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MEMORANDUM FOR: K. Kniel, Chief, Core Performance Branch, DSS

FROM: R. O. Meyer, Leader, Reactor Fuels Section, CPB, DSS

SUBJECT: MEETING SUMMARY OF ANS FISSION GAS GROUP

Enclosed is a meeting summary of the recent ANS-5.4 meeting on fission gas release. That summary is being sent to meeting attendees as committee correspondence. Several items of interest are noted below.

(a) An error in the statement of the low-temperature release model has been identified and corrected. The correct expression is given in the enclosed detailed summary.

(b) The model has been extended to cover cesium and tellurium fission products. The model previously covered noble gases and iodine. Release predictions for non-noble gases are only approximate.

(c) We hope to produce drafts of the standard and the supporting technical document at the committee's next meeting in February.

Ralph O. Meyer

Ralph O. Meyer, Leader
Reactor Fuels Section
Core Performance Branch
Division of Systems Safety

Enclosure:
As stated

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COMMITTEE CORRESPONDENCE

SOCIETY/COMMITTEE:

ANS-5.4

SUBJECT:

Fuel Plenum Gas Activity

AGENDA ITEM:
FILE NO.: N/A

ADDRESS CORRESPONDENCE TO:

R. O. Meyer

U.S. NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

DATE: NOV 21 1978

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Dear Group Members:

Enclosed are (1) a summary of the last Working Group Meeting, (2) an attendance list for the meeting, and (3) handouts presented at the meeting.

Sincerely,

Ralph O. Meyer

Enclosures:
(As stated)

cc: M. E. Remley, AI
M. Weber, ANS

Enclosure 1

Summary of ANS-5.4 Meeting

ANS Working Group 5.4 met on Thursday, November 8, 1978 at NRC Headquarters in Bethesda, Maryland.

An error in the statement of the low temperature release model had been pointed out in Chang Rim's letter of September 4, 1978. The derivation of this model was therefore reviewed in detail. The correct statement of this model is as follows. For stable isotopes,

$$F = 8.5 \times 10^{-8} \times Bu. \qquad (1)$$

For radioactive isotopes,

$$F = 2.0 \times 10^{-12} \times P/\lambda, \qquad (2)$$

where:

F is the release fraction. For radioactive species, F is the fraction of the non-decayed inventory just as it had been defined in the Booth model.

Bu is burnup in megawatt-days per metric ton of heavy metal.

P is specific power in megawatts per metric ton of heavy metal

λ is the decay constant in sec^{-1}.

In eq. (1) the uncertainty in the factor 8.5 is estimated to be +3.5 and -3.0. In Eq. (2) the uncertainty in the factor 2.0 is estimated to be +0.8 and -0.7.

The low temperature model was derived from the assumption of a knock-out mechanism. There is still some confusion as to whether a recoil mechanism is important. Larry Noble and the low-temperature subgroup will review this consideration. Since low-temperature releases are small and relatively large errors can be tolerated, we will probably not get deadlocked over this issue.

Bob Ritzman has extended the consideration of non-noble gases to cesium and tellurium. His handout is enclosed. The important equations in that handout are Eqs. 1 and 5, which relate the diffusion parameters for cesium and tellurium to the diffusion parameter for the noble gases.

Procedures for applying the high-temperature model were discussed. There was general agreement that enough radial nodes should be used such that nodal temperature differences would not exceed 100°C. This probably amounts to about 10 radial nodes as a minimum. No concensus was reached on axial noding, but one rather durable suggestion was as follows. Enough axial nodes should be used such that nodal temperature differences would not exceed 100°C with two exceptions: (1) this condition would not have to be met below 1000°C since releases would be too small to worry about

accuracy, and (2) any 5% axial section of the fuel (e.g., near a control blade tip) could be exempted from fine subdivision since it could not cause a large error in the rod-averaged gas release.

As the meeting neared an end, assignments were discussed. Ralph Meyer agreed to draft the (brief) standard. The (not-so-brief) support document would be similar to the April 1977 Status Report and organized as follows.

