	-ln Ctr	–	ln Ctr	ln ctr	CS
–	ln Ctr	+	-ln Ctr	-ln ctr	-CS



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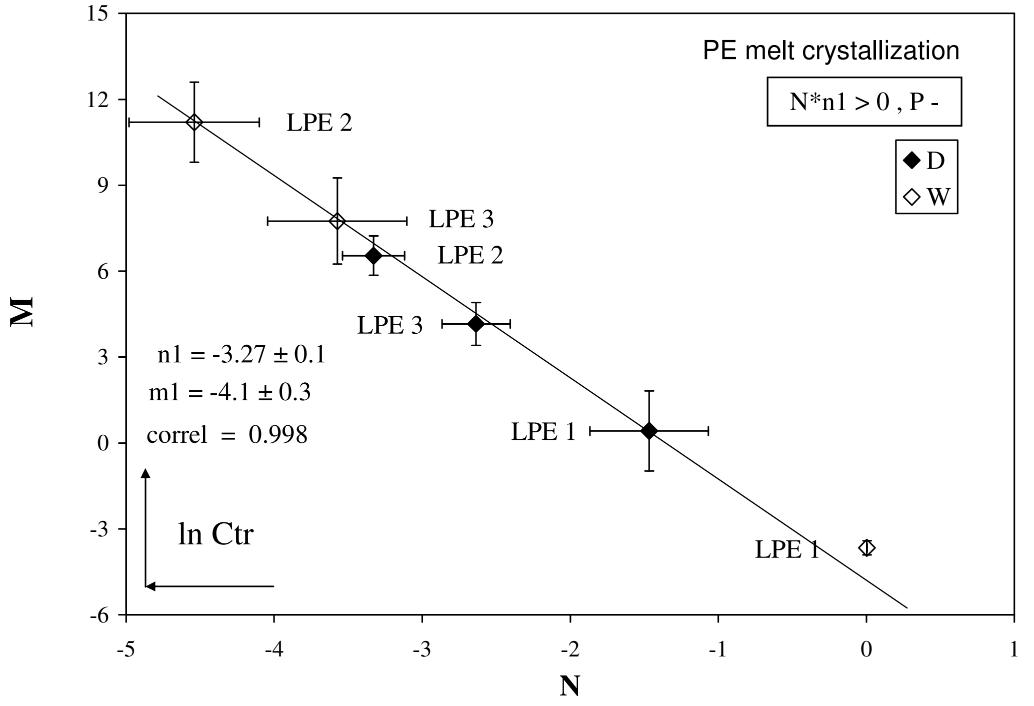

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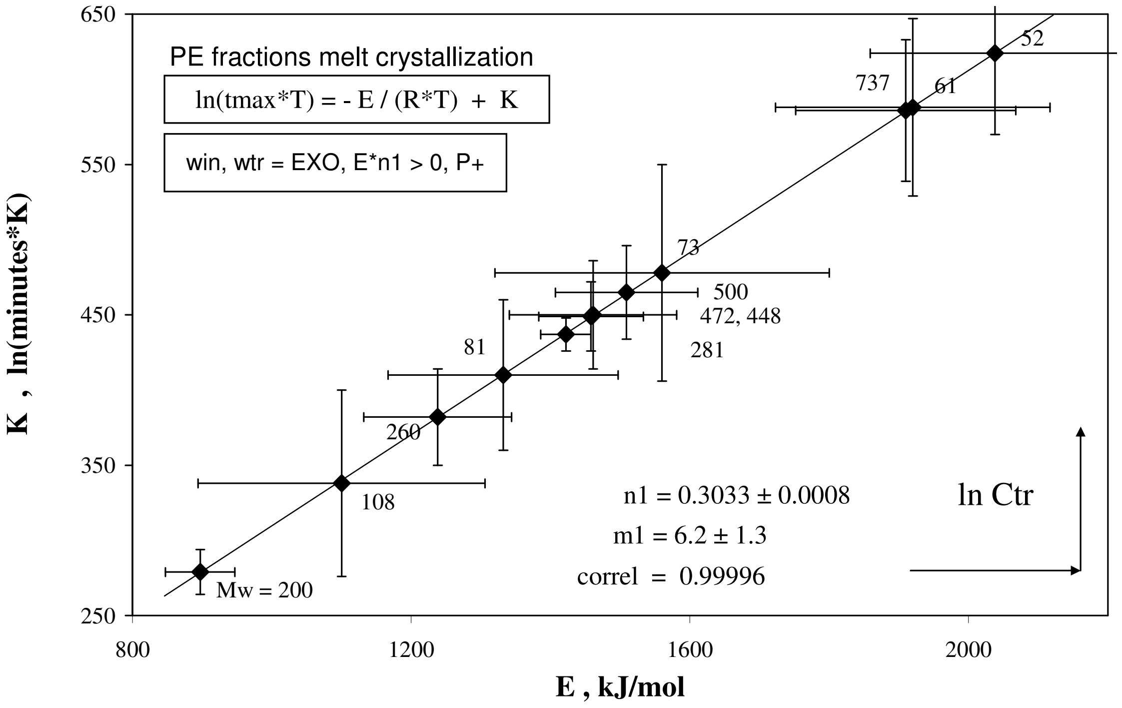

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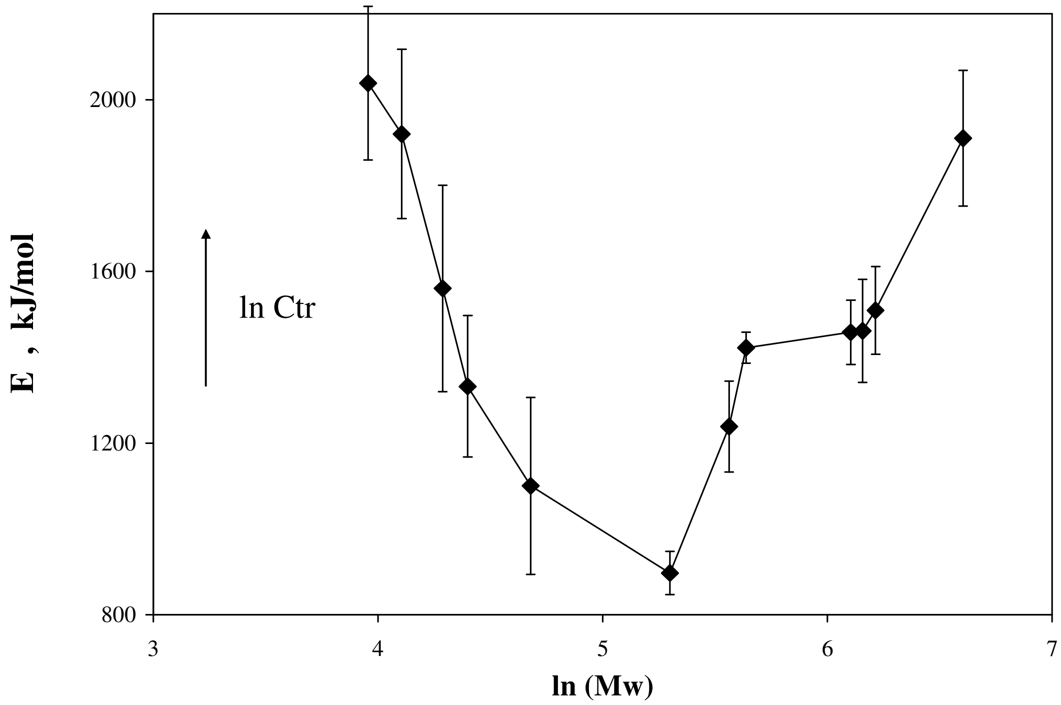

