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TRANSPORT PROPERTIES OF DILUTE GAS MIXTURES

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by Richard S. Brokaw

with the collaboration of Roger A. Svehla and Charles E. Baker

NASA, Lewis Research Center
Cleveland, Ohio, 44135

SYNOPSIS

If you want a quick, overall view, read the Introduction and Summary and Conclusions. The sections entitled Theoretical Considerations, Results and Discussion, and References are for the benefit of Missouri residents!

INTRODUCTION

There is a wealth of experimental data on the transport properties of gas mixtures - diffusion coefficients, mixture viscosities and heat conductivities, as well as results on thermal diffusion. Rather then attempting some sort of statistical assessment of all this information, a few simple systems which typify interactions amongst various types of molecules - monatomic and polyatomic, nonpolar and polar - are examined in the hope of obtaining some insight as to the concordance between theory (rigorous or approximate) and experiment (painstaking or prefunctory).

To this end, five binary gas systems are considered in the neighborhood of room temperature, since precise data are most easily obtained here. For the systems chosen, there is data on the three most important transport properties: the binary diffusion coefficient, mixture viscosity, and thermal conductivity.

There is also thermal diffusion data for some of these mixtures, but it is not analyzed or considered in detail here.

Systems have been selected so as to avoid undue complexity - ternary or multi-component mixtures are not examined, since the behavior of these systems should be understandable in terms of the behavior of binary systems. Similarly, systems involving interacting pairs of complex polyatomic (and possibly polar) molecules have been avoided.

In general we expect that the rigorous Chapman-Enskog theory for monatomic gases should serve rather well in describing the diffusivity and viscous properties of polyatomic gases. In the case of the thermal conductivity, however, a diffusive flux of internal energy (rotation, vibration, etc.) can make a substantial contribution to the heat flux, and must be considered. To this end the recent approximate theory of Mason and Monchick[1] for the heat conductivity of polyatomic and polar gases is invoked and extended (in an approximate fashion) to gas mixtures.

Specifically, the following mixtures have been examined:

Helium-Argon. Since this is a mixture of monatomic gases, obeying a spherically symmetric intermolecular force law, we should expect the rigorous Chapman-Enskog treatment to be fully applicable.

Helium-Hydrogen. A mixture of a monatomic gas with a diatomic molecule whose only internal energy is rotational. The transfer of energy between translation and rotation is slow for hydrogen, and requires several hundred collisions. Thus we should be able to treat the transport of translational and internal energies separately.

Hydrogen-Nitrogen. A mixture of diatomic gases which possess rotational

internal energy only. However, in examining the heat conductivity, the translation-rotation relaxation in nitrogen must be considered.

Argon-Ammonia. This mixture involves the interaction of a monatomic gas with a gas possessing an appreciable dipole moment.

Hydrogen-Ammonia. A mixture involving a polar molecule and a diatomic molecule.

In general, the emphasis here is on mixture properties; hence we try to tie to perfect predictions as to the properties of the pure components. To this end experimental thermal conductivities were corrected to be in accord with theory for the pure gases for those substances where the theory seems sound and near rigorous - helium, argon, and hydrogen. Experimental conductivities of nitrogen and ammonia have been assumed correct, and a rotational relaxation time has been assigned to make the experimental heat conductivity of nitrogen or ammonia correspond to the theory of Mason and Monchick[1].

THEORETICAL CONSIDERATIONS

In general, the transport properties of dilute gas mixtures involve terms characteristic of the interaction of the pure components with themselves, and also all possible pairwise interactions between unlike species. The self-interactions give rise to the properties of the pure species - not the subject of this discourse. The parameterwhich most directly characterizes the unlike interactions is the

Diffusion Coefficient. Such data have been analyzed in terms of the first Chapman-Enskog approximation (Reference 2, Equations 8.2 - 44), in most cases corrected by an average of the Kihara second approximations[3] calculated for the extremes of composition.

Mixture Viscosities have been analyzed in terms of the first Chapman-Enskog approximation (Reference 2, Equations 8.2 - 22). However, in most cases the input quantity η_{12} derived from diffusion has been corrected according to the third C-E approximation for pure gases.

Mixture Thermal Conductivities for monatomic gas mixtures have been obtained from the expression of Muckenfuss and Curtiss[4]. This expression was also used to compute the translational energy contribution to the conductivity for the other mixtures, but with an appropriate correction, as discussed below. The internal energy for mixtures has been computed according to Hirschfelder's[5] Eucken-type approximation for mixtures. Again with an appropriate correction. The use of the Muckenfuss-Curtiss equation for the translational heat conductivity with Hirschfelder's equation for the internal heat conductivity is justified assuming inelastic collisions rare. This assumption is necessary in order that the translational distribution function should not be unduly perturbed so that the translational conductivity is related to the viscosity or diffusion coefficients as in the case of the noble gases. It seems that this assumption is justified, except in the case of rotation-translation interchange which occurs every few collisions (say 2 - 20).

Mason and Monchick[1] have derived approximate expressions for the heat conductivity of pure polyatomic gases taking account of inelastic collisions. They find that the translational conductivity is less than that of a monatomic gas:

$$\lambda_{trans} = \lambda_{mon}\left[1 - \frac{2}{\pi}\left(\frac{5}{2} - \frac{\rho D}{\eta}\right)\frac{C_{rot}}{C_{trans}}\frac{1}{Z}\right] \qquad (1)$$

Here C_{trans}, C_{rot} are the translational and rotational portions of the heat capacity while Z is the collision number for rotational relaxation.

On the other hand, the internal energy contribution to the conductivity is enhanced:

$$\lambda_{int} = \lambda_{int,\,Modified\ Eucken}\left[1 + \frac{2}{\pi}\left(\frac{5}{2} - \rho\frac{D}{\eta}\right)\frac{C_{rot}}{C_{int}}\frac{1}{Z}\right] \qquad (2)$$