Section		Author
I	Introduction	R. O. Meyer
II	High Temperature Model	
	A Data	C. E. Beyer
	B Mathematics	L. D. Noble and C. S. Rim
	C Model Fitting	C. S. Rim
	D Non-Noble Isotopes	R. L. Ritzman
	E Isotopic Precursors	M. J. F. Notley
III	Low Temperature Model	
	A Data	R. A. Lorenz
	B Mathematics	L. D. Noble
	C Model Fitting	L. D. Noble
	D Non-Noble Isotopes	M. J. F. Notley

Draft sections of the support document should be mailed to all members by the authors prior to the next meeting. The next

meeting is tentatively planned for a central location (Dallas) in late February.

We worked until 9:00 p.m. without dinner and then adjourned.

Enclosure 2

ANS-5.4 Meeting Attendees

November 8, 1978

S. E. Turner	So. Science	Chariman
C. E. Beyer	HEDL	
B. J. Buescher	B&W	
R. J. Klotz	C-E	
R. A. Lorenz	ORNL	
W. Leech	W	
R. O. Meyer	NRC	
L. D. Noble	GE	
M. J. F. Notley	AECL	
R. L. Ritzman	SAI	
R. E. Mason	INEL	Guest
J. C. Voglewede	NRC	Guest

8 November 1978

To: ANS 5.4 Subcommittee Members

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Dear Group Members:

Enclosed is a report of work done to extend the ANS 5.4 high temperature model to cesium and tellurium. Comments are welcome.

Very truly yours,

RL Ritzman

Robert L. Ritzman

RLR/imp
Enc

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CESIUM AND TELLURIUM RELEASE FROM UO_2

R.L. Ritzman

The ANS-5.4 Working Group has made a considerable effort to formulate a method for calculating the release of fission product xenon and krypton from UO_2 reactor fuel[1]. An earlier paper described efforts to extend the procedure to fission product iodine[2]. The present paper outlines the results of further work to provide a procedure for the fission products, cesium and tellurium.

The procedure for cesium and tellurium corresponds exactly with the procedure that was used for iodine; i.e., specific data from the literature were used to develop diffusivity ratios for cesium relative to xenon and for tellurium relative to xenon. Then reference noble gas diffusion parameters (D') were multiplied by the ratio to obtain D' values for either cesium or tellurium for use in the ANS-5.4 model. The same four data sources were used to derive cesium/xenon and tellurium/xenon diffusivity ratios as were used to develop the iodine/xenon diffusivity ratios. These are essentially the only sets of experiments in which the release of all of the species were measured under the same conditions for each particular set. The next section presents the data analysis for the cesium/xenon diffusivity ratios. This is followed by a similar section for the tellurium/xenon diffusivity ratios. The last section summarizes all the fission product diffusivity ratio results and tabulates the D' values that are indicated by the applied method.

Cesium/Xenon Diffusivity Ratios

The first data source used was the work of Davies, Long, and Stanaway[3] who measured D'_{Cs}/D'_{Xe} ratios for a series of UO_2 sintered compacts, sintered spheroids, and fused spheroids of various densities at temperatures ranging from $1000^{o}C$ to $2150^{o}C$. A total of 19 determinations of use to this study, as reported by the investigators, are tabulated in Table 1. A similar set of measurements were performed by Parker, Creek, Barton, Martin, and Lorenz[4] with 93-94% dense UO_2 at temperatures ranging from $1400^{o}C$ to $1980^{o}C$. The D'_{Cs}/D'_{Xe} ratios obtained from 12 separate experiments given in this source are listed along with other pertinent data in Table 2. The other two data sources come from investigators who used diffusion theory to interpret their experimental work, and thus obtained values for the limiting diffusion coefficient and the activation energy in the classical Arrhenius equation. The parameter values given by Oi and Takagi[5,6] and by Parker[7] in his analysis of old ORNL data are given in Table 3.

The 31 data points given in Tables 1 and 2 were subjected to a least squares analysis in which $\ln (D'_{Cs}/D'_{Xe})$ was assumed to vary linearly as the reciprocal of the absolute temperature in accordance with a form of the Arrhenius equation. The derivation of the appropriate relationship as given in Reference (2) shows this line will have a slope corresponding to $(Q_{Xe}-Q_{Cs})/R$ and an intercept value at $1/T = 0$ corresponding to $\ln(D'^{o}_{Cs}/D'^{o}_{Xe})$. The result of the least squares analysis is displayed in Figure 1 along with the 31 experimental diffusivity ratio values and two other lines which correspond to the ratio of Arrhenius expressions published by Oi and Takagi and by Parker.