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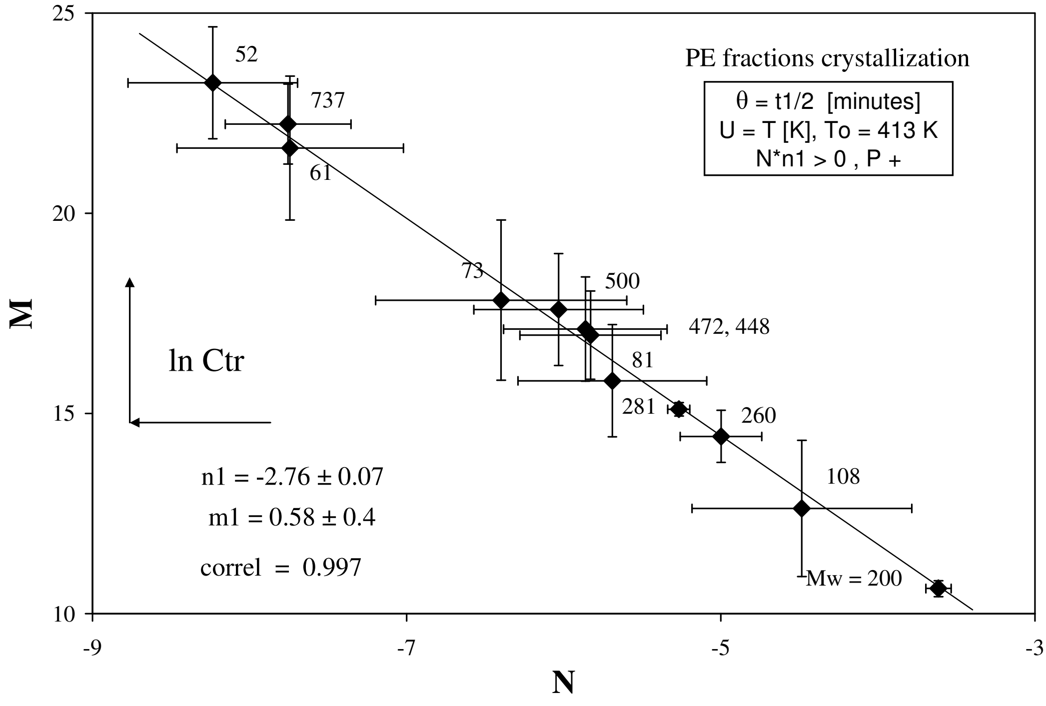

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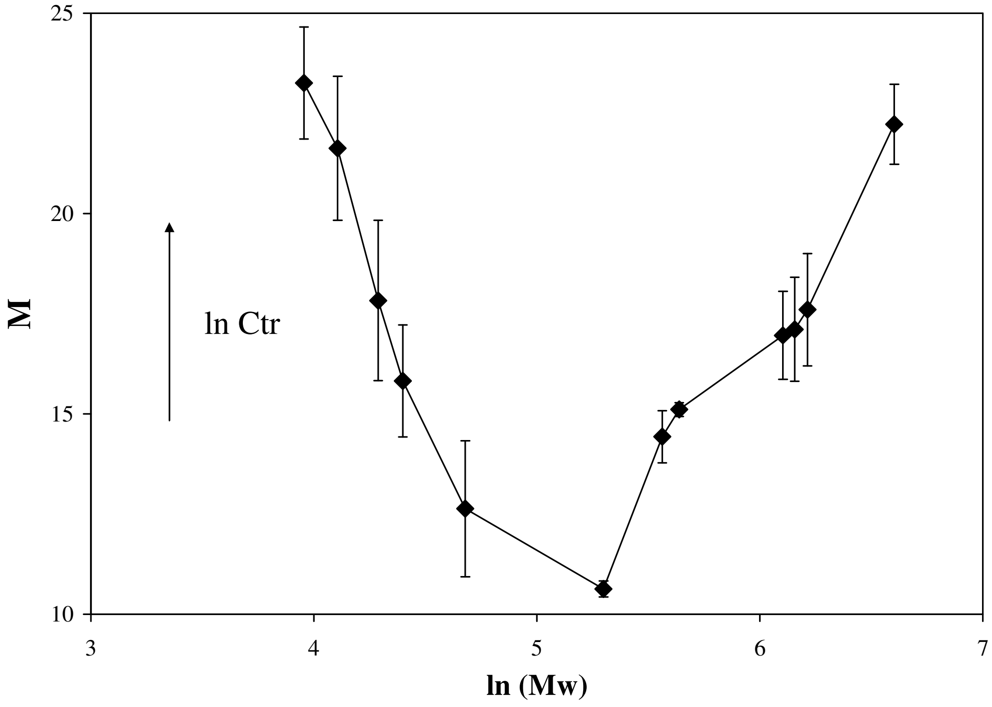

