

## Separation of Chlorine Isotopes in Sodium Chloride Solution Using Thermal Diffusion Column

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### ABSTRACT

The separation of chlorine isotopes in an aqueous solution is demonstrated... The separation took place in a thermal diffusion, column TDC. Separation action was proved using four types of measurements. These measurements include density, electrical conductivity, melting temperature and PH values. The TDC used is 1.5m high. The TDC heating process was that which employs skin effect electromagnetic heating.

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فصل نظائر الكلور في المحلول الألكتروليتي باستخدام عمود الانتشار الحراري

### المخلص

تم بيان إمكانية فصل نظائر الكلور في المحاليل الألكتروليتيّة. جرت عملية الفصل باستخدام برج الانتشار الحراري وتم اثبات حدوث هذا الفصل باستخدام ثلاث طرق للقياس هي، قياس الكثافة والتوصيلية الكهربائيّة ودرجة الانصهار ودرجة الحمضية. جرى استخدام برج انتشار حراري ارتفاعه 1.5 متر واستخدمت فيه طريقة التسخين بظاهرة التوصيل السطحي الكهرومغناطيسي (ظاهرة الجلد).

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### INTRODUCTION

Thermal diffusion is one of many tools that can be used to separate gas mixtures and isotopic mixtures (Perry and Chilton 1953). A large number of papers have been published in this respect (Murry 1983). However thermal diffusion separation has almost entirely concerned itself with isotopic separation in the gaseous states. Recently, Eocenarro et al., (1985,89,89,90,90,91,94,94,96,98,99,99,99) published several papers dealing with the problem of separation of organic liquid mixtures using TDC. Separation of substances in the aqueous solutions states has been limited so far Home and Yuan (1989.) This may be due to

the fact that the TDC itself is not well suited for handling electrically conductive liquids this is because of possible electrical leak between the hot wire and the solution in the TDC

Recently a new method was adopted to carry out the heating process in the TDC. This method involves what is called skin effect heating process, Kasem (1999). In such process the hot wire is replaced by an electrically conductive thick copper wire while the hot tube is made of a ferromagnetic material such as carbon steel. High electric current values are allowed to pass through the hot tube where the electromagnetic induction caused by the AC current will produce the heating action

Separation of lithium isotopes and heavy water was demonstrated using such, column Hilal (2001) in the present work experimental data on sodium chloride solution separation is reported

#### **Experimental Set Up:**

The Experimental setup considered here consists of a 1.5 meter thermal diffusion column which employs skin effect heating process. The geometrical and physical parameters of this column are described elsewhere. Ten samples of sodium chloride solutions with different concentrations were prepared these concentrations ranged between of 0.1-1mole in the steps of 0.1mole the sodium chloride used was of chemical purity. Doubly distilled water was used to prepare the solutions. Each solution was fed into the TDC and the TDC was allowed to run for 24 hours. It is known from experience that thermal equilibrium can be reached after six hours. Thus it is assumed that thermal equilibrium was well established. At the end each run samples of solution were taken from the top and the bottom sampling ports of the TDC. These samples were saved for later analysis.

Four types of analysis were carried out on the samples. These are density measurement, electrical conductivity, PH value and melting points measurements. density measurements were carried out using a high sensitivity weighing equipment model Metler H54.AR with sensitivity of  $10^{-5}$ gm. Electrical conductivity of each sample was obtained using an electrical conductivity meter model (Wesson Schftich che-Werkstatten-D8120) A Metler PH meter was used to get the PH values of each sample.

Melting point measurements were carried out using the standard method. The samples were put in the deep freeze until becoming solid. Each sample was taken out separately, allowed to settle at room temperature. The temperature of the sample is recorded continuously until all solid converts into liquid. The value of the constant temperature during the melting is recorded to be the melting temperature.

It may be worth stating that good care was taken during all runs and measurements. The procedure involved flushing the TDC system and all vessels used with alcohol and distilled water before every new run to avoid any contamination. The more or less systematic behavior of the data is a fair indication that contamination effects do not represent a major problem.

