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Thermal Diffusion Factors and Intermolecular Potentials for Noble Gas - SF₆ Systems

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ABSTRACT

Experimental thermal diffusion factors for equimolar mixtures of He-, Ne-, Ar-, Kr-, and Xe-SF₆ have been measured in the temperature range from 225 to 500 K. The data were obtained in a 20-tube trennschaukel, or "swing separator". The systems containing the four lighter noble gases all exhibited a "normal" thermal diffusion factor, α_T , that is concentration of the heavy species, SF₆, in the cold region of the apparatus and increase of α_T with temperature. Xe-SF₆, the system with the smallest mass difference, exhibited "abnormal" behavior. The spherically symmetric Pack potentials were used to calculate the thermal diffusion factor with reasonable success. Recently published dipole-dipole dispersion coefficients were used to construct intermolecular potentials of the Hartree-Fock-Dispersion functional form with individually damped attractive terms. The potentials, when tested against the available transport and thermodynamic data, improved the fit to experiment in almost all cases.

I. INTRODUCTION

In a binary mixture in the gaseous phase in which the temperature is steady but not uniform, transport of both matter and heat may be observed. If the higher temperature is at the top of the apparatus so that convection is not present, the degree of separation is known as the thermal diffusion factor, α_T . The separation is a molecular migration of the components due to the temperature gradient which normally manifests itself as a concentration of the heavy species in the cold region and the light species in the hot region. The convention is, that when this occurs, a positive value is assigned to α_T which also customarily increases in magnitude with increasing absolute temperature. By far the majority of systems behave in this manner but occasionally one finds "abnormal" behavior in which either the light species enriches in the cold region (negative α_T) or α_T decreases in magnitude with increasing temperature. This is quite rare and it is generally observed that one of the components is a diatomic or polyatomic molecule and also that the masses are relatively close together. It is assumed that the internal degrees of freedom contribute in some fashion to α_T and that when the masses are close the inelastic contribution overshadows the normal statistical separation process.

This laboratory has been engaged in measuring thermal diffusion factors for a number of years and has studied only one "abnormal" system (Taylor and Pickett, 1986) which had already been reported in the literature. The present work reports ^{on} another such system which, to the best of our knowledge, has not yet been observed. This is the Xe-SF₆ system. In addition, this investigation covers the entire homologous noble gas - SF₆ series, presenting

thermal diffusion factors and intermolecular potentials for the five systems.

II. EXPERIMENTAL

a) The Apparatus

The experimental device used for the determination of the thermal diffusion factor was a trennschaukel, or "swing separator", developed by Clusius and Huber (1955). In principle this device is a number, n , of two-bulb experiments connected in series as shown in Figure 1. The top and bottom ends of the tubes (approximately 1/3) are embedded in isothermal regions held at temperatures T_H and T_C , respectively. The temperature gradient, ΔT , is established in the middle portion of the tubes. By connecting each successive tube top to bottom with a capillary and gently moving the gas to and fro by means of a pump, the concentrations of the components of the gas mixture in the top of one tube are made essentially identical with those at the bottom of the next tube. Because the temperature gradient is applied in the vertical direction with the hot end at the top, the effect of convection is eliminated. The separation factors of the individual tubes are multiplicative, so an n -fold increase in the separation is realized.

The actual apparatus utilized is a 20-tube trennschaukel which has been described in detail by Taylor and Pickett (1986). It consists of two solid nickel blocks in the shape of a cylinder with a hole in the middle. Twenty equally spaced Inconel tubes and capillaries are welded into the nickel blocks and connected in series. The gas mixture is swung by a bellows pump which, along with the trennschaukel, is located in an environmental chamber.

Both bellows pump and trennschaukel are connected to an external vacuum and gas handling system.

b) Conducting the Experiments

The present measurements were carried out in the temperature range of approximately 200 to 500 K using gases of research grade purity. Experiments were attempted at higher temperatures but above approximately 575 K some breakdown of the SF₆ was observed, probably due to catalysis by the nickel in the apparatus. Duplicate experiments were conducted in many cases to insure reproduce ability of the results. Some low temperature experiments were conducted in a different apparatus which had only ten tubes. Both systems produced similar results confirming the reported values.

Suitable consideration was given to the operating parameters of the trennschaukel such as operating time, pump period, pumped volume and pressure, for each of the experiments. As simple as the apparatus in Figure 1 appears, it is still subject to certain restrictions which, if violated, will invalidate the results. For example, one must operate sufficiently long so that steady state concentrations in the regions under and over the pump are approached. The frequency of pumping lies within certain limits. If one pumps very slowly, backward diffusion in the capillaries sets in, but if pumping is too rapid, the motion of the gas disturbs the thermal diffusion balance in the tubes. The theory of material transport in the device was derived by van der Waerden (1957). Explicit expressions involving the physical characteristics and operating parameters for the present device have been worked out in detail

by Taylor (1989) and these conditions and corrections were applied to the present experiments.

The compositions at the hot and cold ends of the trennschaukel were measured at the end of the experiments on a Finnegan Model 271 mass spectrometer. Three corrections for approach to equilibrium, back diffusion in the capillaries, and disturbance due to pumping were applied to the measured composition differences, Δx_i , obtained between the hot and cold blocks. The overall separation factor, Q , is

$$Q = \frac{1 + [\Delta x_1 (corr) / x_1 (T_H)]}{1 + [\Delta x_2 (corr) / x_2 (T_C)]} \quad (1)$$

where x_1 and x_2 are the mole fractions of the light and heavy species, respectively. The experimental thermal diffusion factor is given by

$$\alpha_T = \frac{1}{n} \frac{\ln Q}{\ln (T_H / T_C)} \quad (2)$$

where n is the number of stages (or tubes) in the trennschaukel.

c) Other Experimental Data

In addition to the present thermal diffusion measurements, several other transport and thermodynamic properties were used as available in fitting parameters for the intermolecular potentials, using data found in the literature. These properties are the viscosity, η , the second virial coefficient, B , and the binary diffusion coefficient, D_{12}^0 .