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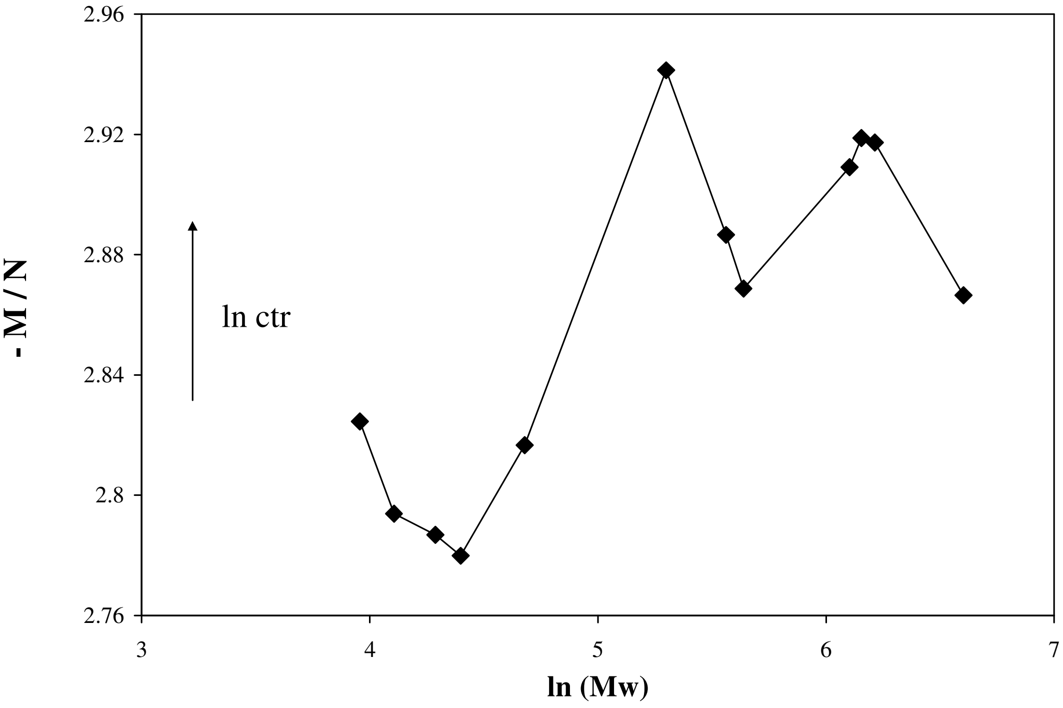

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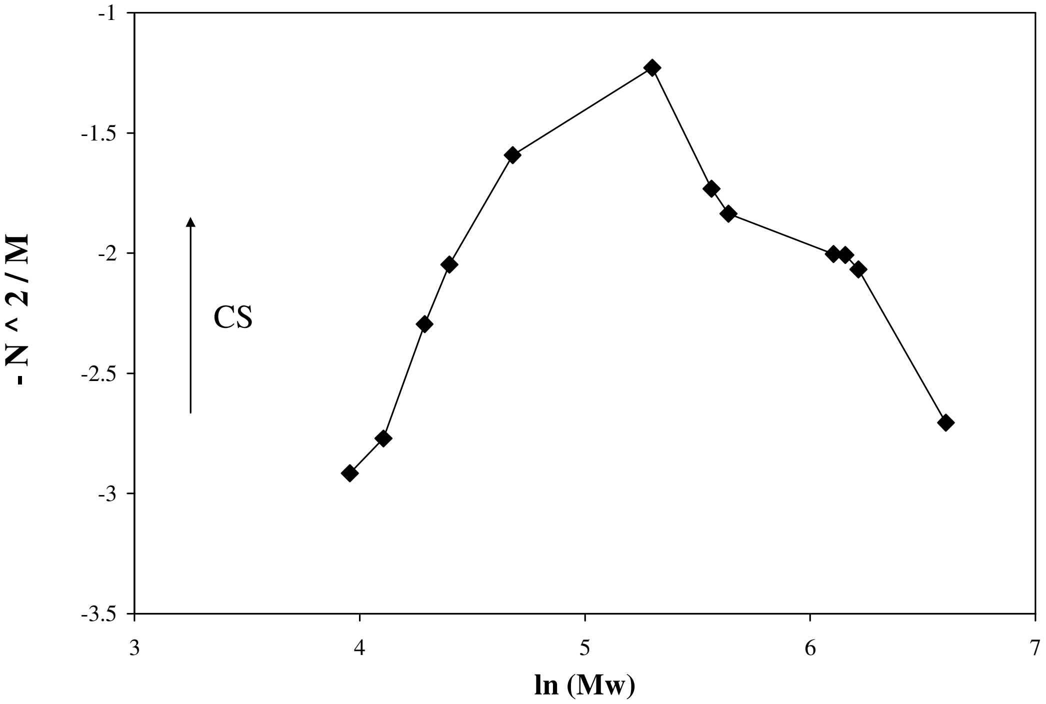

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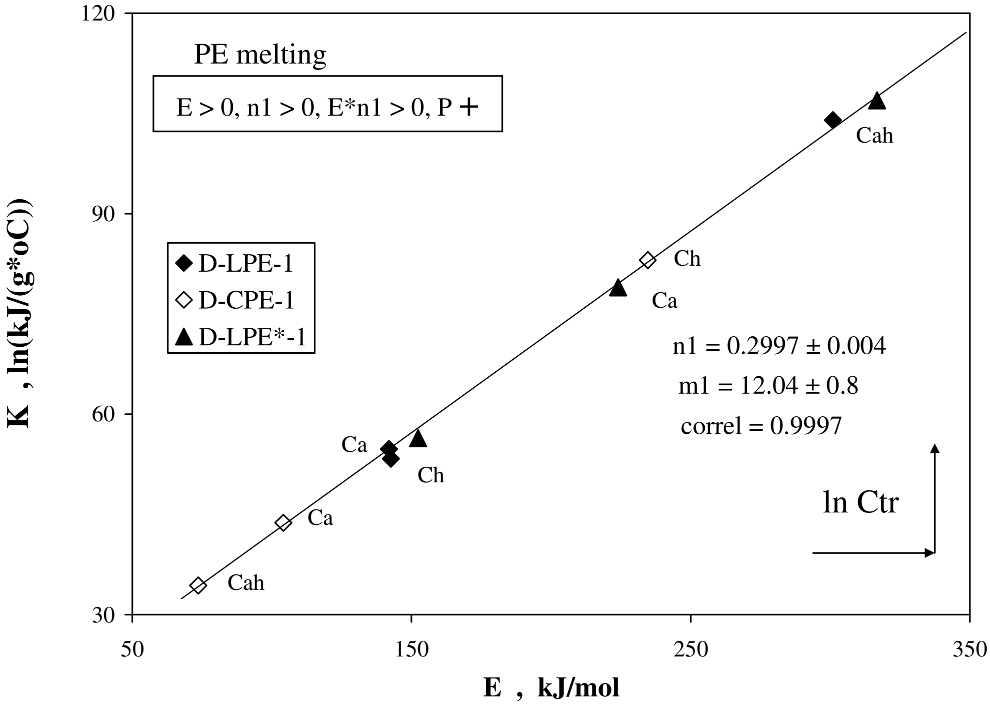