As yet no comparable theory for mixtures has been published. Consequently we use an extension to binary mixtures arrived at intuitively as follows: Assume that the mixture conductivity may be written

$$\lambda_{mix} = \lambda_{mix,\,Modified\ Eucken} + \Delta\lambda_1 + \Delta\lambda_2, \qquad (3)$$

where $\Delta\lambda_1$, $\Delta\lambda_2$ are the deviations attributable to the separate components. An approximate formula[6] for the conductivity of a monatomic gas mixture is

$$\lambda_{mix,mon} \cong \frac{\lambda_{1,mon}}{1 + \Psi_{12}\frac{\chi_2}{\chi_1}} + \frac{\lambda_{2,mon}}{1 + \Psi_{21}\frac{\chi_1}{\chi_2}}, \qquad (4)$$

where $\lambda_{1,mon}$, $\lambda_{2,mon}$ are the conductivities of the pure components, χ_1, χ_2 are mole fractions, and Ψ_{12}, Ψ_{21} are functions of the molecular weights and cross sections[6]. Hirschfelder's formula for the internal heat conductivity is of the same form as Equation (4), but with $\lambda_{int,\,Modified\ Eucken}$ in place of the λ_{mon} and the Ψ_{ij} replaced by Di/Dij. We assume that in a mixture the terms Z_i^{-1} must be replaced by

$$Z_{i,mix}^{-1} = \chi_i Z_i^{-1} + \chi_j Z_{ij}^{-1} \qquad (5)$$

Hence, from Equations (1), (2), (4), (5) and Hirschfelder's internal conductivity formula, we infer

$$\Delta\lambda_1 = \frac{2}{\pi}\left(\frac{5}{2}-\frac{\rho D}{\eta}\right)_1 C_{rot}\left[\frac{\chi_1}{Z_1}+\frac{\chi_2}{Z_2}\right]\cdot$$

$$\left[\frac{\lambda_{1,int,\,Modified\,Eucken}/C_{1,int}}{1+\frac{D_1}{D_{12}}\frac{\chi_2}{\chi_1}} - \frac{\lambda_{1,mon}/C_{1,trans}}{1+\Psi_{12}\frac{\chi_2}{\chi_1}}\right] \qquad (6)$$

The formula for $\Delta\lambda_2$ is obtained from Equation (6) by interchanging subscripts 1 and 2. Thus Equations (3) and (6) were used in analyzing data on mixtures involving nitrogen and ammonia.

As mentioned in the introduction, mixture conductivities were corrected so as to match theoretical values for helium, argon, and hydrogen. Hot wire thermal conductivity cells measure in essence the reciprocal of the conductivity. Thus if λ_1^*, λ_2^* are the experimentally reported conductivities of the pure components, and λ_m^* is the experimental mixture value, while λ_1, λ_2 are the theoretical conductivities of the pure components, then the corrected mixture conductivity λ_m is

$$\lambda_m^{-1} = \left(\lambda_m^{*-1} - \lambda_1^{*-1}\right)\frac{\lambda_2^{-1}-\lambda_1^{-1}}{\lambda_2^{*-1}-\lambda_1^{*-1}} + \lambda_1^{-1} \qquad (7)$$

RESULTS AND DISCUSSION

Helium-Argon. Diffusion coefficients for the helium-argon system were calculated assuming the exponential -6 potential with the force constants r_m = 3.505Å, α = 13.21, and ϵ/k = 33.4°K. The values for α and ϵ/k are taken from Mason[3]

while r_m was adjusted to fit the experimental data of Walker and Westenberg[7] at 25°C and 1 atmosphere. Comparison between theory and experiment are shown below:

TABLE 1

Helium Concentration	Diffusion Coefficient		$f^{(2)}_D$
	Experiment	Calculated	
0.0 (trace He)	0.754	0.750	1.0284
1.0 (Trace Argon)	0.725	0.729	1.0001

The factor $f^{(2)}_D$ is the ratio of the second Kihara approximation to the first Chapman-Enskog approximation. Of course the agreement between experiment and theory is not surprising, since r_m was chosen to obtain a good fit. The experimentally observed variation of diffusion coefficient with concentration is somewhat larger than that predicted theoretically, but it is not clear if this is a real effect or merely experimental error.

Viscosities for helium-argon mixtures were calculated again assuming the (exp-6) potential with force constants as follows:

TABLE 2

	r_m	α	ϵ/k
He - He	3.109	12.4	9.16
Ar - Ar	3.858	14.0	123.2
He - Ar (combining rules)	3.471	13.21	33.4
(Diffusion)	3.494	"	"

Again the α and ϵ/k are due to Mason[3]; for pure helium and argon r_m has been chosen to fit the recent, precise viscosity determinations of Iwasaki and Kestin[8]. Two values of r_m for the unlike interaction have been tested - the first from Mason's[3] empirical combining rules, and the second obtained from the experimental diffusion coefficients as follows:

$$r_{m\,\text{viscosity}}^2 = r_{m\,\text{Diffusion}}^2 / f_\eta^{(3)} \qquad (8)$$

Here $f_\eta^{(3)}$ is the correction for the third approximation for the viscosity of a <u>pure</u> gas (in this case 1.0062). This procedure lacks elegance, but is probably in the right direction. Theory and experiment are compared in Figure 1. Although the combining rule is not bad - the worst deviation is only 0.7% - the force constants derived from diffusion are clearly to be preferred. All in all, the agreement between the diffusion and viscosity data is heartening indeed, and seems to fully confirm the rigorous Chapman-Enskog theory. In fact the viscosity data are apparently of a quality such as to justify a proper calculation of the second approximation.

Thermal conductivities have been computed using the force constants of TABLE 2, ($r_m = 3.494$ for the He-Ar interaction); deviations of experiment[9] from theory are shown in Figure 2. Clearly the deviations are much more serious, amounting to 1.5% in the best case (the data of Von Ubisch), with errors approaching 10% at worst (Baker and Thornton). In an overall sense, the errors seem random rather than systematic, and there seems no cause for questioning the rigorous theory. Indeed, the computed conductivities are perhaps more reliable than the experimental values for this system.

Data on thermal diffusion in helium-argon mixtures has been analyzed by Mason[4]; agreement between theory and experiment seems satisfactory.

Helium-Hydrogen. Diffusion coefficients have been measured by Bunde[10] with what appears to be good precision and accuracy. Here they have been calculated assuming the exponential -6 potential with force constants $r_m = 3.289\text{Å}$, $\alpha = 13.22$, and $\epsilon/k = 18.27^\circ K$ (α and ϵ/k again from Mason[3]), with r_m adjusted to bracket Bunde's data at 25° and 1 atmosphere, as shown below:

TABLE 3

Range of Helium Concentration	Diffusion Coefficient Experiment	Calculated	$f^{(2)}_D$
0 (trace He)	- - -	1.543	1.0036
0 - 1.00	1.549	- - -	- - -
0.1490 - 1.000	1.550	- - -	- - -
0.4537 - 1.000	1.572	- - -	- - -
1.00 (trace H_2)	- - -	1.571	1.0221

Again the experimental variation of diffusion coefficient with composition seems somewhat larger than theory, but in this case the experimental compositions embrace a range of values, so that the comparision is less meaningful.