He-SE₆: Viscosity data of Kestin *et al.* (1977a) [298 - 483 K]; second virial coefficients

of Martin *et al.* (1982) [290 - 320 K] and Bell and Dunlop (1981) [293 - 320]; binary diffusion coefficients of Trengove *et al.* (1984) [280 - 320 K], Ivakin and Suetin (1964) [290 - 455 K], and Loiko *et al.* (1981) [192 - 572 K]; and thermal diffusion factors of Trengove *et al.* (1984) [255 - 300 K].

Ne-SF₆: Viscosity data of Kestin *et al.* (1977a) [298 - 483 K]; second virial coefficients of Martin *et al.* (1982) [290 - 320 K]; and binary and thermal diffusion data by Trengove *et al.* (1984) [255 - 300 K].

Ar-SF₆: Viscosity data of Kestin *et al.* (1977a) [298 - 483 K]; second virial coefficients by Martin *et al.* (1982) [290 - 320 K], Bell and Dunlop (1981) [293 - 320 K], Bellm *et al.* (1974) [300 - 550 K], and Santafe *et al.* (1974) [273 - 323 K]; and binary diffusion coefficients by Trengove *et al.* (1984) [280 - 320 K], Ivakin and Suetin (1964) [287 - 472 K], and Loiko *et al.* (1981) [186 - 555 K].

Kr-SF₆: Viscosity data of Kestin *et al.* (1977b) [298 - 473 K]; second virial coefficients of Martin *et al.* (1982) [290 - 320 K] and Santafe *et al.* (1974) [273 - 323 K]; and binary diffusion coefficients of Trengove *et al.* (1984) [280 - 320 K].

Xe-SF₆: Second virial coefficients by Martin *et al.* (1982) [290 - 320 K], and binary diffusion coefficients of Trengove *et al.* (1984) [277 - 323 K].

III. THEORETICAL

Pack *et al.* (1982a, 1984) and El-Sheikh *et al.* (1990) have made a study of anisotropic noble gas - SF₆ intermolecular potentials derived from molecular beam differential scattering. Angle dependent calculations were made by these workers using a Morse-Spline-van der

Waals (MSV) potential function wherein the adjustable parameters were adjusted to fit the experimental differential cross sections and other properties. It was concluded that the anisotropic effects were small, probably on the order of a few percent. Prior to publishing the potential, Pack (1982b) had calculated approximate values of the first van der Waals dispersion constants for each of the systems thus fixing the C_6^* parameters.

Recently Kumar *et al.* (1985) have redetermined the first van der Waals dispersion coefficients for the five systems and obtained considerably different values from those calculated by Pack. Because of the small anisotropic effect, the present work utilizes a spherically symmetric potential of the Hartree-Fock-Dispersion (HFD) type with the new dispersion coefficients to fit the new thermal diffusion data and the previous transport and thermal property data. The potential used has individually damped dispersion coefficients and is given in reduced form in the following.

Ahlrichs *et al.* (1977) originally proposed the HFD potential form which was extensively used for noble gas interactions. Aziz and Chen (1977) found it necessary to add a squared term to the exponent of the repulsive portion of the potential. The reduction parameter for HFD-type potentials is generally taken to be the location of the minimum in the well, so here $x = r/r_m$, and

$$[V^*(x)]_{rep} = A \cdot \exp^{-\alpha \cdot x + \beta \cdot x^2} \quad (3)$$

Feltgen (1981) has constructed a model formula to represent the dispersion interaction from the interatomic electron correlations, which individually damps each term in the

multipole expansion. In the present case the first six terms of the expansion were used, yielding for the attractive part of the potential, the truncated series

$$[V^*(x)]_{att} = -\sum_{n=6}^{16} f_n(x) \frac{C_n^*}{x^n} \quad (n \text{ even}) \quad (4)$$

The damping function, $f_n(x)$, is extremely complicated. McConville (1984) has suggested keeping the form of the damping function as presented by Ahlrichs *et al.* (1977) but constructing individual damping functions, whose magnitude and range depend upon the multipole expansion coefficients, as shown by Feltgen *et al.* (1982). The damping function is

$$f_n(x) = \exp\left\{-\left\{\frac{D_n}{x}-1\right\}^2\right\} \quad x < D_n$$

$$f_n(x) = 1.0 \quad x \geq D_n \quad (5)$$

where the constants, D_n , are evaluated at the mid point of the damping function. Feltgen has derived the relationship

$$f_n(x_n) \approx 0.5, \text{ for } x_n = (C_{n+2}^* / C_n^*)^{1/2} \quad (6)$$

which yields

$$D(n) = 1.8326 (C_{n+2}^* / C_n^*)^{1/2}. \quad (7)$$

The complete potential is

$$V^*(x) = A^* \exp^{-\alpha^*x + \beta^*x^2} - \sum_{n=6}^{16} f_n(x) \frac{C_n^*}{x^n} \quad (n \text{ even}) \quad (8)$$

The transport properties were calculated in the classical approximation using collision integrals generated by Program COLL (Taylor, 1979). The third order kinetic approximation to the thermal diffusion factor was calculated (Taylor *et al.*, 1985) while the second order diffusion and mixture viscosity expressions from kinetic theory were used (Hirschfelder *et al.*, 1954). A semi-classical expression was employed for the second virial coefficient (Hirschfelder *et al.*, 1954).

IV. RESULTS

a) Experimental

The present experimental results for the five noble gas-SF₆ systems which were measured in the 20-tube trennschaukel described previously, are presented in Table I. The data for He-, Ne-, and Ar-SF₆ are shown in Figure 2 and those for Kr- and Xe-SF₆ are shown in Figure 3. The four lighter noble gas systems exhibited "normal" behavior in that α_T increased with increasing temperature and was positive; e.g. the heavy SF₆ molecule enriched in the cold region. This was not unexpected because "abnormal" behavior usually occurs in systems with very similar masses. The Xe-SF₆ system however, with a relative mass difference of approximately 10%, demonstrated abnormal behavior. The thermal diffusion factor decreased moderately over the entire temperature range investigated, yielding a very small negative value at the highest temperature. Following the procedure used by Taylor (1980), polynomials of the type