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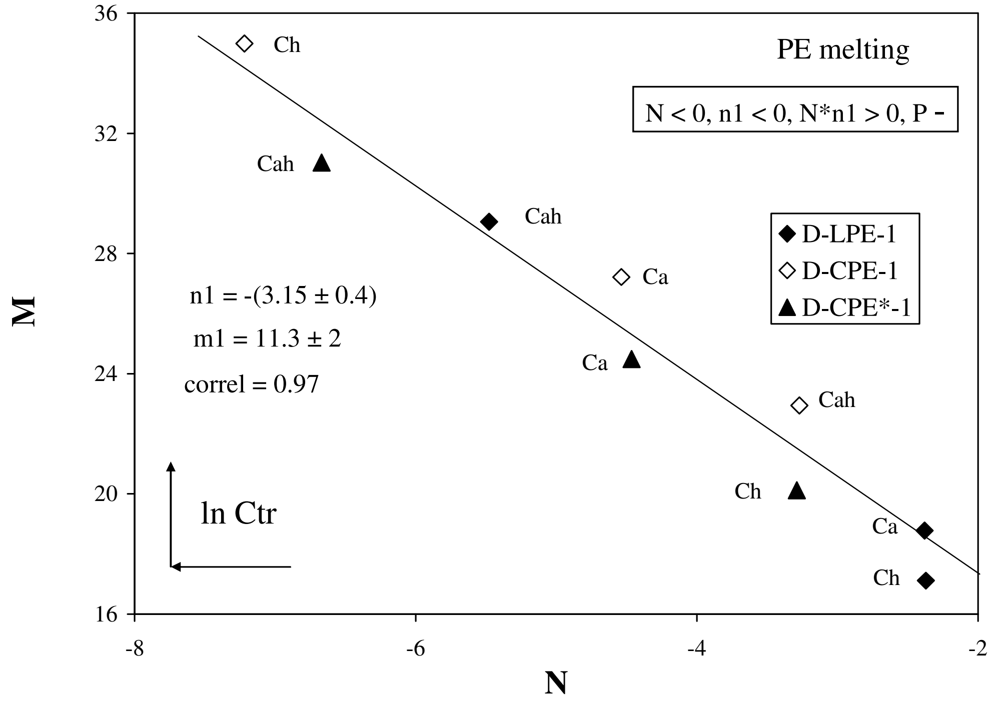

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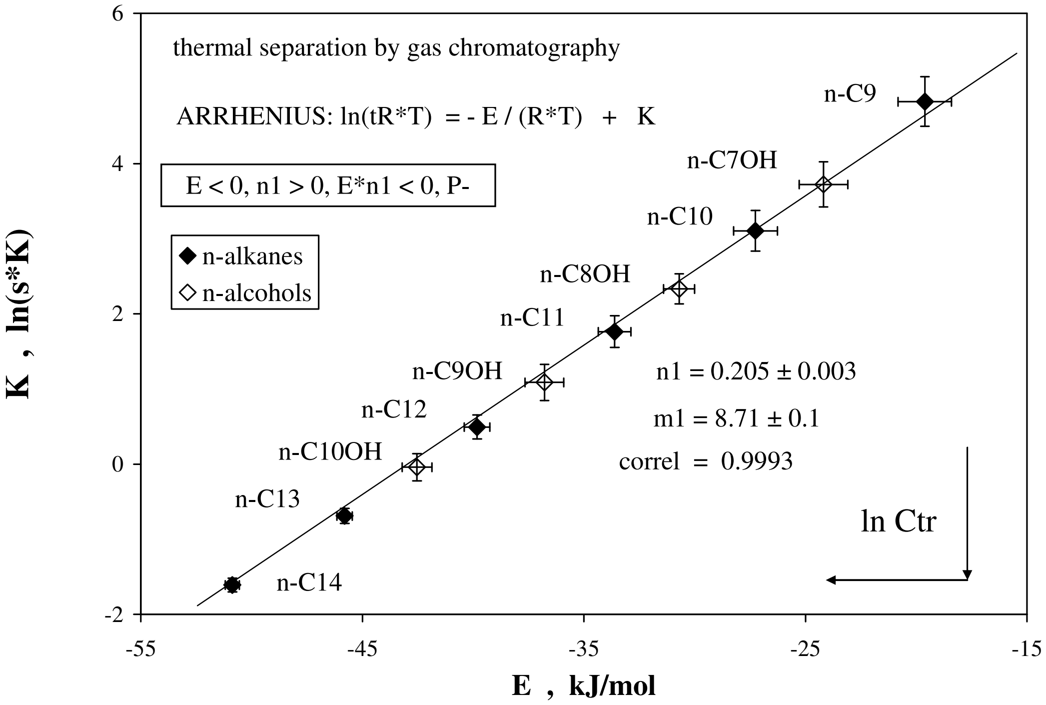

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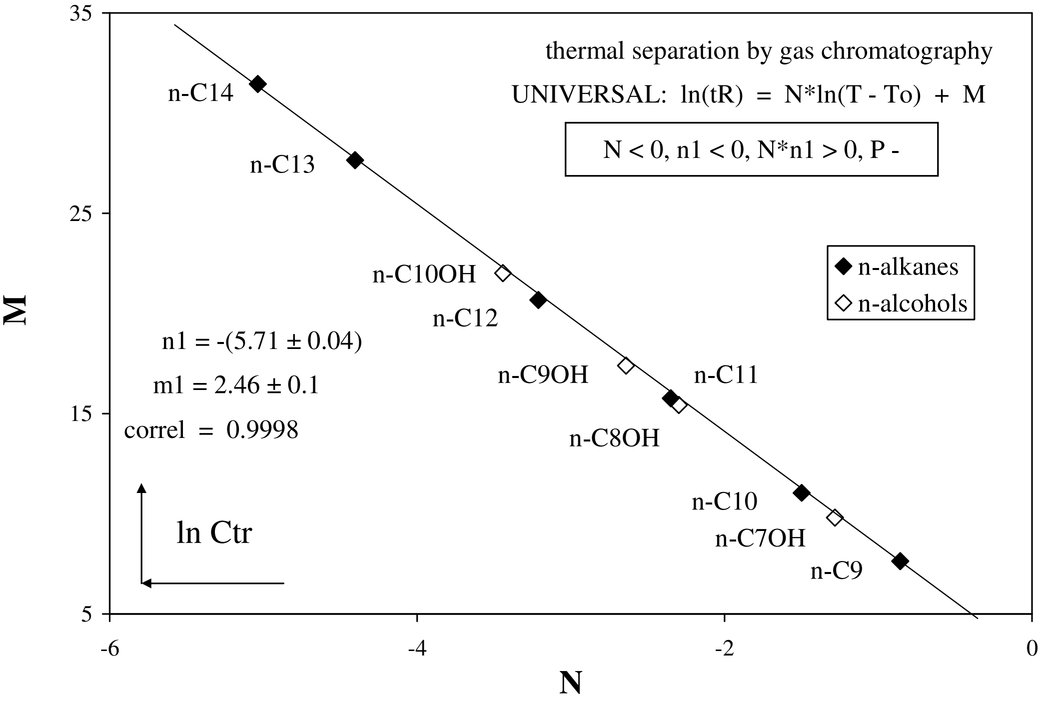