Table 1. Diffusivity Ratios Obtained for UO_2 in Reference (3)

Sample Type	Density g/cm^3	Surface Area cm^2/g	Temperature ^oC	D'_{Cs}/D'_{Xe}
Sintered	10.3	140	1000	5.76
Compact	10.3	140	1000	26.0
Compact	10.3	140	1200	1.96
Compact	10.3	140	1400	1.96
Compact	-	∿100	1600	5.29
Compact	10.3	10	1600	39.7
Compact	10.8	5	1600	0.64
Compact	10.7	7	2000	4.84
Compact	10.7	7	2050	0.83
Compact	10.7	7	2150	0.141
Sintered	-	25	1200	1.0
Spheroids	-	25	1400	21.2
Spheroids	-	25	1600	0.36
Spheroids	-	25	1600	6.25
Spheroids	-	25	1600	0.64
Spheroids	-	25	1600	0.36
Spheroids	-	25	1400	6.25
Fused	10.6	77	1200	1.0
Spheroids	10.6	77	1600	3.24

Table 2. Diffusivity Ratios Calculated From Release Data for UO_2 in Reference (4)

Sample Type	% Theo. Density	Temperature °C	Fraction Released in 5.5 Hours Cs	Fraction Released in 5.5 Hours Xe	D'_{Cs}/D'_{Xe}
PWR-UO_2	93-94	1515	0.014	0.013	1.16
PWR-UO_2	93-94	1610	0.017	0.027	0.396
PWR-UO_2	93-94	1710	0.027	0.026	1.08
PWR-UO_2	93-94	1800	0.032	0.037	0.748
PWR-UO_2	93-94	1900	0.086	0.097	0.786
PWR-UO_2	93-94	1980	0.15	0.12	1.56
EGCR-UO_2	97	1400	0.026	0.008	10.6
PWR-UO_2	93-94	1400	0.005	0.005	1.0
PWR-UO_2	93-94	1400	0.21	0.061	11.9
EGCR-UO_2	97	1610	0.12	0.026	21.3
PWR-UO_2	93-94	1610	0.20	0.060	11.1
PWR-UO_2	93-94	1780	0.032	0.037	0.748

Table 3. Arrhenius Equation Parameters for Cesium and Xenon from References (5), (6), and (7)

Reference	Specie	Pre-Exponential Factor	Activation Energy
(5)	Xe	$3.0 \times 10^{-3} cm^2/sec$	63.0 Kcal/g-atom
(6)	Cs	$8.5 \times 10^{-9} cm^2/sec$	6.1 Kcal/g-atom
(7)*	Xe	$2.32 \times 10^{2}\ sec^{-1}$	79.6 Kcal/g-atom
	Cs	$1.37 \times 10^{0}\ sec^{-1}$	52.3 Kcal/g-atom

*Parameters taken from values uncorrected for the "burst effect".