$$\alpha_T = \sum_{n=1}^3 C_n T^{n/2} \quad (9)$$

Table I
Thermal Diffusion Factors for the Noble Gas - SF₆ Systems

He - SF ₆			Ar - SF ₆ (Cont.)		
T _{av} (K)	α _T	%Unc.	T _{av} (K)	α _T	%Unc.
200	0.4294	9.6	389	0.1494	4.5
225	0.4624	7.3	446	0.1744	6.9
230	0.4678	7.3	446	0.1843	6.9
250	0.4742	2.9	501	0.1923	7.3
280	0.4917	4.1	502	0.1928	7.3
310	0.4826	7.1	Kr - SF₆		
311	0.4974	7.1	T _{av} (K)	α _T	%Unc.
352	0.4969	5.0	233	0.0308	4.6
352	0.5028	5.0	234	0.0319	4.6
429	0.5264	7.4	277	0.0376	20.5
429	0.5313	7.4	341	0.0503	3.6
497	0.5304	7.1	381	0.0575	6.8
497	0.5378	7.1	396	0.0608	6.6
Ne - SF₆			451	0.0689	6.9
T _{av} (K)	α _T	%Unc.	496	0.0763	7.9
233	0.2381	6.8	502	0.0779	7.9
275	0.2779	8.0	Xe - SF₆		
352	0.3052	4.3	T _{av} (K)	α _T	%Unc.
401	0.3613	6.7	231	0.0181	12.1
450	0.3845	7.9	235	0.0172	12.1
500	0.4014	9.9	269	0.0124	13.7
Ar - SF₆			276	0.0126	13.7
T _{av} (K)	α _T	%Unc.	309	0.0099	13.8
230	0.0841	5.6	310	0.0100	13.8
233	0.0699	5.6	352	0.0034	25.2
235	0.0724	5.6	352	0.0034	25.2
275	0.0929	4.8	427	0.0019	27.0
277	0.1029	4.8	427	0.0090	27.0
331	0.1287	2.5	429	0.0034	27.0
331	0.1247	2.5	502	-0.0020	25.2
331	0.1231	2.5	502	-0.0026	25.2
389	0.1488	4.5			

were used to find a function which yielded a satisfactory least squares fit to the experimental data for each system. For the normally behaving systems the function $\alpha_T = A + B/T^{1/2} + C/T$ was either the best choice, or very nearly the best, and well within the estimated experimental uncertainty. The search was limited to three parameter functions because of the relatively low number of points available. The results were:

He-SF₆

$$\alpha_T = 0.61403 - 0.58676/T^{1/2} - 26.571/T; \quad rms \ dev. = 1.7\% \quad (10)$$

Ne-SF₆

$$\alpha_T = 1.2173 - 25.131/T^{1/2} + 155.99/T; \quad rms \ dev. = 3.9\% \quad (11)$$

Ar-SF₆

$$\alpha_T = 0.74217 - 16.704/T^{1/2} + 99.776/T; \quad rms \ dev. = 5.7\% \quad (12)$$

Kr-SF₆

$$\alpha_T = 0.34650 - 8.6166/T^{1/2} + 58.029/T; \quad rms \ dev. = 1.4\% \quad (13)$$

The above function was not satisfactory for the Xe-SF₆ system which did not behave in the usual fashion. The best function was

Xe-SF₆

$$\alpha_T = 0.44476E-2 - 0.64478E-10T^3 + 0.18138E+6/T^3; \quad rms \ dev. = 40.8\% \quad (14)$$

If the Xe-SF₆ datum point at 429 K is excluded from the correlation, the rms percent deviation can be reduced to approximately 24% with A=0.40815E-2; B=-0.64100E-10; and

$C=0.18724E+6$. However, the average estimated experimental uncertainty of the data is approximately 19% and neither equation is considered a completely satisfactory representation of the data. The least squares equations are shown as the solid lines in Figures 2 and 3. The range of validity of the above equations is approximately 200 - 500 K.

b) Theoretical

Potential functions were obtained for the five systems by the procedure outlined below and the parameters obtained are given in Table II. We used the dipole-dipole dispersion coefficients given by Kumar *et al.* (1985) and retained the zero of the potential as closely as possible to that obtained from differential cross sections. For Kr and Xe the exact values were used; for Ar the value was slightly larger; and for He and Ne the values were slightly smaller, but in all cases within $\pm 0.8\%$ of Pack's values. First the repulsive walls were approximated from the diffusion data by fitting to the actual values and adjusting the slope of the repulsive wall to predict the logarithmic plot of D_{12} versus T. The most accurate data were those of Trengove *et al.* (1984) but unfortunately the temperature range of their data was very small, being only about 50 K.

The dipole-quadrupole and quadrupole-quadrupole terms, C_6^* and C_8^* , which affect the shape of the well, were varied to simultaneously fit thermal diffusion and the second virial coefficient. The remaining dispersion coefficients needed, C_{12}^* through C_{18}^* , were obtained from the combining rules of Douketis *et al.* (1982) and are given by:

$$\begin{aligned}
C_{12}^* &= 1.028 C_6^* (C_{10}^*/C_8^*)^3 \\
C_{14}^* &= 0.975 C_8^* (C_{12}^*/C_{10}^*)^3 \\
C_{16}^* &= C_{10}^* (C_{14}^*/C_{12}^*)^3 \\
C_{18}^* &= C_{12}^* (C_{16}^*/C_{14}^*)^3
\end{aligned}
\tag{15}$$

As for diffusion, the temperature range of the available experimental second virial coefficient data was rather narrow, except for Ar-SF₆, in which case the data ranges for B and α_T were comparable and spread over several hundred degrees. The theoretical prediction of properties by the Ar-SF₆ potential was very accurate for all properties. In general it was

Table II
Reduced Parameters for the HFD-CID Potentials
Describing Noble Gas - SF₆ Interactions