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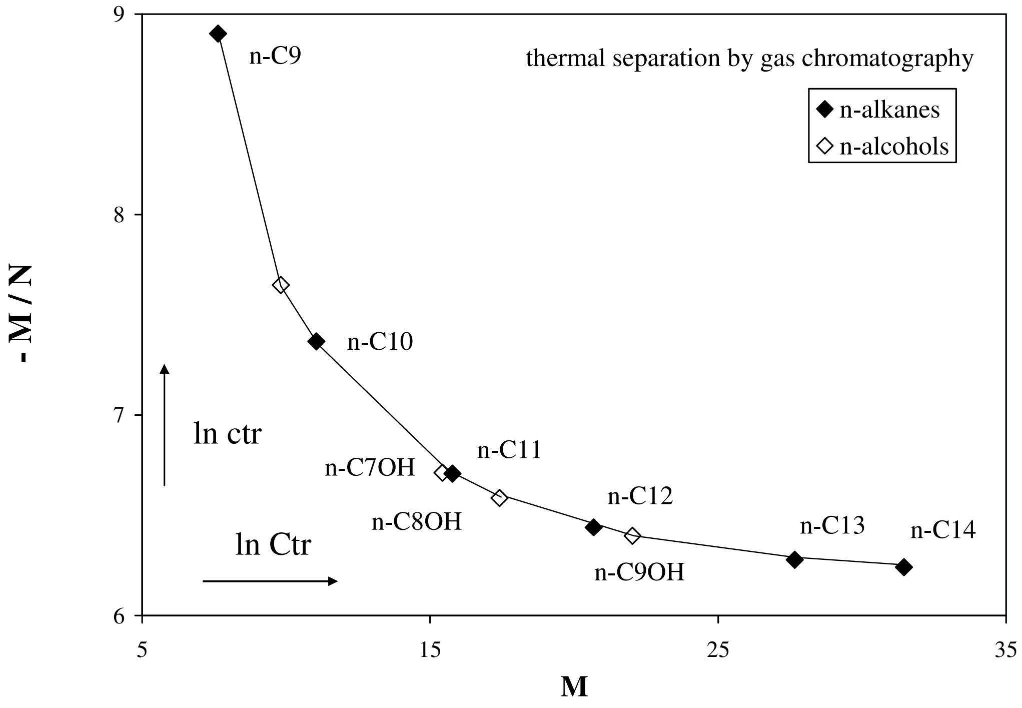

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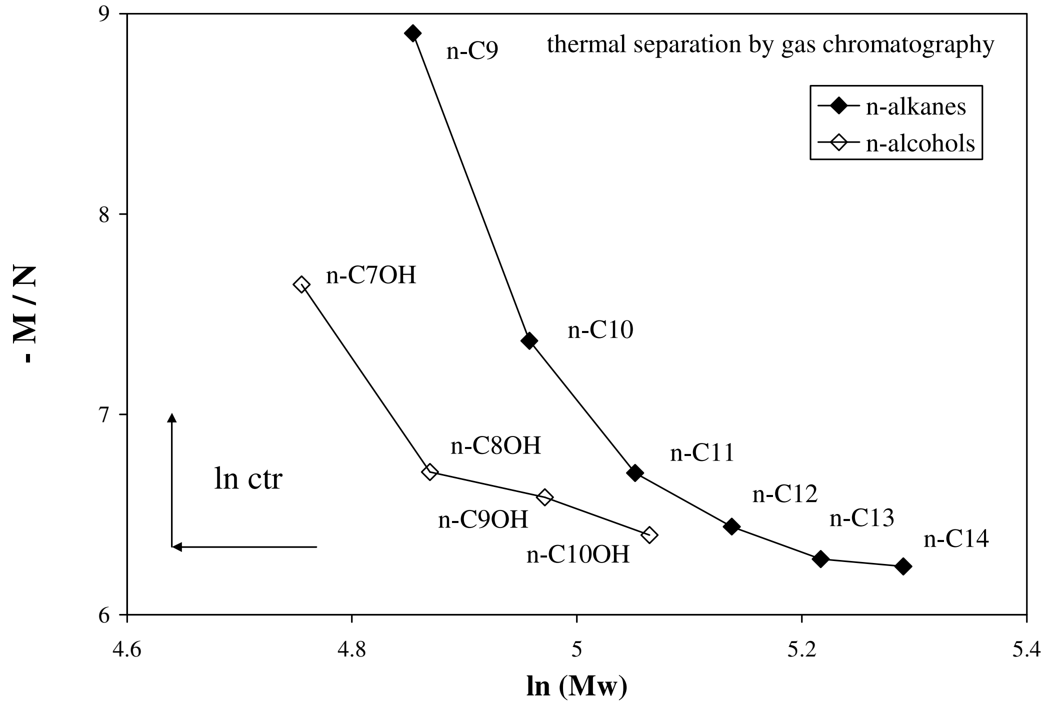