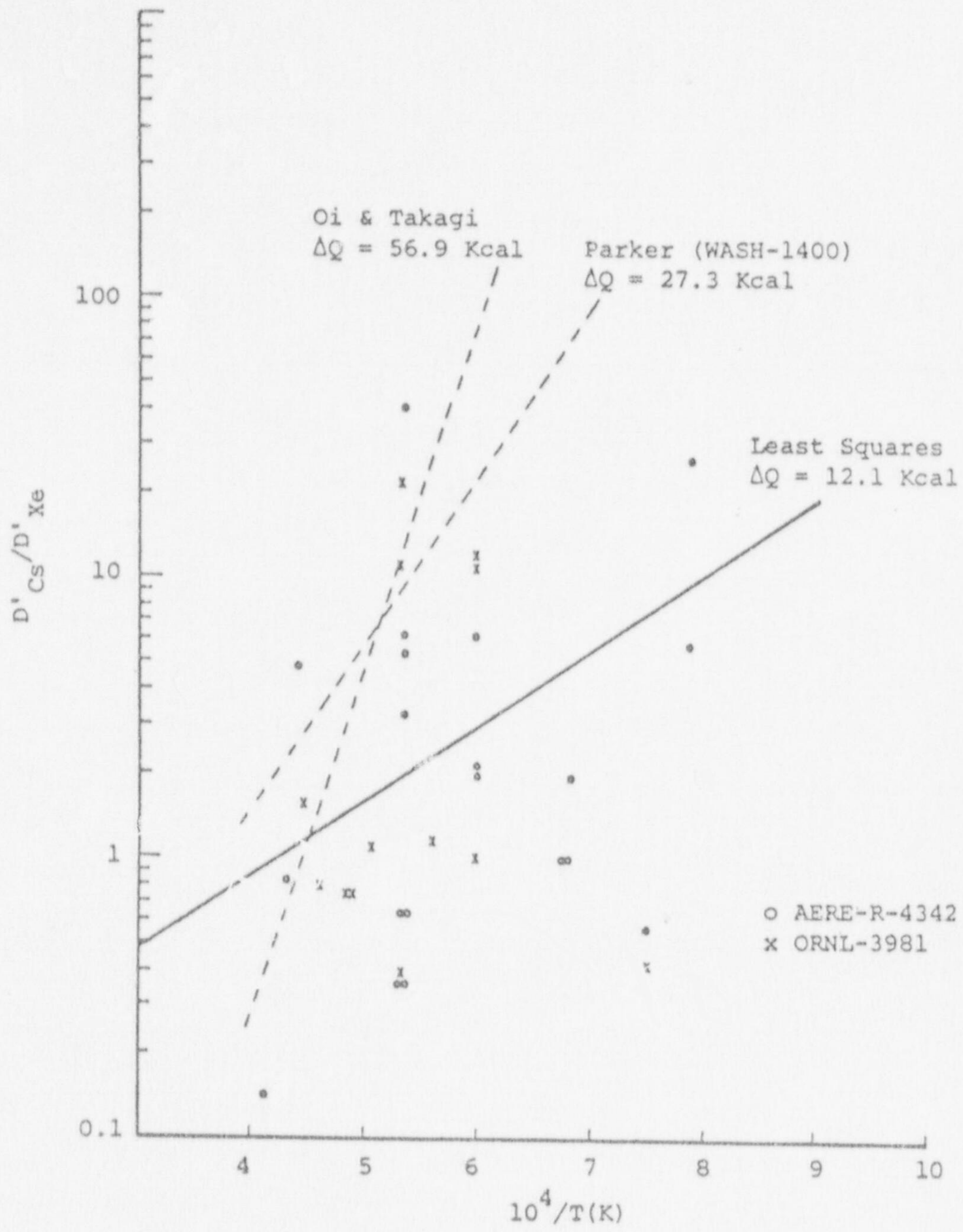

Figure 1. Diffusivity Ratio Data and Curves for Cesium/Xenon

Inspection of Figure 1 reveals considerable scatter in the 31 data points. The least squares analysis resulted in a positive value for $\Delta Q = (Q_{Xe} - Q_{Cs})$ of 12.1 Kcal/g-atom which is considerably less than the value of 56.9 Kcal/g-atom obtained from the Oi and Takagi expressions and the value of 27.3 Kcal/g-atom obtained from the Parker expressions. The complete equation for the least squares line is:

$$(D'_{Cs} / D'_{Xe}) = 7.58 \times 10^{-2} e^{12100/RT} \tag{1}$$

Likewise the complete equations for the Oi and Takagi line and the Parker line are respectively:

$$(D'_{Cs} / D'_{Xe}) = 2.83 \times 10^{-6} e^{56900/RT} \tag{2}$$

and

$$(D'_{Cs} / D'_{Xe}) = 5.89 \times 10^{-3} e^{27300/RT} \tag{3}$$

It is noteworthy that all three lines indicate a positive value for the difference in activation energies, but the wide range of values is rather disappointing. It does appear that neither the Oi and Takagi line nor the Parker line provide a very good fit to the data points in Figure 1. On this basis, and to maintain consistency with the approach used for iodine, it was decided to rely on the least squares fit to indicate the diffusivity of cesium relative to xenon in UO_2. The data scatter and the disagreement in ΔQ values diminish the confidence that can be attached to applications involving this approach, but it represents the best that can be done with presently available information. Multiplying Equation (1) above by the reference equation for xenon diffusion in UO_2,

the following equation, which estimates cesium diffusion, is obtained.

$$D'_{Cs} = 1.68 \times 10^{-4} e^{-37600/RT} \quad (4)$$

Use of this equation in conjunction with the diffusion model and acceptable fuel thermal performance models thus provides a means to estimate high-temperature release of cesium from operating reactor fuel.