	He - SF ₆	Ne - SF ₆	Ar - SF ₆	Kr - SF ₆	Xe - SF ₆
A*	1.4592409E+07	1.402631E+06	2.2934800+E05	1.0165219E+04	1.1352081E+05
α*	11.432508500	11.674363100	9.814840320	4.536530490	9.80225754
β*	-7.006056790	-3.654314990	-3.520080570	-5.491277690	-2.31121159
C ₆ *	0.6599639060	0.6089537140	0.6882240180	0.7170730230	0.764062524
C ₈ *	0.8686320190	0.7477934360	0.7461425070	0.7426090240	0.699833691
C ₁₀ *	1.3033266100	1.0284051900	0.9302735330	0.8844109770	0.737154365
C ₁₂ *	2.2917394600	1.6282625200	1.3711595500	1.2451944400	0.917934299
C ₁₄ *	4.6044282900	2.8937826200	2.3294735000	2.0311164900	1.324280740
C ₁₆ *	10.570228600	5.7728233300	2.3294735000	3.8383665100	2.213417050
σ(Å)	3.800	3.860	4.080	4.178	4.299
r _m (Å)	4.185	4.275	4.550	4.687	4.8075
ε/k (K)	57.00	113.00	218.10	247.10	291.685

properties by the Ar-SF₆ potential was very accurate for all properties. In general it was found that, with a given set of potential parameters and if the slope of the diffusion plot was close, the high temperature thermal diffusion was predicted accurately. The low temperature thermal diffusion and second virial coefficients were consistent in that minimum percent deviations were simultaneously achieved with the same parameters.

The potentials were then used to predict the viscosity with excellent success for Ar-SF₆ and Kr-SF₆ ($< \pm 1\%$). It should be noted that no experimental viscosity data are available for Xe-SF₆. The predicted viscosities for He-SF₆ and Ne-SF₆ were not as close, although they were within approximately $\pm 2\%$ of the reported values. Possibly, because the property of viscosity is manifested by momentum exchange, the magnitude of the viscosity coefficient is more strongly influenced by the heavy SF₆ collisions.

In all of the above calculations it was the unlike interactions which were of interest and certainly for ordinary and thermal diffusion and the interaction virials, the unlike collisions dominated the magnitude of the property. The like-like collisions do, however, have a significant role. A new HFD-type potential obtained by Taylor and Hurly (1990) was used and made a large improvement over any inverse power type of potential which was tried. For the noble gases, the like-like potentials used were: He-He (Aziz *et al.*, 1987); Ne-Ne (Aziz *et al.*, 1983); Ar-Ar (Aziz and Slaman, 1986a); Kr-Kr (Aziz and Slaman, 1986a); and Xe-Xe (Aziz and Slaman, 1986b).

V. CONCLUSIONS

Thermal diffusion factors were measured for equimolar mixtures of each of the five

noble gases with SF₆ in the temperature range from approximately 200 to 500 K. It is noteworthy that an heretofore unobserved example of "abnormal" thermal diffusion behavior was discovered for Xe-SF₆. We conclude that, at least for the properties considered, collisional averaging by and large smooths out the anisotropy in the SF₆ collisions so that a spherically symmetric potential can be used. It was also found that accurate potentials for the like-like collisions, especially for SF₆, were necessary to obtain self consistent results in the prediction of properties. Thus collision integrals calculated with the presently derived spherically symmetric potentials appear to predict macroscopic properties better than any of those previously published.

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FIGURE CAPTIONS

Figure 1 - Principle of the trennschaukel or "swing separator." A given number of tube/capillary pairs are connected top-to-bottom and the contained gas mixture is swung to-and-fro by a pump. A temperature difference, $\Delta T = T_H - T_C$, is imposed across the n tubes by embedding the top approximately 1/3 of the tubes in an isothermal region at T_H and the bottom 1/3 in another isothermal region at T_C .

Figure 2 - Temperature dependence of the thermal diffusion factor for He-, Ne-, and Ar-SF₆. The experimental points are: ●, present data; ■, Trengove *et al.* (1984); and ▲, Heintz *et al.* (1975). The solid lines are the least squares fit to the present data given by Equations (10), (11), and (12), respectively.

Figure 3 - Temperature dependence of the thermal diffusion factor for Kr- and Xe-SF₆. The experimental points are: ●, present data. The solid lines are the least squares fit to the data given by Equations (13) and (14).

Figure 4 - Percent deviation of experimental thermal diffusion factors for the five systems from the theoretical values (base line) calculated using the presented HFD potentials in Table II. The experimental points are the same as in Figures 2 and 3. The dashed curves are the MSV (spherically symmetric) potentials reported by Pack *et al.* (1982a, 1984) and El-Sheikh *et al.* (1990).

Figure 5 - Percent deviation of experimental ordinary diffusion coefficients for the five systems from the theoretical values (base line) calculated using the presented HFD potentials in Table II. The experiential points are: ●, Trengove *et al.* (1984); ▲, Ivakin and Suetin (1964); and ■, Loiko *et al.* (1981). The dashed curves are the same as in Figure 4.

Figure 6 - Percent deviation of experimental viscosity data for the four systems from theoretical values (base line) calculated using the presented HFD potentials in Table II. The experimental points are: He-, ●, $x_1 = 0.3856$, ○, $x_1 = 0.6547$; ○, $x_1 = 0.8697$ (Kestin *et al.*, 1977a); Ne-, ●, $x_1 = 0.3428$, ○, $x_1 = 0.7505$ (Kestin *et al.*, 1977a); Ar-, ●, $x_1 = 0.3758$, ○, $x_1 = 0.7949$ (Kestin *et al.*, 1977a); and Kr-, ●, $x_1 = 0.2173$, ○, $x_1 = 0.5135$ (Kestin *et al.*, 1977b). x_1 denotes the mole fraction of the noble gas. The dashed curves are the same as in Figure 4.

Figure 7 - Deviation of experimental second virial coefficients for the five systems from theoretical values (base line) calculated using the presented HFD potentials in Table II. The experimental points are: ●, Martin *et al.*, (1982); ■, Bell and Dunlop (1981); ○, Santafe (1974); and □, Bellm (1981). The dashed curves are the same as in Figure 4. For Kr the dashed curve is the Pack (D) potential and the dotted curve is the Pack (S) potential.