### RESULTS AND DISCUSSION

Table (1) summarizes all numeral results obtained. These results include values of density, electric conductance and solution melting points for all samples obtained from the top and the bottom of the TDC. The estimated experimental error on each type of measurement depending on equipment accuracy is also shown.

Table 1: Changes in some physical properties of solutions in the TDC.

Conc. Mole	PH values ± 0.001		Melting point °C ± 0.01		Electric conductance mho.cm ± 0.01		Density Gm/cm <sup>3</sup> ± 0.0001	
	Top	Bot	Top	Bot	Top	Bot	Top	Bot
0.1	8.163	8.535	-0.1	-0.25	0.58	0.61	.9949	0.9951
0.2	7.920	8.660	-0.25	-0.5	1.0	1.0	.9989	0.9991
0.3	8.314	9.160	-0.7	-1.0	1.4	1.3	1.0021	1.0018
0.4	8.031	8.432	-1.1	-1.5	1.7	1.6	1.0085	1.0082
0.5	7.943	8.312	-1.75	-2.0	2.2	2.2	1.0104	1.0106
0.6	7.865	8.107	-1.0	-2.0	2.6	2.6	1.0141	1.0141
0.7	7.832	8.060	-1.76	-2.75	2.9	3.0	1.0179	1.0182
0.8	7.560	7.995	-2.0	-2.5	3.3	3.3	1.0218	1.0218
0.9	7.803	7.895	-3.5	-4.25	3.65	3.7	1.0286	1.0288
1.0	7.882	7.970	-4.5	-5.5	3.95	4.0	1.0295	1.0300

Density measurements result of top and bottom samples are shown in Fig(1). Although the results indicate a systematic, but only slight difference between top and bottom samples, no solid conclusions can be drawn about the separation effect. This is because that even the differences being systematic; with the top samples density being lower than those at the bottom, however, these differences are not far away from the margin of experimental errors.

More clear evidence concerning the separation effect can be drawn from Fig (2). It is clear that top samples melting points are well above the corresponding bottom ones. This is a good evidence that molar concentrations at the top are always lower than those at the bottom.

In order to establish the nature of the separation effect, electrical conductivity measurements were carried out on all samples. No differences could be detected. This indicates that the total number of ion pairs did not change during the separation process. This later result appears to be in some contradiction with that deduced from the results of Fig (2). This contradiction can be explained if one considers the results of Fig (3). This fig shows the results of PH values measurements. It is clear again here that some sort of separation effect is taking place.

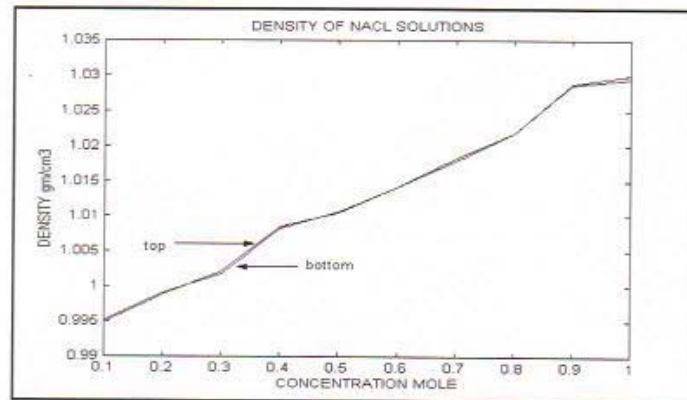


Fig 1: Density versus concentration mole for NaCl solution.