Viscosities and thermal conductivities for helium-hydrogen were calculated assuming force constants as follows:

TABLE 4

	r_m	α	ϵ/k
He - He	3.100	12.4	9.16
H_2 - H_2	3.337	14.0	37.3
He - H_2 (combining rules)	3.226	13.22	18.27
(Diffusion)	3.278	"	"

Again α and ϵ/k are from Mason, with r_m for the pure components adjusted so as to reasonably reproduce the experimental data for these gases, and r_m for the He - H_2 interaction has been adjusted according to Equation (8) with $f^{(3)}_{\eta} = 1.0063$. Theoretical viscosity calculations are compared with experiment[11] in Figure 3. Again the agreement between the diffusion and viscosity data is encouraging. (We should note, however, that the viscosities seem considerably less precise than in the helium-argon case, with deviations of 1 - 2% rather than 0.2%) Thermal conductivities computed according to the modified Eucken approximation for mixtures are compared with Barua's[12] experimental results in Figure 4. (The modified Eucken approximation assumes that the collision numbers Z_{H_2} and $Z_{H_2\text{-}He}$ are sufficiently large that the correction of Equation (6) is negligible.) Clearly these data seem to favor the empirical combining rules over unlike interaction force constants determined from diffusion coefficients or mixture viscosities. (It might be noted that these experimental data are in even closer accord with the theoretical analysis accompanying the data.)

However, from research on gas chromatography using thermal conductivity detectors it is well-known that small amounts of hydrogen <u>decrease</u> the heat conductivity of helium-hydrogen mixtures[13]. From this fact alone one can establish a maximum

slope to the conductivity-concentration plot at zero hydrogen concentration; these slopes are shown in Figure 4. Further, the data of Schmauch and Dinerstein[14] indicate that a mixture containing 17.3% hydrogen has the same thermal conductivity as pure helium; the consequences of this fact are indicated as an "X" on Figure 4.

Summarizing, it seems that the rigorous Chapman-Enskog theory properly represents the relation between diffusion coefficients and viscosities of helium-argon mixtures. It is not clear whether or not the modified Eucken approximation describes the thermal conductivities of these mixtures. Experimental data of enhanced accuracy are clearly required.

Thermal diffusion data have been analyzed in reference 2., p 584.

Hydrogen-Nitrogen. Diffusion coefficients have been measured by Bunde[10] using a Loschmidt-type cell; he obtained no information on concentration dependence. Values have been calculated assuming $r_m = 3.686\text{Å}$ (to fit the experiment), $\alpha = 15.56$ and $\epsilon/k = 58.97^\circ K$ (from Mason[3]). Comparisons at 1 atmosphere are as follows:

TABLE 5

	Hydrogen Concentration		
	0	0 - 1	1
Diffusion Coefficient	(calculate)	(Experiment)	(calculated)
25°C	0.7842	0.7835	0.7599
55°	0.9231	0.9079	0.8944
85°	1.071	1.052	1.037
$f^{(2)}_D$ (25°)	1.03220	- - -	1.00006

The correction for the second approximation was calculated at 25°C and then applied at the other temperatures as well - however, this factor should vary only very slightly with temperature.

Viscosities and thermal conductivities for hydrogen-nitrogen mixtures were calculated from the following force constants:

TABLE 6

	r_m	α	ϵ/k
$H_2 - H_2$	3.337	14.0	37.3
$N_2 - N_2$	4.011	17.0	101.2
$H_2 - N_2$ (combining rules)	3.690	15.56	58.97
$H_2 - N_2$ (Diffusion)	3.675	"	"

All values were taken from Mason[3] except for r_m from diffusion, again adjusted according to Equation (8) with $f^{(3)} = 1.0063$. In Figure 5 experimental viscosities[11,15] are compared with theoretical values calculated using $r_{m\,H_2-N_2}$ from diffusion. (Values from combining rules were only slightly different and hence are not shown.) The agreement between viscosity and diffusion data again seems satisfactory.

Thermal conductivities have been computed from Equations (3) and (6) with $Z_{H_2} = Z_{H_2-N_2} = \infty$ and $Z_{N_2} = Z_{N_2-H_2} = 7.2$. Theory and experiment[16] are compared in Figure 5. Also shown as a dashed line is the modified Eucken-type approximation corresponding to $Z_{N_2} = Z_{N_2-H_2} = \infty$. It would seem that the apparent discord between theory cannot be rationalized by the choice of larger collision numbers. Once again better experimental conductivity data would be most welcome.

Argon-Ammonia. Diffusion coefficients for this system have recently been determined by Srivastava and Srivastava[17]. They fitted their experimental data to the Lennard-Jones (12-6) potential with σ = 3.286Å and ϵ/k = 224.65. These values agree quite closely with estimates based on empirical combining rules using viscosity force constants of the pure components (σ = 3.271Å and ϵ/k = 221°K). Viscosities of argon-ammonia mixtures have been measured by Iwasaki, Kestin, and Nagashima[18]. These data are analyzed here by choosing viscosities of pure ammonia and argon and calculating the parameters characterizing the unlike interaction from the force constants derived from diffusion; results are shown in Figure 6. Clearly there is a serious discrepancy, with experimental viscosities as much as 7% larger than theory. It is possible to obtain agreement by arbitrarily taking σ = 3.072Å for the unlike interaction. This is equivalent to assuming that the measured diffusion coefficients are too low (should be some 14.4% larger). The lower portion of Figure 6 shows that accord between theory and experiment is much improved; errors are generally in the range of 1 - 2%. (It might be noted that these experimental data appear much less precise than the same authors' results[8] for the helium-argon system.)

In view of the serious discrepancy between the viscosity and diffusivity data we set about to measure the heat conductivities of these mixtures at 300°K. A hot-wire thermal conductivity was used and calibrated with helium and argon assuming conductivities of these gases to be 370.9 and 42.42 microcalories $cm^{-1}sec^{-1}{}^{o}K^{-1}$ respectively. (These values were obtained by considering literature data on the thermal conductivity and, more especially, viscosity of pure helium and argon[19].) Data were analyzed using Equations (3) and (6) after

correcting the coefficient for the diffusion of internal energy for the resonant exchange of rotational energy postulated by Mason and Monchick[1]. The collision number Z_{NH_3} was taken as 2.26 so as to fit the pure ammonia datum. An upper limit to the mixture conductivities was calculated assuming Z_{NH_3} - Ar = ∞ . Calculations are compared with experiment in Figure 7 - it is seen that the experimental conductivities are compatible with the viscosity results but incompatible with the diffusion coefficients. As a matter of fact by assuming $Z_{NH_3\text{-}Ar} = 1.4Z_{NH_3}$ the experimental data can be fitted almost perfectly. Thus the experimental diffusion coefficients[17] for the ammonia-argon system are clearly suspect and should be remeasured.