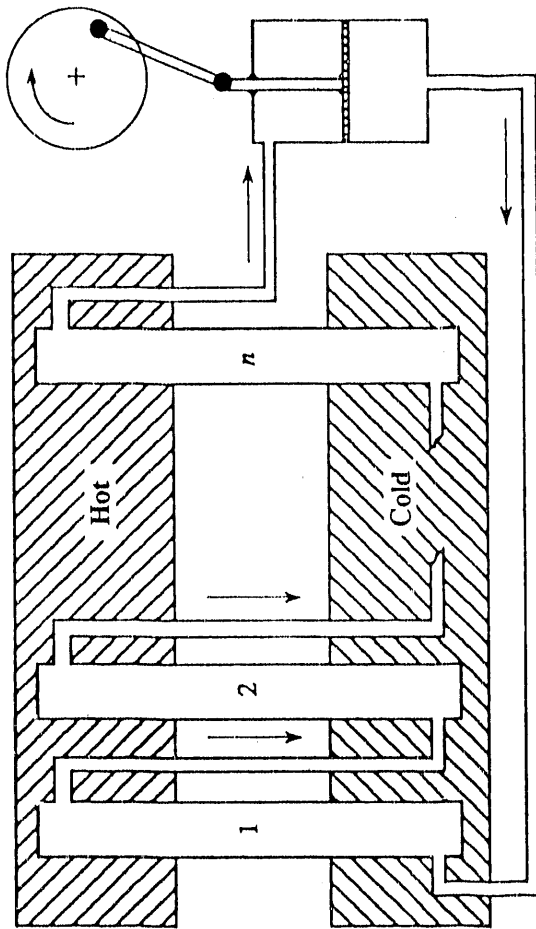


Fig. 1

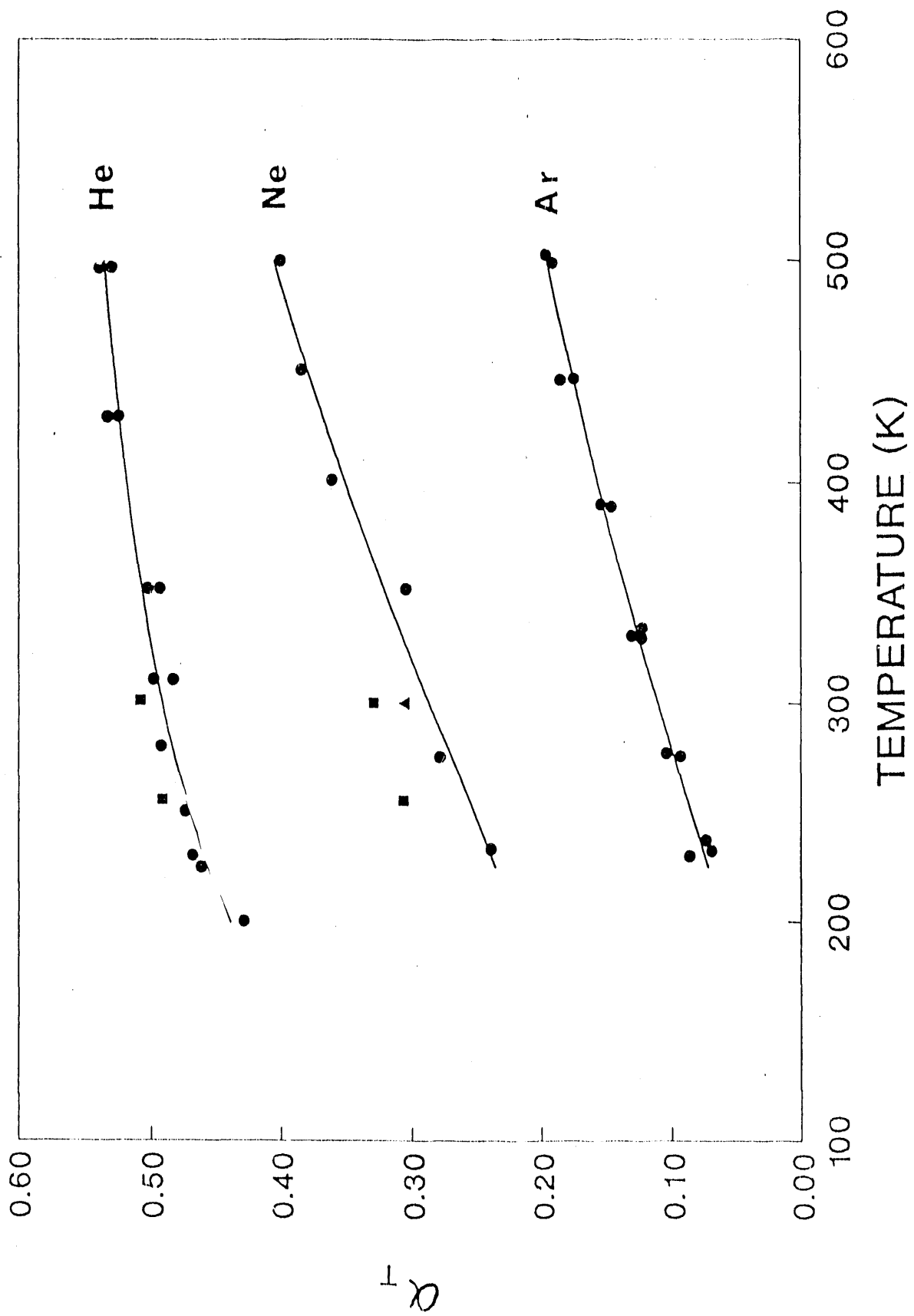


Fig. 2