Tellurium/Xenon Diffusivity Ratios

From the work of Davies, Long, and Stanaway[3], a set of 17 determinations of D'_{Te}/D'_{Xe} ratios useful to the present study were made. These ratios and associated data are listed in Table 4. The fission product release studies of Parker, Creek, Barton, Martin, and Lorenz[4] provide 10 additional D'_{Te}/D'_{Xe} ratio values which are listed along with other relevant data in Table 5. In Table 6 the Arrhenius equation parameters as determined by Oi and Takagi[5,6] and by Parker[7] for xenon and tellurium diffusion from UO_2 are given. The values in these three tables constitutes the body of data used here to obtain a diffusivity ratio expression for the tellurium/xenon combination.

The 27 tellurium/xenon diffusivity ratio data points given in Tables 4 and 5 were subjected to a least squares analysis of the same type that was performed for the cesium/xenon and the iodine/xenon data. The result of the analysis is shown by the solid line in Figure 2 along with the 27 experimental data points and two other dashed lines which correspond

Table 4. Diffusivity Ratios Obtained for UO_2 in Reference (3)

Sample Type	Density g/cm^3	Surface Area cm^2/g	Temperature °C	D'_{Te}/D'_{Xe}
Sintered	10.3	140	1000	7.84
Compacts	10.3	140	1000	24.0
Compacts	10.3	140	1200	15.2
Compacts	10.3	140	1400	7.29
Compacts	10.3	19	1600	100.
Compacts	10.8	5	1600	121.
Compacts	10.16	4	1300	44.9
Compacts	10.7	7	2000	13.0
Sintered	-	25	1200	6.76
Spheroids	-	25	1400	39.7
Spheroids	-	25	1600	110.3
Spheroids	-	25	1600	196.
Spheroids	-	25	1600	441.
Spheroids	-	25	1600	441.
Spheroids	-	103	1400	10.9
Fused	10.6	77	1200	32.5
Spheroids	10.6	77	1600	259.

Table 5. Diffusivity Ratios Calculated from Release Data for UO_2 in Reference (4)

Sample Type	% Theo. Density	Temperature °C	Fraction Released in 5.5 Hours		D'_{Te}/D'_{Xe}
			Te	Xe	
PWR-UO_2	93-94	1515	0.029	0.013	4.98
PWR-UO_2	93-94	1610	0.12	0.027	19.75
PWR-UO_2	93-94	1710	0.20	0.026	59.2
PWR-UO_2	93-94	1800	0.21	0.037	32.2
PWR-UO_2	93-94	1400	0.039	0.008	23.8
EGCR-UO_2	97	1400	0.008	0.008	1.0
PWR-UO_2	93-94	1400	0.012	0.005	5.76
PWR-UO_2	93-94	1400	0.16	0.061	6.88
EGCR-UO_2	97	1610	0.12	0.026	21.3
PWR-UO_2	93-94	1780	0.21	0.037	32.2

Table 6. Arrhenius Equation Parameters for Tellurium and Xenon from References (5), (6), and (7)

Reference	Specie	Pre-Exponential Factor	Activation Energy
(5)	Xe	$3.0 \times 10^{-3} cm^2/sec$	63.0 Kcal/g-atom
(6)	Te	$6.6 \times 10^{-3} cm^2/sec$	70.0 Kcal/g-atom
(7)*	Xe	$2.32 \times 10^{2} sec^{-1}$	79.6 Kcal/g-atom
	Te	$5.33 \times 10^{1} sec^{-1}$	66.6 Kcal/g-atom

*Parameters taken from values uncorrected for the "burst effect".