Hydrogen-Ammonia. Diffusion coefficients for this system have been measured by Bunde[10]; his results have been fit assuming a Lennard-Jones (12-6) potential with σ = 2.933Å and ϵ/k = 142.7. (The value of ϵ/k was obtained by Bunde from a fit of the first Chapman-Enskog approximation to his data over a range of temperature; σ has been deduced by applying an average correction for the second Kihara approximation to the diffusion coefficient to σ = 2.927 deduced by Bunde.) Comparisons at 1 atmosphere are as follows:

TABLE 7

	Hydrogen Concentration		
	0	0 - 1	1
Diffusion Coefficient	(calculate)	(Experiment)	(calculated)
25°C	0.7891	0.7830	0.7825
55°	0.9403	0.9426	0.9324
85°	1.1018	1.0933	1.0926
$f^{(2)}_D$ (25°)	1.00846	- - -	1.00005

The correction factor $f^{(2)}_D$ calculated at 25°C was applied at all three temperatures.

Viscosities and thermal conductivities for hydrogen-ammonia have been analyzed by assuming viscosity cross sections to exactly fit the viscosities of pure hydrogen and pure ammonia. Parameters characterizing the unlike interaction were taken from the combining rules of reference 2, p 600 (σ = 3.022, ϵ/k = 123°K) and also from diffusion (σ = 2.931, ϵ/k = 142.7). (The value of σ from diffusion has been corrected according to Equation 8 with $f^{(2)}_\eta$ = 1.0015.) Experimental viscosities of Trautz and Heberling[20] are compared with theory in Figure 8. The agreement is seen to be very good, with the diffusion force constants somewhat to be preferred. Thus it appears that the rigorous Chapman-Euskog theory for monatomic gases is quite acceptable even for mixtures involving polar gases.

Thermal conductivities were calculated from Equations (3) and (6) with the resonant correction of Mason and Monchick and assuming Z_{NH_3} = 2.11 so as to fit the experimental value of Gray and Wright[16] for pure ammonia at 25.3°C. A theoretical upper limit to the heat conductivities was calculated assuming $Z_{NH_3\text{-}Ar} = \infty$. Calculations are compared with experiment in Figure 8. Also shown as a dashed line is the modified Eucken-type approximation corresponding to $Z_{NH_3\text{-}H_2} = \infty$. In this case the correction for inelastic collisions it quite large, and amounts to about 15% for pure ammonia. It does not seem possible to draw any firm conclusions concerning the theory from Figure 8. The deviations are certainly larger than one would hope, but show a large random, rather than systematic, fluctuation. The agreement is scarcely worse that in the case of the H_2 - N_2 data of the same authors shown in Figure 5.

SUMMARY AND CONCLUSIONS

Experimental transport properties of five selected binary systems have been investigated in terms of the best theoretical formulations presently available. This analysis leads to the following tentative conclusions:

Regarding Theory:

1. The diffusion coefficients and viscosities of gas mixtures, including mixtures with polyatomic and polar molecules can be very well described in terms of the Chapman-Enskog theory, despite the fact that this theory is strictly rigorous only for monatomic gases.
2. There is no reason to doubt the validity of the Chapman-Enskog theory for the thermal conductivity of mixtures of monatomic gases; however, none of the experimental data are good enough to provide a really definitive test of the theory.
3. There are approximate methods which take account of the effects of internal energy on the heat conductivity of mixtures involving polyatomic and polar gases. These formulations seem promising, but again, experimental precision is too low to permit a meaningful evaluation of these methods.

Regarding Experimental Data:

1. There is already a large amount of mixture data of modest accuracy and precision (say errors larger than 1 - 2%). Further experimentation of this sort will do little to further our testing

and understanding of theory.

2. The very best determinations of diffusion coefficients and mixture viscosities seem good enough to provide really meaningful tests of theory, including the second Chapman-Euskog approximation for diffusion and perhaps viscosity as well. More data of this quality would be most welcome.
3. There is a crying need for thermal conductivity data of high accuracy and precision - little or none exists. It is proposed that such data may be acquired from precise measurements relative to the noble gases. Accurate conductivities for the monatomic gases can be computed from the best viscosity measurements by means of rigorous kinetic theory.

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FIG 1. Deviation of Theoretical from Experimental Viscosities for Helium – Argon Mixtures.