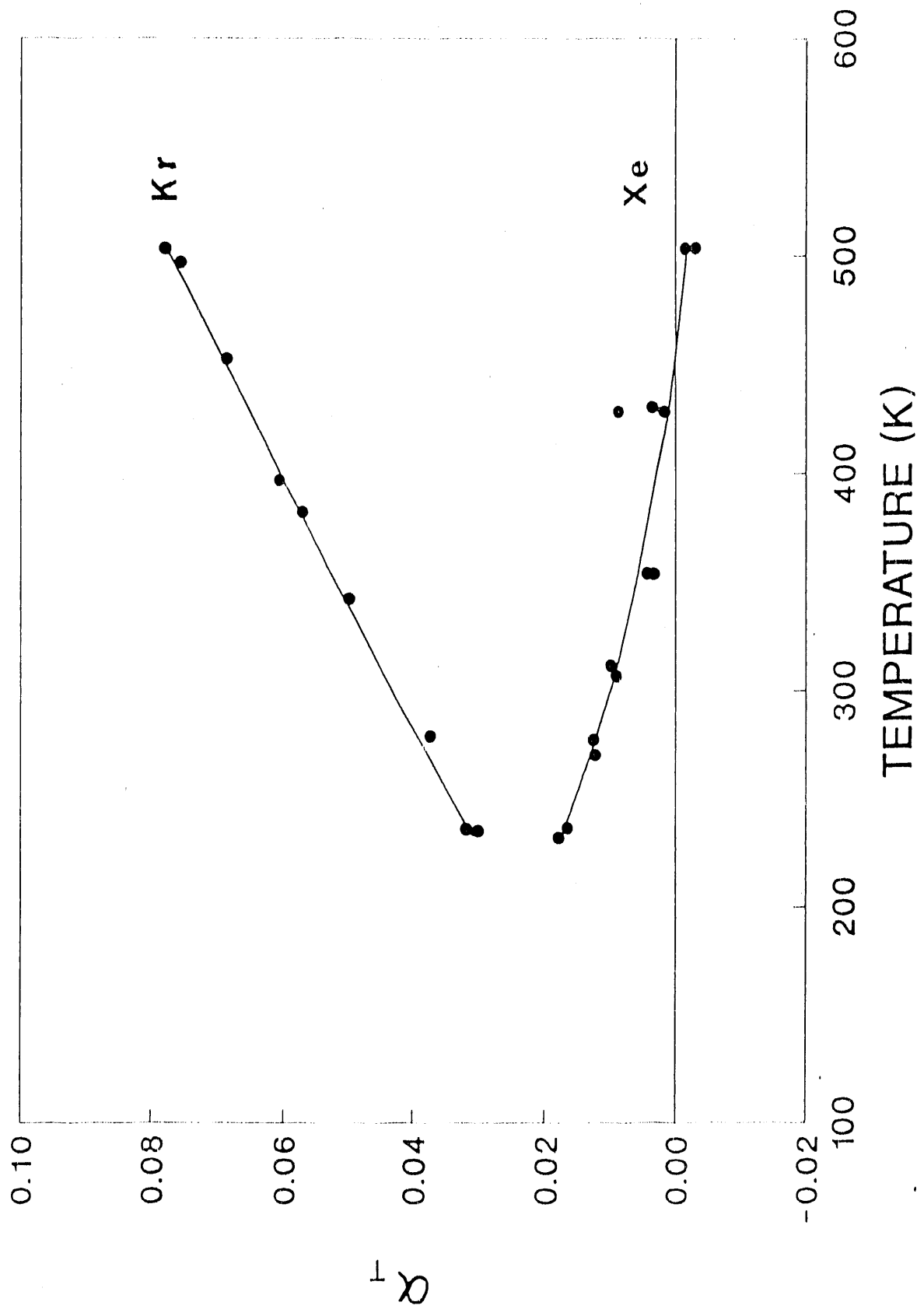


Fig. 3

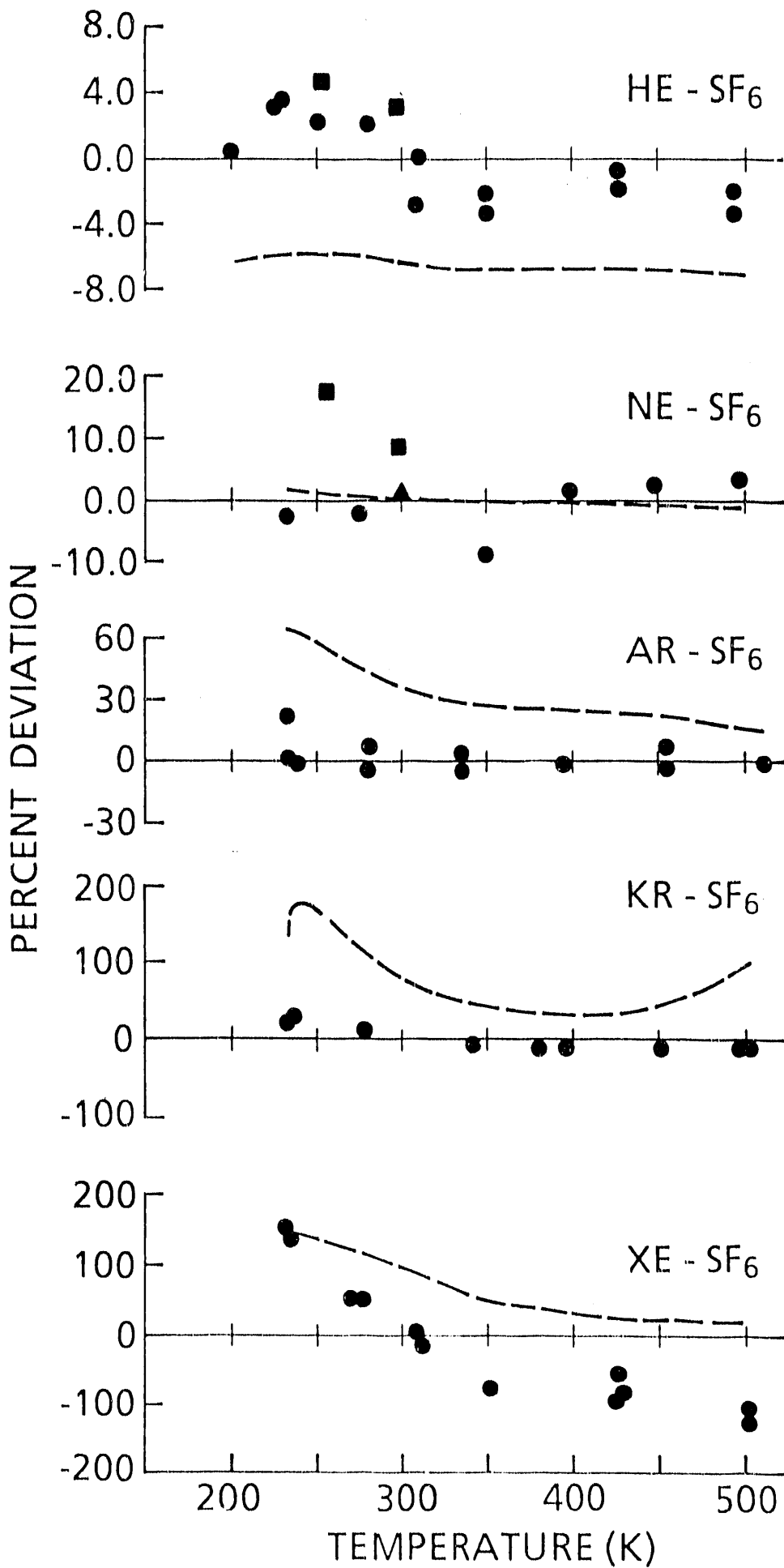


Fig 4

PERCENT DEVIATION