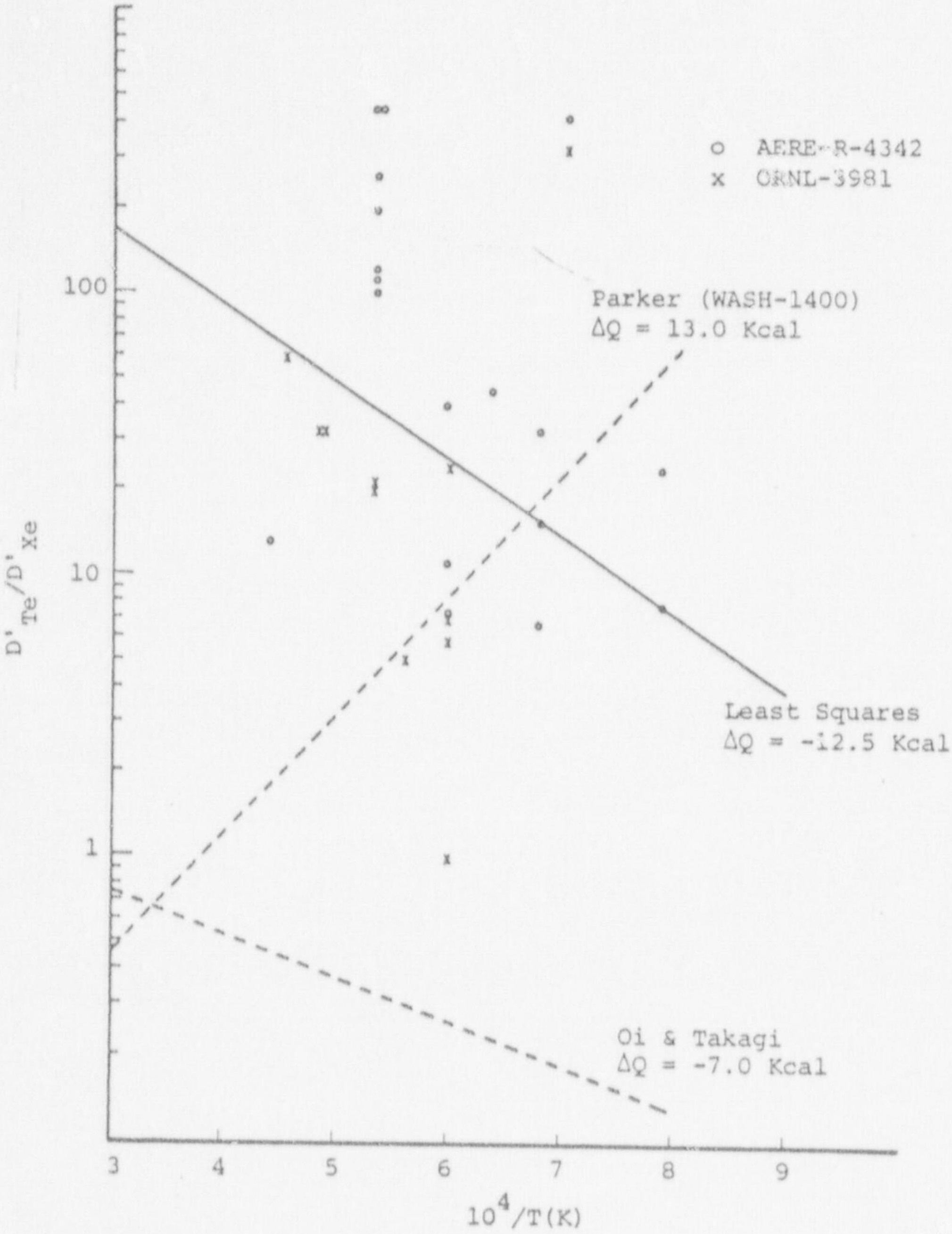

Figure 2. Diffusivity Ratio Data and Curves for Tellurium/Xenon

to the ratio of Arrhenius expressions published by Oi and Takagi and by Parker. Inspection of Figure 2 reveals considerable scatter in the 27 data points. The least squares analysis resulted in a negative value for $\Delta Q = (Q_{Xe}-Q_{Te})$; i.e., -12.5 Kcal/g-atom in contrast to the positive values which had been obtained for both $(Q_{Xe}-Q_{Cs})$ and $(Q_{Xe}-Q_{I})$. The ratio of Arrhenius expressions from Oi and Takagi also yielded a negative ΔQ but the value was only -7.0 Kcal/g-atom. However, the ratio of Arrhenius expressions from Parker gave a positive ΔQ value of 13.0 Kcal/g-atom. The complete equation for the least squares line is:

$$(D'_{Te}/D'_{Xe}) = 1.10 \times 10^{3} e^{-12500/RT} \quad (5)$$

Likewise the complete equations for the Oi and Takagi line and the Parker line are respectively:

$$(D'_{Te}/D'_{Xe}) = 2.2 e^{-7000/RT} \quad (6)$$

and

$$(D'_{Te}/E'_{Xe}) = 2.29 \times 10^{-1} e^{13000/RT} \quad (7)$$

The considerable disagreement between ΔQ values and the wide distances between equation lines in Figure 2 indicate that tellurium behavior in UO_2 is not very well defined. In any case, however, it is apparent that the Oi and Takagi line is a poor fit for the data points in Figure 2. The positive slope of the Parker line is probably due to the fact that it was derived from a collection of data which did not include the measurements of Davies, Long, and Stanaway. Therefore, it is not really applicable to the present analysis. This leaves

the least squares line as the best available indicator of the diffusivity of tellurium relative to xenon in UO_2. Multiplying Equation (5) above by the reference equation for xenon diffusion in UO_2 produces the following equation for tellurium:

$$D'_{Te} = 2.43e^{-62200/RT} \qquad (8)$$

Use of this equation in conjunction with the diffusion model and acceptable fuel thermal performance models provides a means for estimating the high-temperature release of tellurium from operating reactor fuel.

Summary of Diffusivity Ratio Results

The diffusivity ratio approach has been used to derive diffusion parameter expressions for fission products iodine, cesium, and tellurium from a limited body of experimental data. Each analysis considered approximately 30 data points obtained from two principal sources. Although the data were quite scattered, least squares analyses were performed to obtain Arrhenius type expressions for $D'_{Fission\ Product}/D'_{Xenon}$ which were then multiplied by the ANS-5.4 reference equation for D'_{Xenon} in UO_2 fuel to yield the diffusion parameter expressions for the other fission products. The complete set of diffusion parameter expressions are:

$$D'_{Xe} = 2.21 \times 10^{-3} \exp(-49700/RT) \qquad (9)$$

$$D'_{I} = 1.26 \times 10^{-3} \exp(-40800/RT) \qquad (10)$$

$$D'_{Cs} = 1.68 \times 10^{-4} \exp(-37600/RT) \qquad (11)$$

$$D'_{Te} = 2.43 \exp(-62200/RT) \qquad (12)$$

The temperature range of applicability for these expressions extends from about 1000°C to about 2200°C. The expression for xenon is that developed by the ANS-5.4 Working Group as described in Reference (1).

A plot of the four diffusion parameter expressions is given in Figure 3 to provide a visual comparison of the results of the study. Except at rather high temperatures the indicated D'values for the different fission products lie within roughly a factor of 20 of one another. It should be noted again that these results are based on a limited set of data and a particular interpretation of those data. While the results probably represent the best that can be done with the currently available information, they should be applied with caution. On this basis it would be advisable to pursue verification studies if the results are considered for incorporation in a standard procedure for predicting volatile fission product release from operating reactor fuel.