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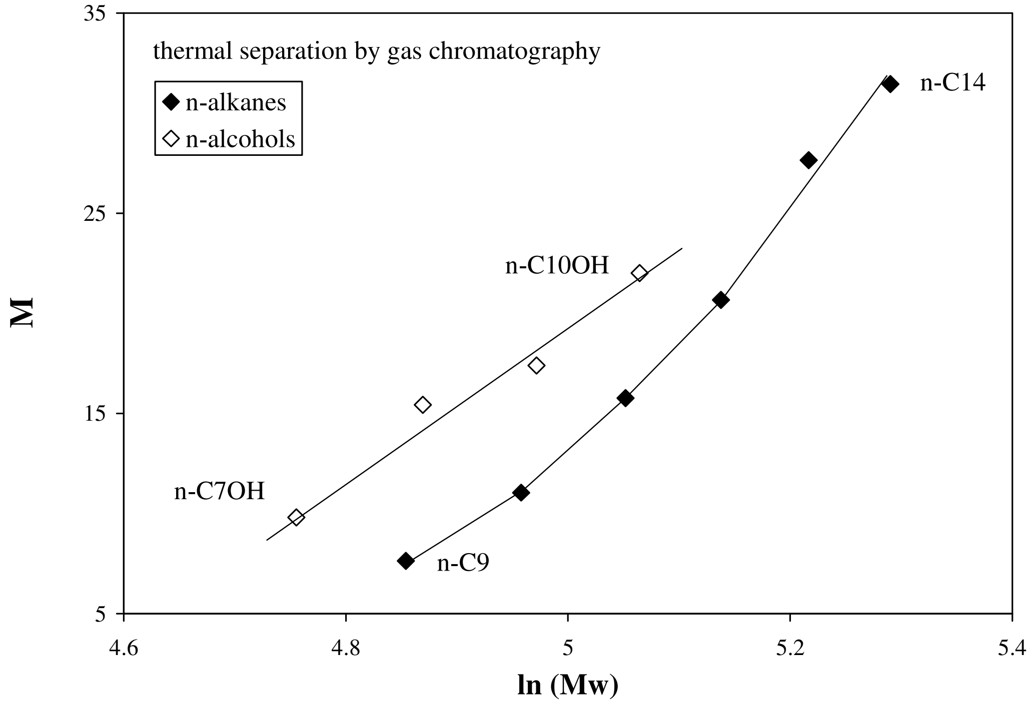

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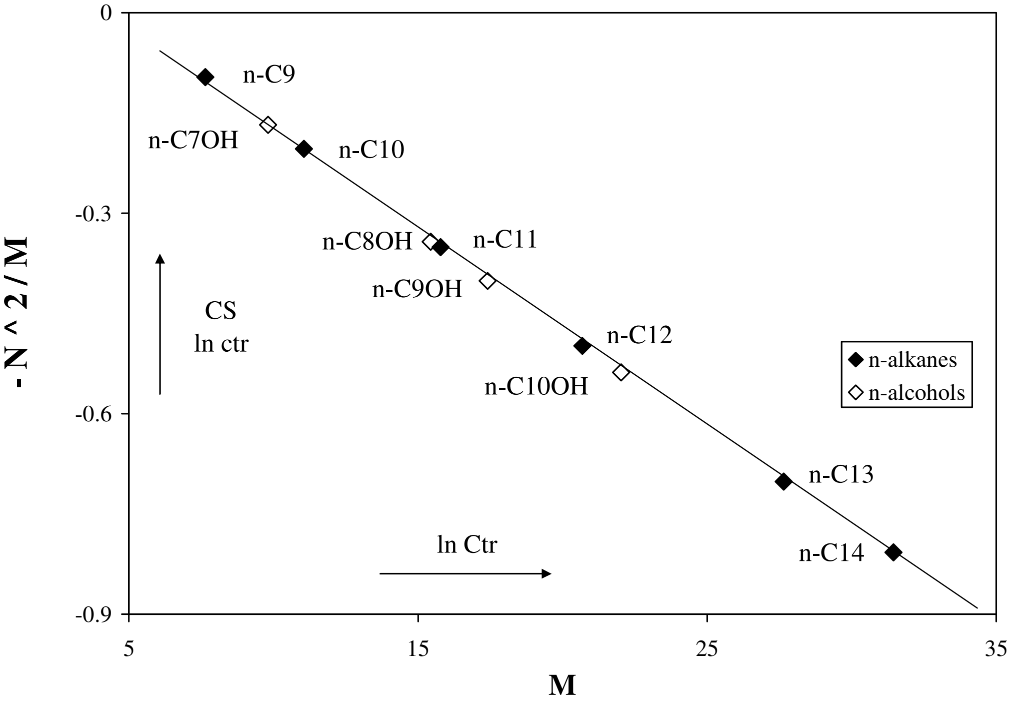

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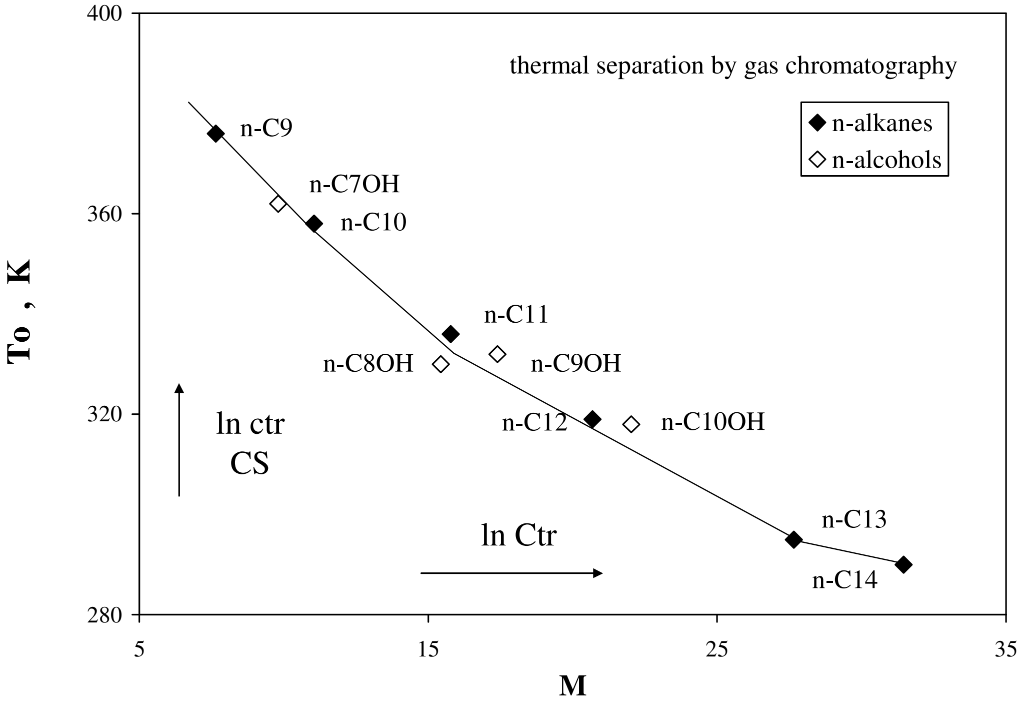