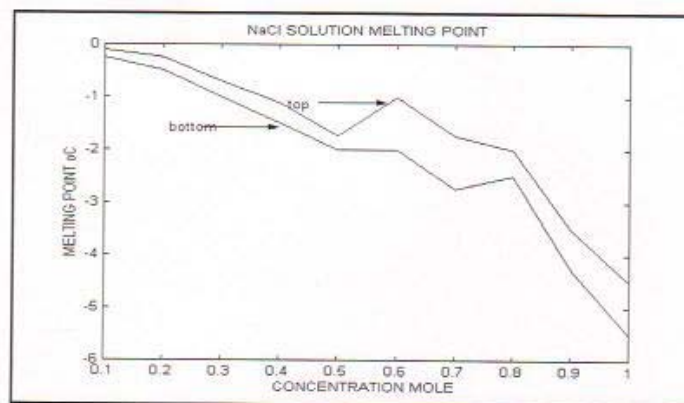


Fig 2: Melting point versus concentration mole for NaCl solution.

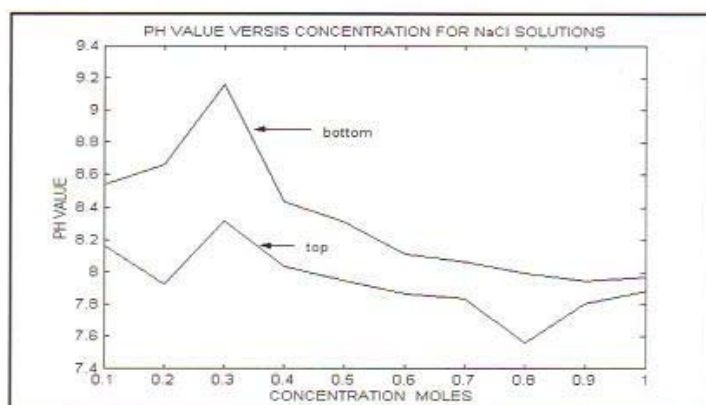


Fig 3: pH value versus concentration mole for NaCl solution.

In order to explain the separation effects taking place within the solution, one has to consider two effects that might be taking place:

- 1- If the separation effect is taking place between the light water molecules and the heavier NaCl molecules, one would expect that the melting points at the top should indeed be higher than those at the bottom. Such an effect is present in Fig (2). However the PH value should not be that sensitive to this type of separation. This contradicts what we observe in Fig (3)
- 2- Another separation effect could be due to ionic migration. Heavier ions tend to migrate toward the bottom while lighter ones to the top. As sodium ions are all identical because sodium has only one isotope  $\text{Na}^{23}$ , no migration of these ions is expected to take place.

However, Chlorine has two main isotopes,  $\text{Cl}^{35}$  and  $\text{Cl}^{37}$ . Thermal diffusion action in the column would act to separate these two isotopes to the top and to the bottom respectively. Because  $\text{Cl}^{35}$  has the larger natural abundance, a larger proportion of Chlorine would accumulate at the top. This effect will be reflected on the PH value curve. The lower part of the solution would be of less acidic reaction than that at the top

This hypothesis is consistent with the results of Fig (2). This can be understood when looking at hand books of physical chemistry. (1976) it can be noticed that the lowering of melting point of an aqueous NaOH solution per one mole is greater than the corresponding one on HCl. Thus it

Is not so surprising that bottom samples containing less Cl ions (more NaOH) should have lower melting points than those at the top containing more Cl ions (less NaOH).

Thus, the two effects concerning the melting points and PH values could be explained on the bases of chlorine isotopic separation in the TDC. This may not be very surprising if we remember that TDC's has been used long ago to separate isotopes in the gaseous state.

In order to establish if the separation effect is somehow related to the original concentration of the solution, a linear fit was attempted between these two parameters. The separation effect was represented by the difference in melting points  $\Delta T$ . The best  $\chi^2$  fit obtained using MATLAB computer program fitting facility is:

$$\Delta T = 0.07 + 0.89C$$

Where:  $C$  is the molar concentration.

A similar fit using PH values differences  $\Delta PH$  gives the following correlation relation:

$$\Delta PH = -0.7 + 0.57C$$

From both cases above, it is clear that the separation effect is strongly related to the solution concentration.