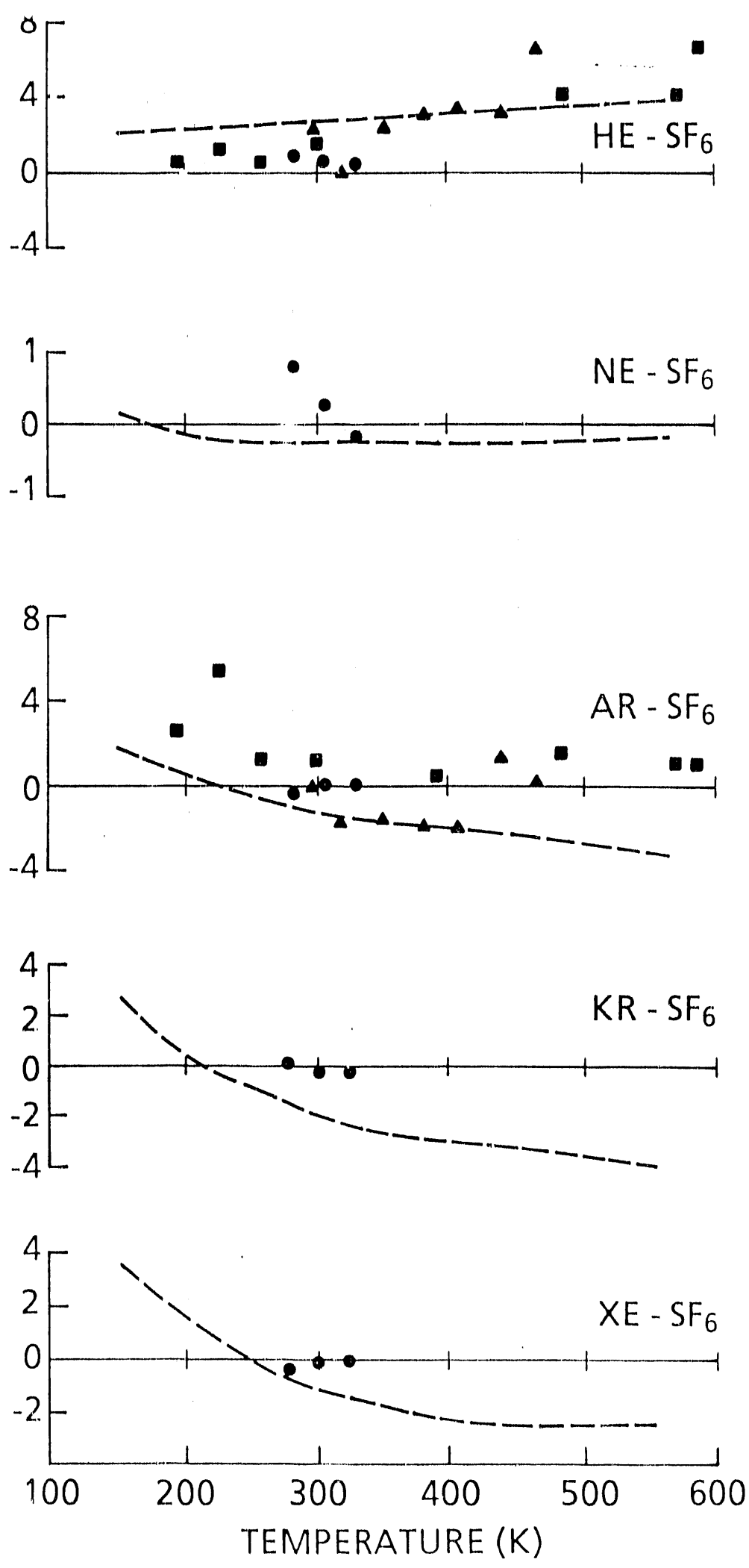


Fig. 5

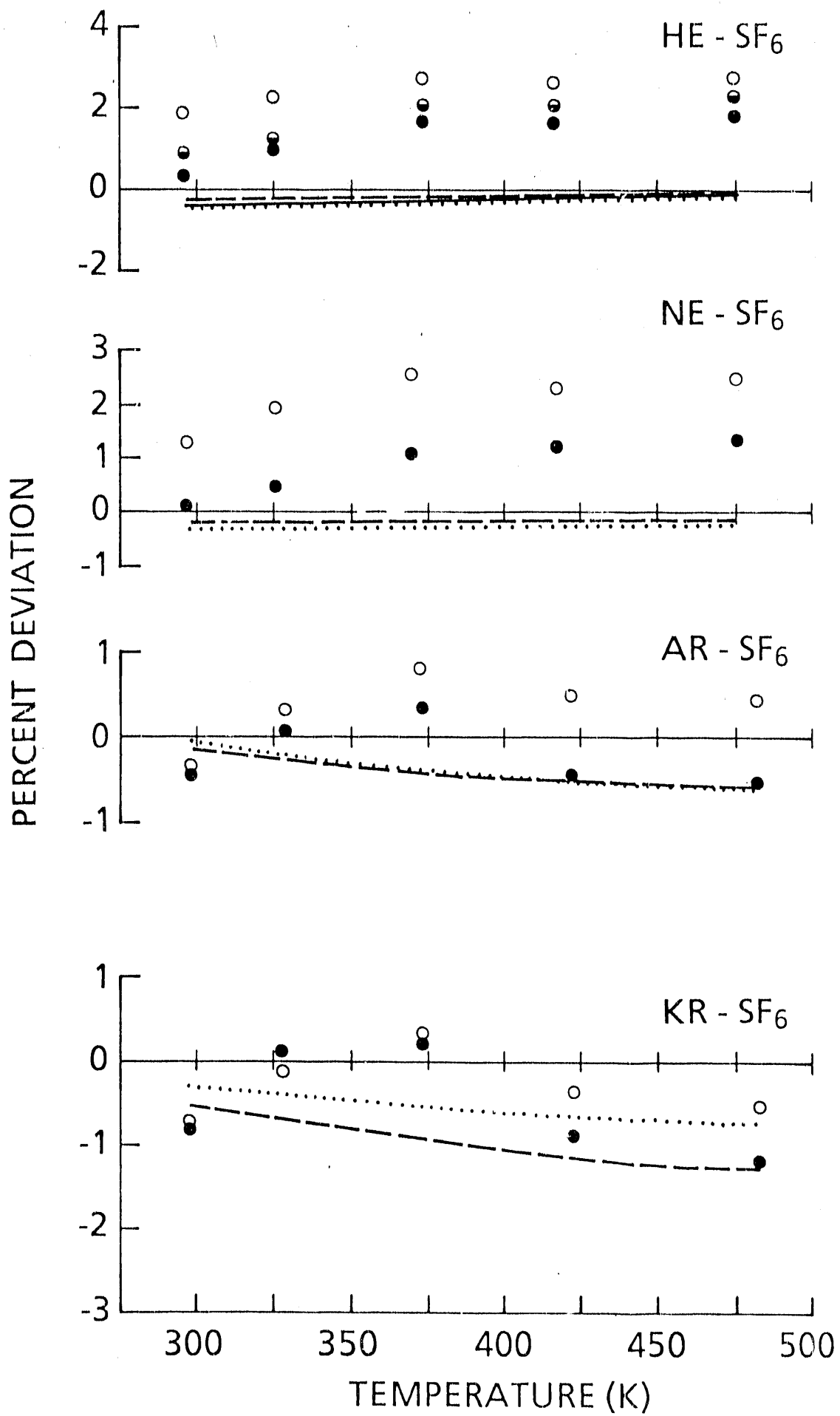


Fig. 6