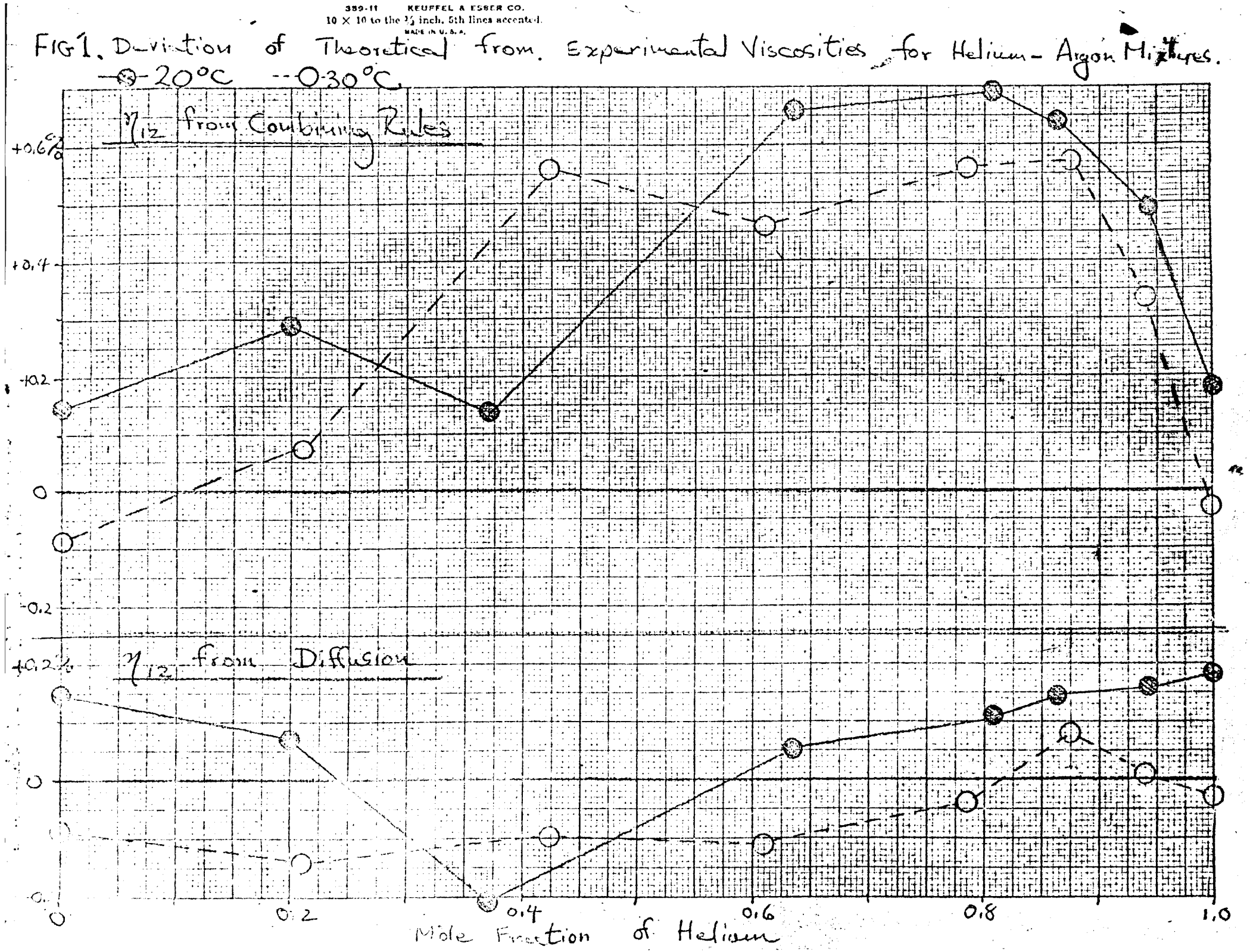

FIG 2. Deviation of Experimental from Theoretical Thermal Conductivities for Helium-Argon Mixtures

λ_{12} from Diffusion

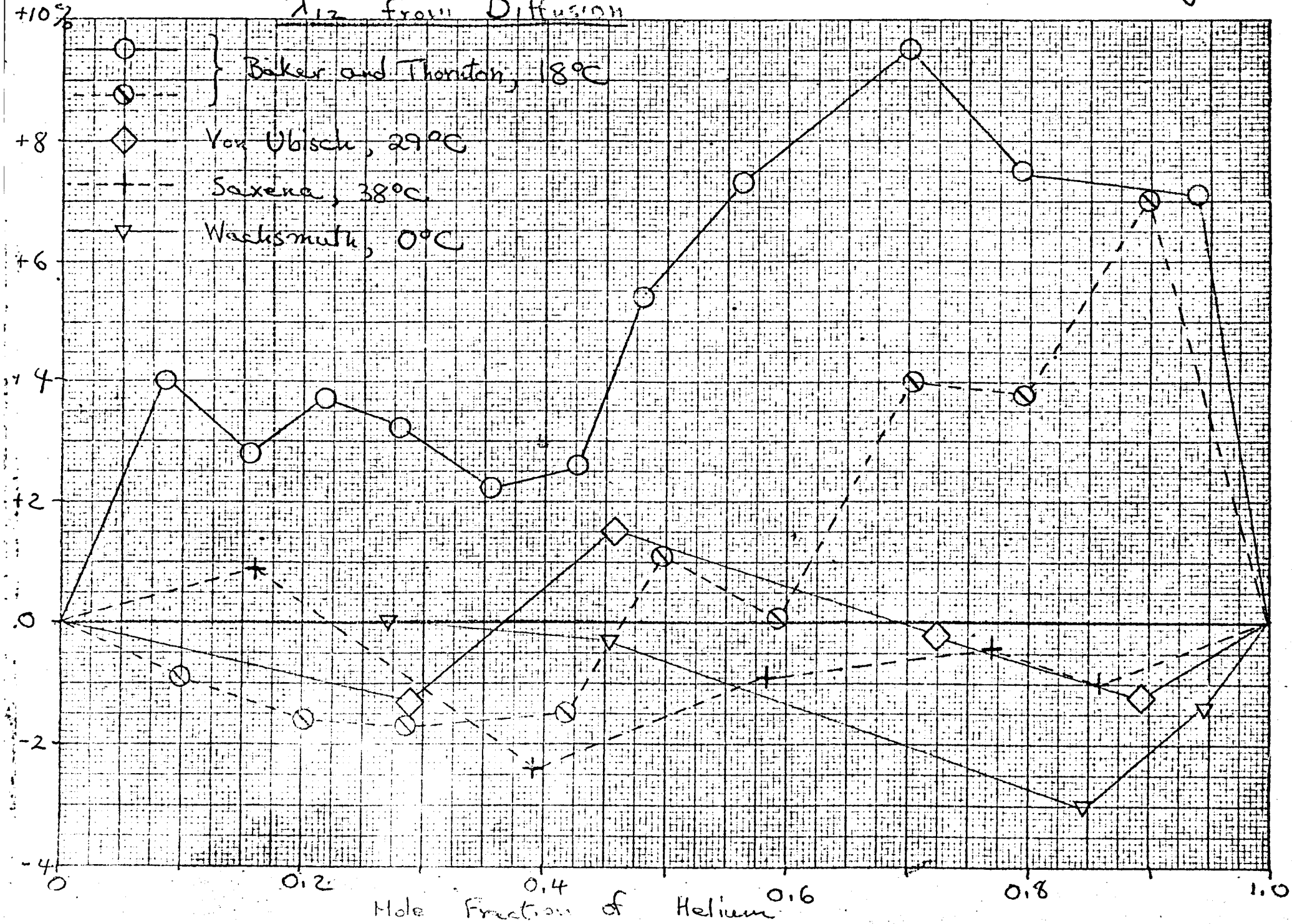

FIG. 3. Departure of Experimental from Theoretical Viscosities for Hydrogen-Helium Mixtures