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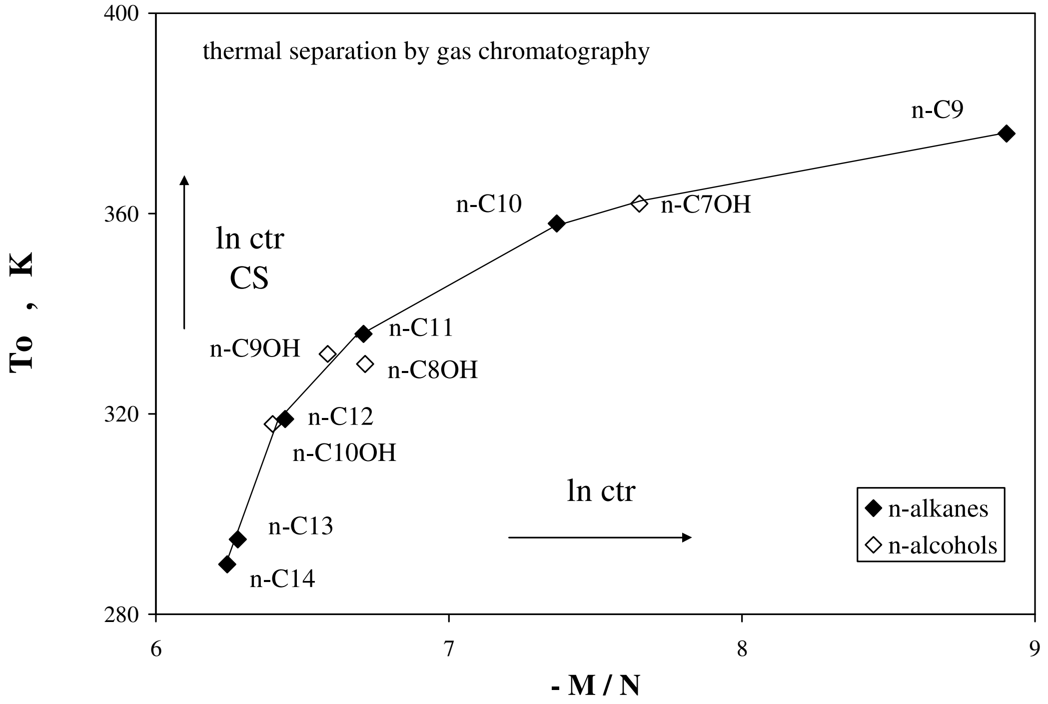

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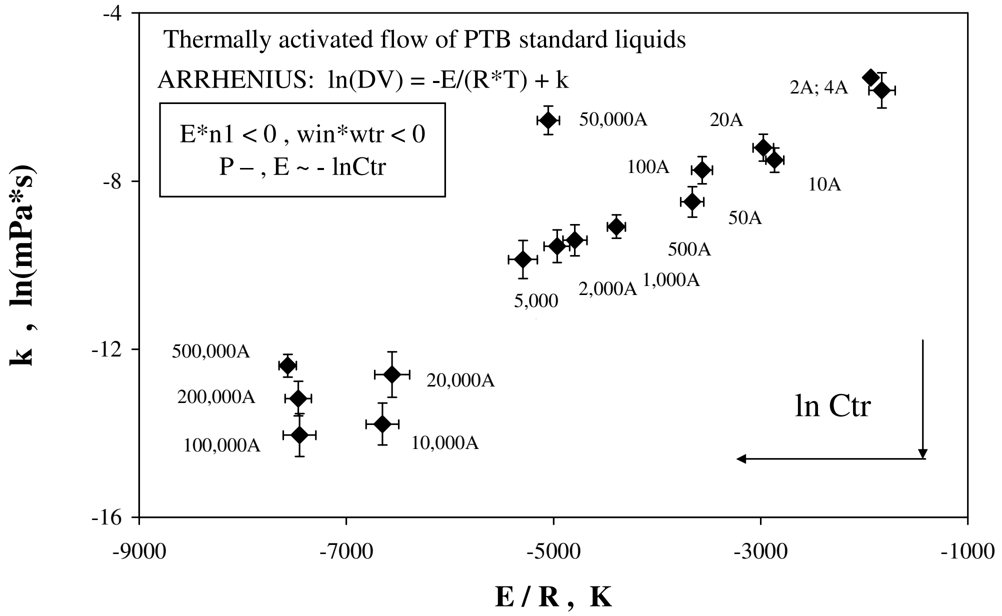

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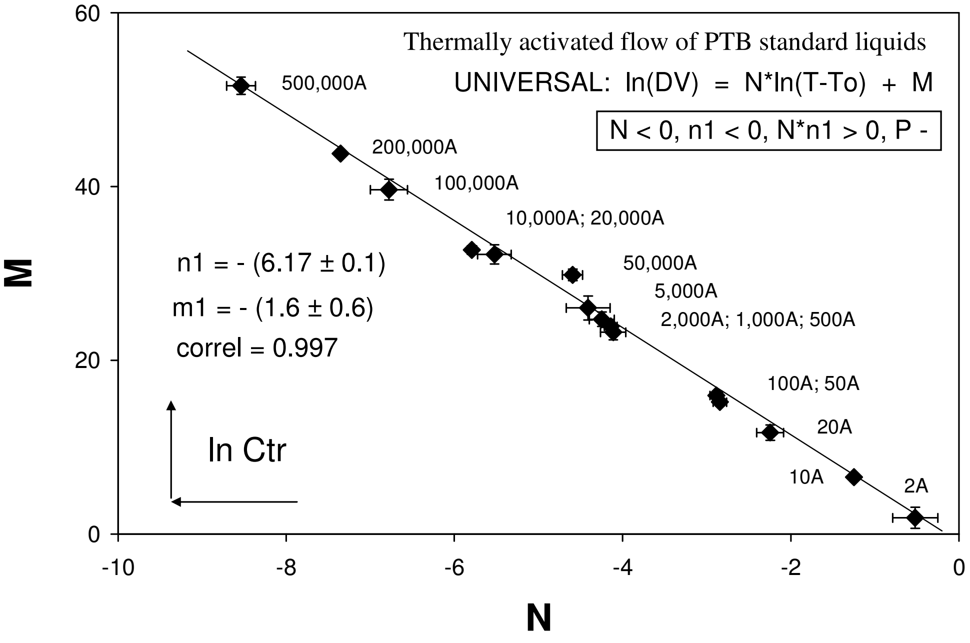