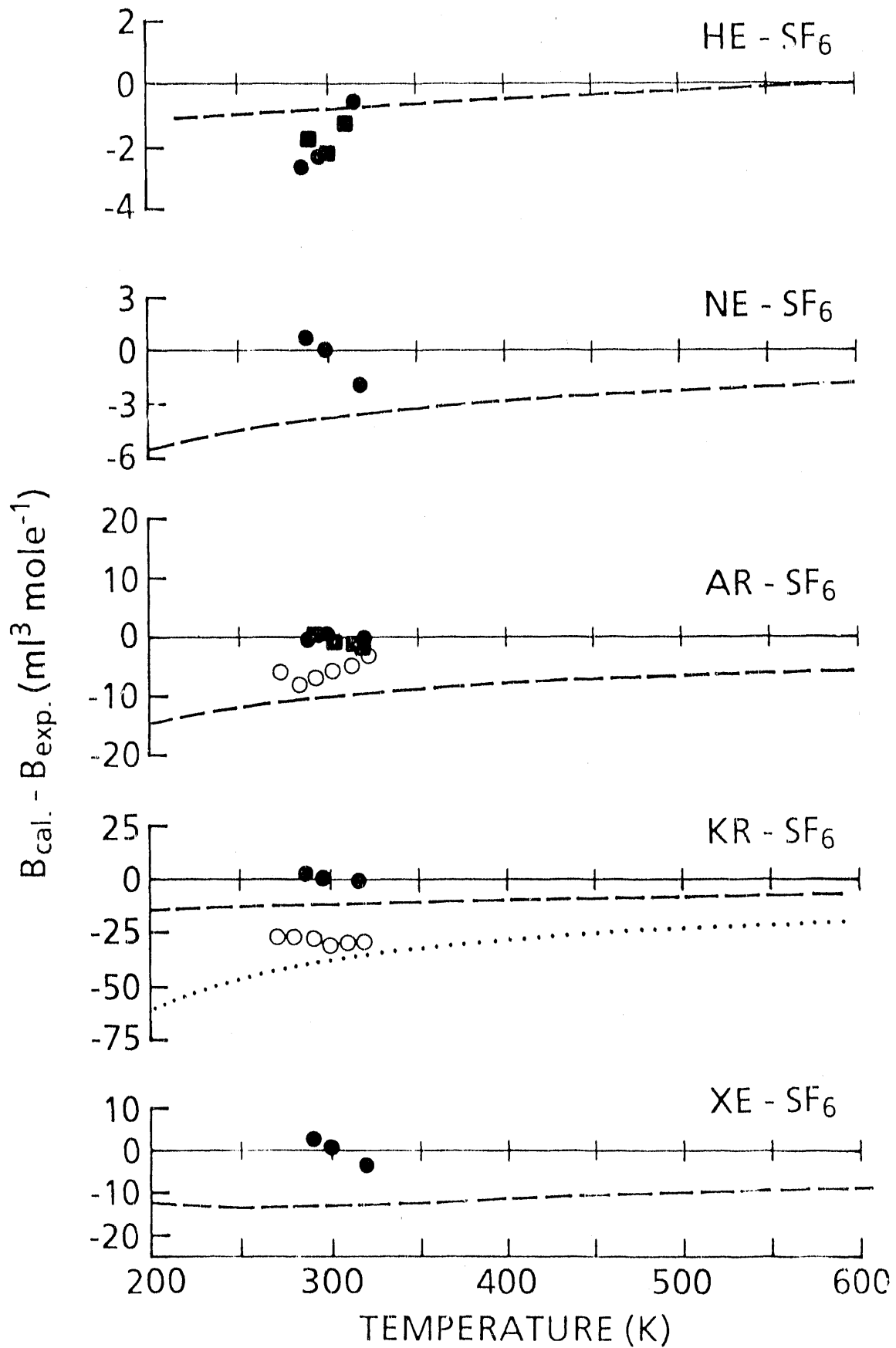


Fig. 7

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