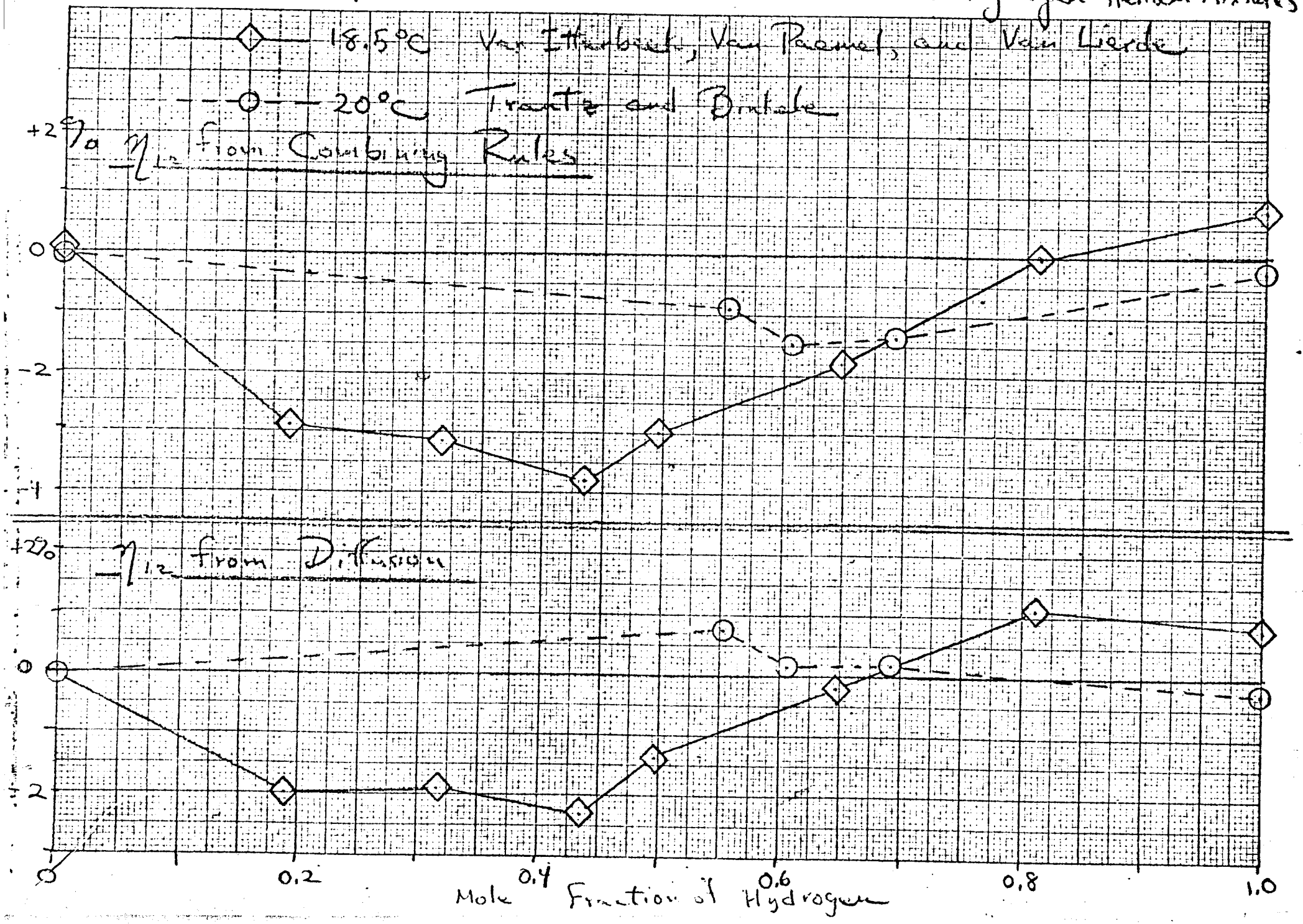

FIG. 4.

Departure of Experimental from Theoretical Thermal Conductivities for Hydrogen-Helium Mixtures