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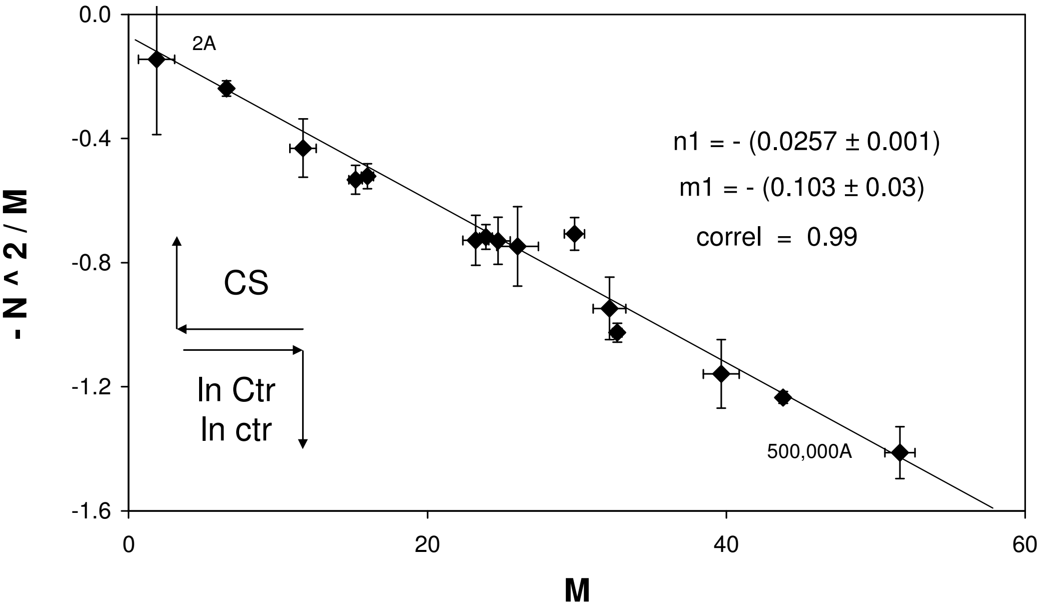

Figure 21.

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1997	1	2	Guide of good practice in metrology (Romanian)	AFI
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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1999	3	2	Discussions on Applied Metrology	AFI
2000	4	1	Editorial: Laboratory accreditation and inter-laboratory comparisons (Virgil Badescu) Doctoral Theses – important data banks. GDF intends to open new series of experiments on thermo-physical properties. Some comments on uncertainty: global budget and DFT analysis. Events: The 9th International Metrology Congress, Bordeaux, France, 18-21 October 1999.	F
2000	4	2	Measurement and Calibration.	AFI
2001	5	1	Editorial: Metrology ensures moral and technological progress. Topoenergetic aspects of amorphous-crystalline coupling. I. Composite behavior of water and aqueous solutions (paper presented at nanotubes and Nanostructures 2001, LNF, Frascati, Rome Italy, 17-27 October 2001). Events: Nanotubes and nanostructures 2000.School and workshop, 24 September – 4 October 2000, Cagliari, Italy.	F
2001	5	2	Editorial: Viscosity – a symptomatic problem of actual metrology. Visco-Dens Calorimeter: general features on density and viscosity measurements. New vision on the calibration of thermometers: ISOCALT® MOSATOR: Topoenergetic databanks on molten salts properties driven by temperature and composition.	F

continued

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2002	6	1	MOSATOR-01: Topoenergetic databanks for one component molten salts; thermally driven viscosity and electrical conductance.	AFI
2002	6	2	Editorial: HuPoTest - Operator calibration or temporal scale psychic test. MOSATOR: topoenergetic databanks of one component molten salts; thermally driven viscosity and electrical conductance.	F
2002	6	3	Editorial: Quo vadis Earth experiment? ISOCALT® : Report on metrological tests	F
2003	7	1	Editorial: Time – an instrument of the selfish thinking. 1st NOTE: Homoeopathy: upon some efficient physical tests revealing structural modifications of water and aqueous solutions. I. Mixing experiments.	F
2004	8	1	Metrological verification and calibration of thermometers using thermostats type ISOCALT® 21/70/2. Metrological verification and calibration of thermometers using thermostats type ISOCALT® 2.2R.	F
2004	8	2	Aspects of correct measurements of temperature. I. measurement of a fixed point according to ITS-90. Physics and Homoeopathy: some physical requirements for homoeopathic practice.(Plenary lecture at the 19th SRH National Congress, 21-22 September 2004, Bucharest, Romania)	F
2005	9	1	AWARD for ISOCALT® at the International Fair TIB-2004, October 2004, Bucharest. ISOCALT® 3/70/21 was awarded in a selection of 20 products by a commission of experts from the Polytechnic University of Bucharest. Upon some aspects of temperature measurements. (12th International Metrology Congress, 20-23 June 2005, Lyon, France)	F
2005	9	2	A new technique for temperature measurement and calibration. National Society of Measurements (NSM). Important warning for T-calibrator users: MSA has chose metrology well calibrators from Fluke (Hart Scientific).	F
2005	9	3	Universal representation of Cancer Diseases. 1. First sight on NSW-2003 report. Universal representation of Cancer Diseases. 2. UK cancer registrations on 1999-2002. Vital Potential can estimate our predisposition for cancer diseases.	F
2006	10	1	NTC – thermistors -1	AFI
2007	11	1	HuPoTest - 40 years of continuous research Basic rules for preventing and vanishing cancer diseases Climate change = change of mentality Hot nuclear fusion – a project of actual mentality	F
2007	11	2	MT – Introduction to Mental Technology HuPoTest – general procedure, assignments of results, specimen of complete test , order and obtain your complete HuPoTest report	F

Year	VOL	NO	Content (titles)	$*)
2007	11	3	TRESISTOR© - data banks of materials with thermally driven electric and magnetic properties TRESISTOR© - NTC -1 - data bank of NTC thermistors	AFI
2008	12	1	Australian population: life, death and cancer	F
2008	12	2	Pattern of Cancer Diseases	F
2008	12	3	Adiabatic calorimetry – summary description of the demo prototype	F
2008	12	4	Flight QF 30 and even more… Temperature calibration of NTC-thermistors. 1.Preliminary results.	F
2009	13	1	Proposal for interlaboratory comparisons. Calibration of NTC-thermistors (The 14th International Metrology Congress, Paris, France, 22-25 June 2009)	F
2009	13	2	Sudoku – un algoritm de rezolvare (Sudoku – an algorithm for solution)	AFI
2009	13	3	Cancer and Diabetes – as social diseases (Open letter to all whom it may concern)	F
2010	14	1	Studies on cement hydration by High Resolution Mixing Calorimetry (HRMC)	F
2010	14	2	Measuring tools for subtle potentials; pas-LED: an efficient measuring tool for subtle potentials.	F
2010	14	3	Upon some features of cancer in Australia: 1982 - 2006	F
2010	14	4	Cancer as an erosion process in human society	F
2010	14	5	Cancer erosion in Australian human society: 1982 - 2006	F
2010	14	6	Cancer erosion in German human society:1980-2008	F
2011	15	1	Procedures and devices for energy and water saving. (I) (in Romanian)	F

*) F=free, AFI=ask for invoice.

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