### CONCLUSIONS

Separation effects concerning Chlorine isotopes in a thermal diffusion column are demonstrated. Separation took place when chlorine was in the form of sodium chloride aqueous solution. The effect seems to be related to the concentration. The effect is clear from both melting point and PH values measurements.

### REFERENCES

- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L. and Saviron, J.M., 1985. Temperature difference effects in liquid thermal diffusion columns. *J. Non-Equilib. Thermodyn.* Vol. 10, pp. 185-192.
- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L. and Santamaria, C.M., 1989. Thermogravitational column as a technique for thermal diffusion factor measurement in liquid mixtures. *Separation Science and Technology*, 24 (7 and 8), 555p.
- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L.; Santamaria, C.M.; Carrion, J.A. and Saviron, J.M., 1989. Non-steady-state density effects in liquid thermal diffusion columns. *J. Phys. Condens. Matter*, 1: pp. 9750-9741.
- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L.; Santamaria, C.M.; Navarro, J.L.; Carrion, C.M.; Carrion, J.A. and Saviron, J.M., 1990. Mass transport coefficient from liquid thermal diffusion columns. *Ber. Bunsenges. Phys. Chem.* Vol. 94, pp. 378-377.
- Ecenarro, Carrion, J.A., Madariaga, J.L., Navarro and C.M. Santamaria and J.M. Saviron (1990), Tickian and thermal diffusion coefficient from liquid Thermo gravitational column's *J. Phys. Condens. Matter* 2, pp. 2289-2296.
- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L.; Santamaria, C.M.; Carrion, J.A. and Saviron, J.M., 1991. Direction of Separation and Dependence of Feed concentration in Liquid Thermo gravitational columns. *Separation science and Technology*, 26(8), pp. 1065-1067.
- Ecenarro, O.; Madariaga, J.A.; Navarro, J.L. and Santamaria, C.M.; Carrion, J.A.; Saviron, J.M.; Bou-Ali, M.M., 1994. Separation by Thermo gravitational diffusion in free media. *Entropie* no 184/185.
- Ecenarro, O.; Madariaga, J.A.; Santamaria, C.M. and Bou-Ali, M.M., 1996. Thermo gravitational diffusion in liquid mixtures with negative Soret effect. *Entropie* no 198/199.

- Ecenarro, O.; Madariaga, J. A.; Santamaria, C. M. and Bou-Ali, M. M., 1998. Thermo gravitational measurement of the soret coefficient of liquid mixtures. *J. phys. Condens. Matter* 10, pp.3321-3331.
- Ecenarro, O.; Madariaga, J. A.; Santamaria C. M. and Bou-Ali, M. M. and Valencia, J. J., 1999. Influence of the Grashof number on the stability of the thermo gravitational effects in liquid mixtures with negative thermal diffusion factor. *Entropie* no 218.
- Ecenarro, O.; Madariaga, J.A.; Santamaria, C.M. and Bou -Ali, M.M. and Valencia, J.J., 1999. Stability of convection in a vertical binary fluid layer with an adverse density gradient. *Physical Review E*, Vol. 59, No. 1.
- Ecenarro, O.; Madariaga, J.A.; Santamaria, C. M. ; Bou-Ali, M. M. and Valencia, J. J., 1999. Soret Coefficient of same binary liquids mixtures. *J. Non-Equilib. Thermodyn.* Vol. 24, pp.228-233.
- Home, F. H. and Yuan Xu, 1989. Thermo gravitational thermal diffusion in Electrolyte solutions, steady state. *J. phys Chem.* Vol.93, pp6539
- Hilal, H. S., 2001. M.Sc. Theses,mosul university. Skin effect heating of Thermal diffusion column and separation of some isotopes
- Kasem, A.W., 1999. M.Sc Theses mosul university. Skin effect heating of industrial pipes.
- Murray, R. L., 1983. *Nuclear Energy: An Introduction to the Concepts system and Applications of nuclear processes.* 2nd edition.
- Perry, R. H. and Chilton, C.H., 1953. *Chemical Engineer's Handbook*, Fifth Edition.