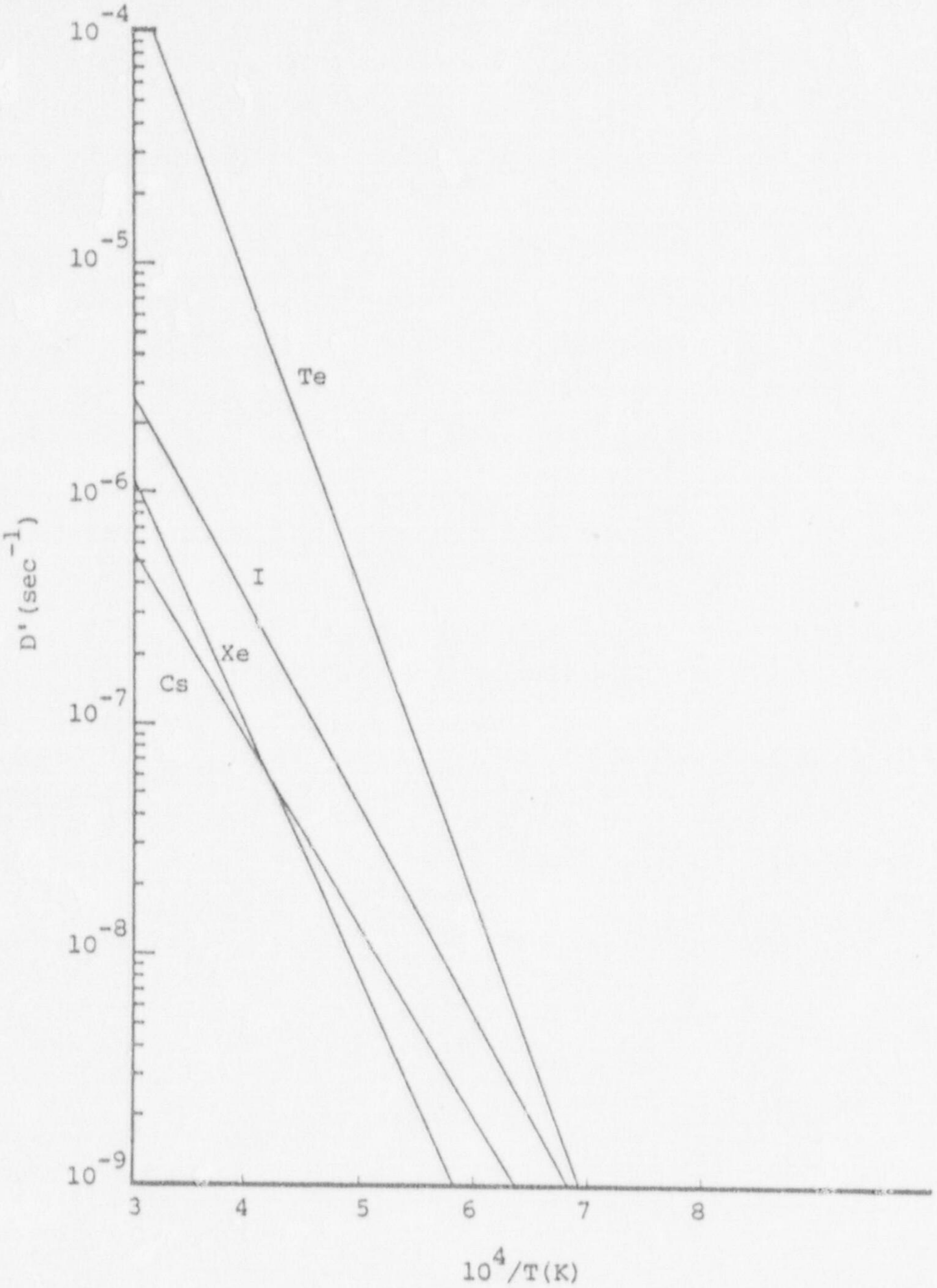

Figure 3. Diffusion Parameters as a Function of 1/T for Xenon, Iodine, Cesium, and Tellurium

REFERENCES

1. "Status Report - ANS 5.4 Fuel Plenum Gas Activity", (218) (Fission Product Release from UO_2 Fuel), S.E. Turner, Chairman, April, 1977.

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3. D. Davies, G. Long, and W.P. Stanaway, "The Emission of Volatile Fission Products form Uranium Dioxide", British Report AERE-R-4342, June, 1963.

4. G.W. Parker, C.E. Creek, C.J. Barton, W.J. Martin, and R.A. Lorenz, "Out-of-Pile Studies of Fission-Product Release from Overheated Reactor Fuels at ORNL, 1955-1965", ORNL-3981, July, 1967.

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7. G.W. Parker, "Calculation of Gap Release of Radioactive Fission Products", Appendix C of Appendix VII of WASH-1400, October, 1975.