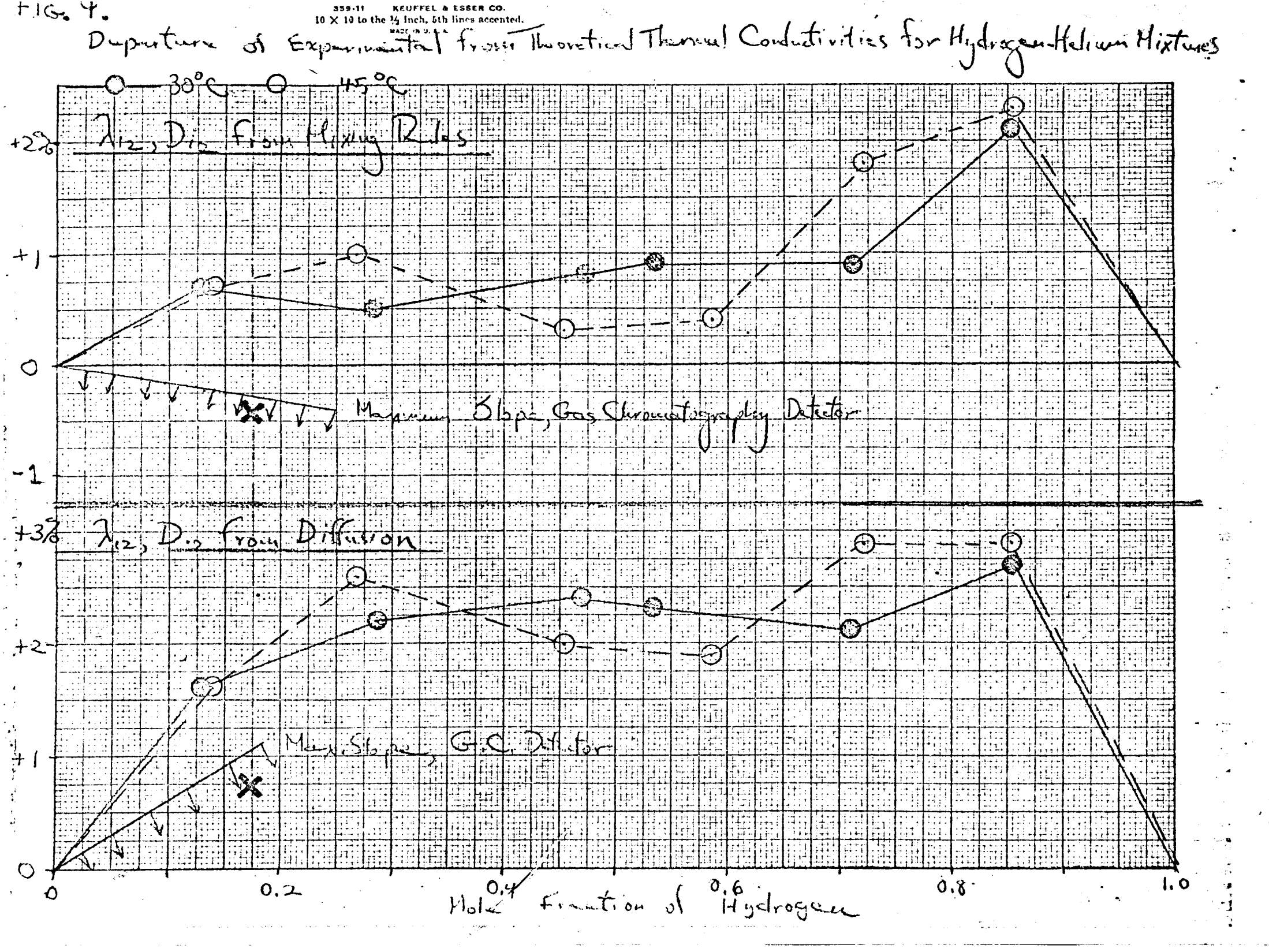

FIG 5. Deviation of Experimental from Theoretical Viscosities and Thermal Conductivities for Hydrogen–Nitrogen Mixtures (η_{12}, D_{12}, λ_{12} from Diffusion)

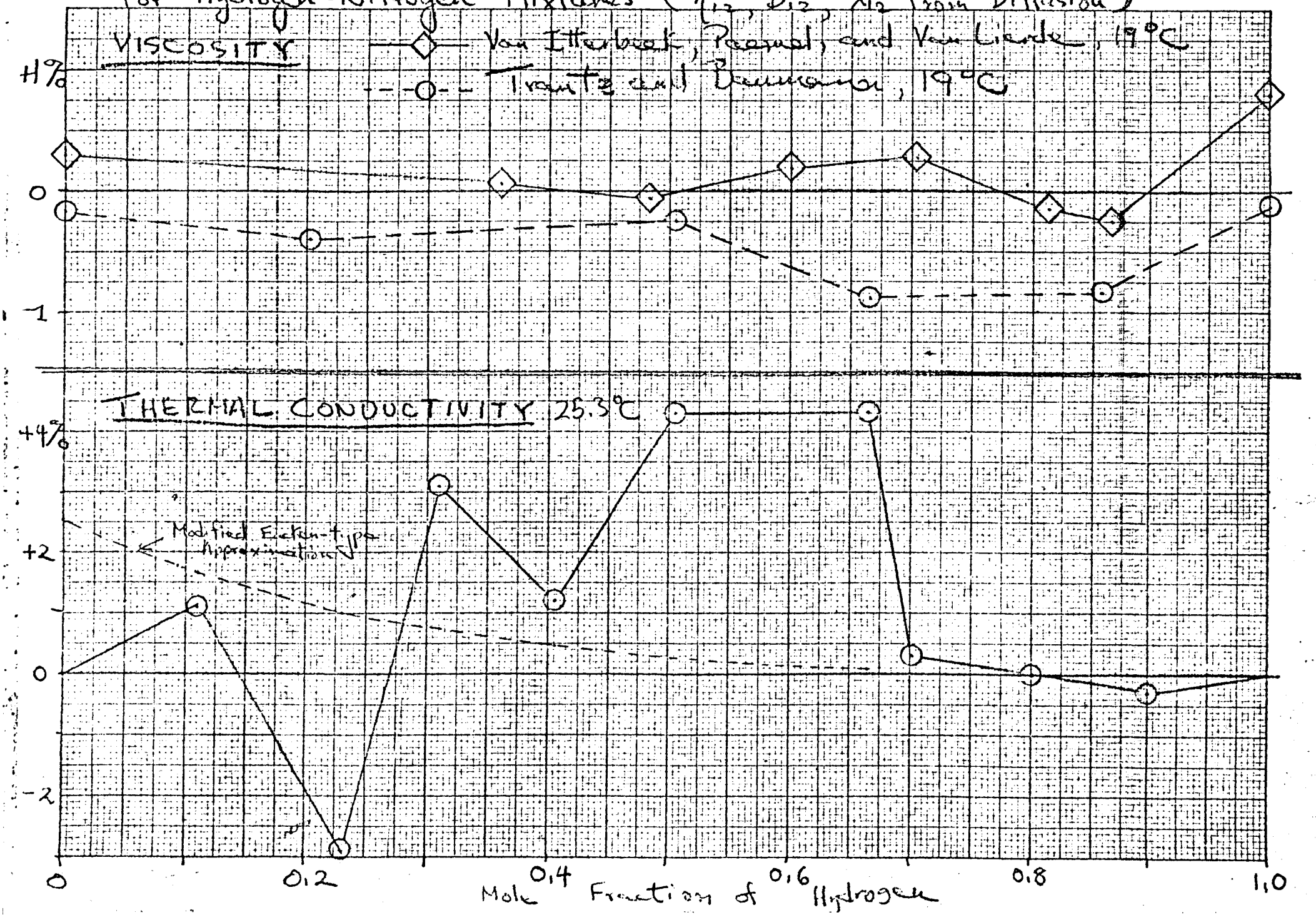

FIG 6. Deviation of Experimental from Theoretical Viscosities for Ammonia-Argon Mixtures

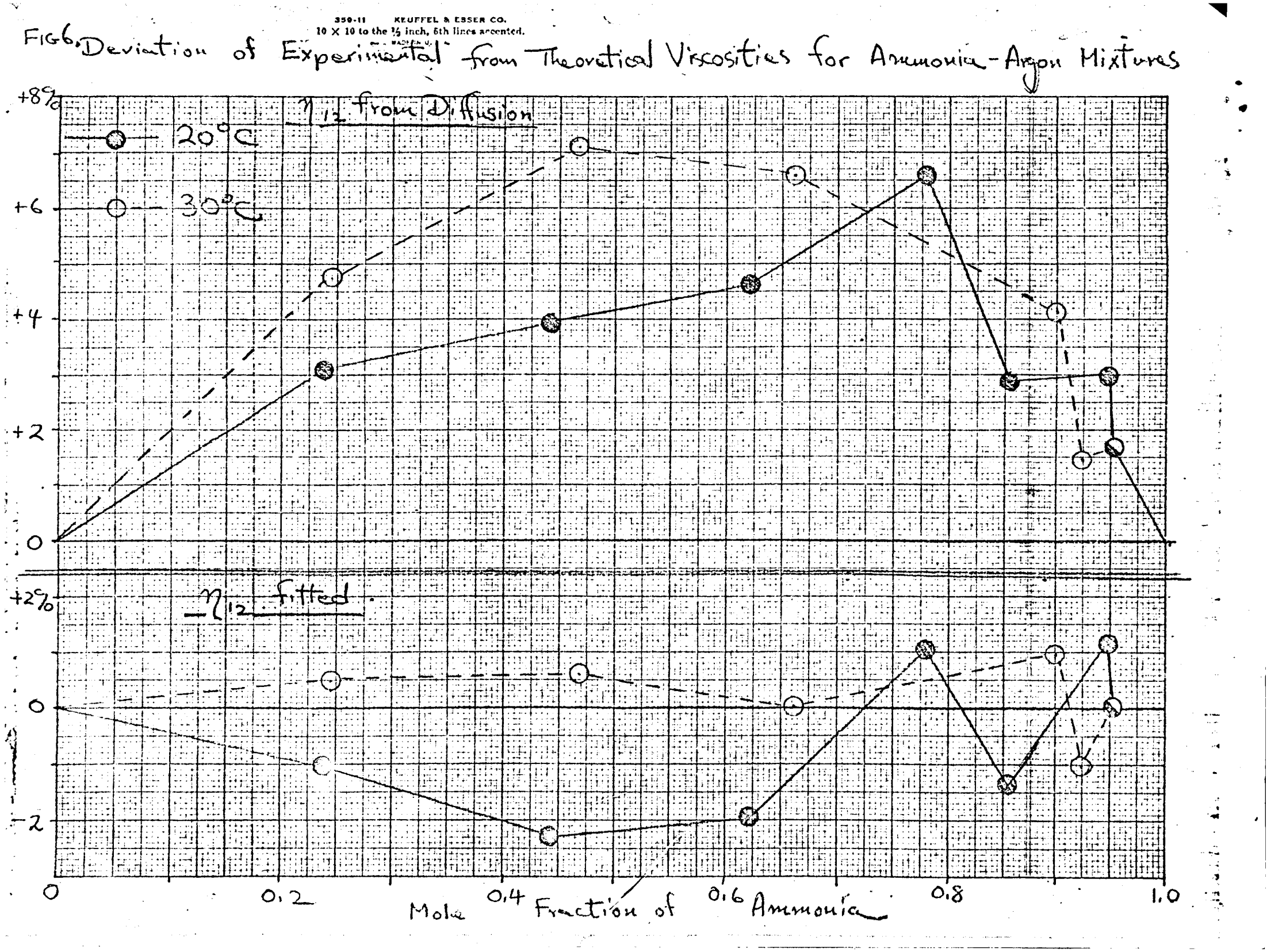

Fig. 7 Thermal Conductivity of Ammonia – Argon Mixtures

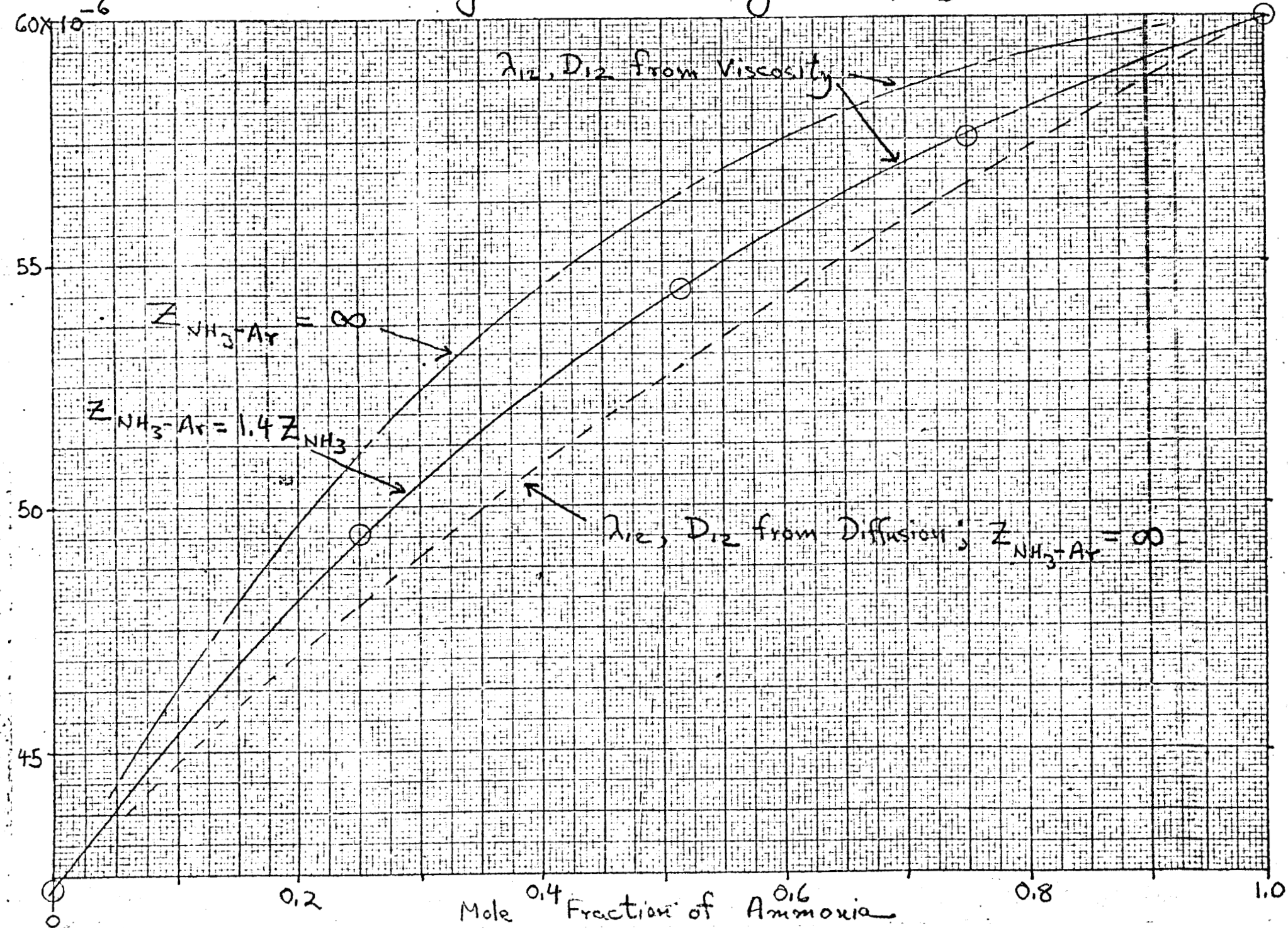

FIG. 8 Deviation of Experiment from Theory for Hydrogen – Ammonia Mixtures