NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Technical Report 32-1077

Estimates of Doppler Coefficients for In-Pile Thermionic Reactor Materials

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Foreword

This work represents one phase of studies on in-pile thermionic-space-reactor power-plant dynamics and stability. This work is conducted by the Energy Sources Group of the JPL Propulsion Research and Advanced Concepts Section for the National Aeronautics and Space Administration. The author, a professor in the Nuclear Engineering Department at the University of Arizona, was retained as a consultant from July 18 to August 26, 1966, to investigate doppler coefficients associated with in-pile thermionic reactors. This report summarizes the pertinent points of the author's investigations.

Acknowledgments

The author gratefully acknowledges the cooperation of C. Till of Argonne National Laboratory and S. Carpenter, R. Tuttle, T. Springer, and L. Mountford of Atomics International. These investigators have kindly consented to the publication here of recent doppler measurements which are as yet unreported in the open literature. The author acknowledges, also, the support provided by the Jet Propulsion Laboratory (JPL) and its personnel.

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Abstract

Recent experimental data have been used to estimate temperature coefficients of reactivity due to doppler broadening of resonances in materials of interest for compact in-pile thermionic-reactor-core concepts. Materials considered are U-235, U-238, and the refractory metals, tungsten, tantalum, molybdenum, and niobium. Equations giving the magnitude and temperature dependence of the doppler coefficient are given for each of these elements. Estimates are given of the possible error in the doppler coefficient introduced by extrapolating the experimental data to the higher temperatures of interest in thermionic reactor concepts. A comparison of this method with the usual theoretical method is made for the Fermi reactor.

Estimates of Doppler Coefficients for In-Core Thermionic Reactor Materials

I. Introduction

Concern for the stability of compact in-core thermionic fast reactors (Ref. 1) has led to an investigation of anticipated temperature coefficients of reactivity arising from the doppler broadening of resonances in fuel, emitter, collector, and cladding materials. Of particular concern is the magnitude of the positive doppler coefficient of U-235. A thermionic-reactor concept typically uses a high degree of enrichment, and a positive coefficient in the fuel can conceivably lead to unstable reactor operation.

The detailed theoretical calculation of doppler coefficients for fast reactors is well known to be a relatively difficult and intricate task (Refs. 2, 3, 4). Even in very hard spectrum reactors, a considerable portion of the doppler coefficient may be attributable to the lower lying resonances and, consequently, accurate specification of the reactor spectrum is very important. The calculations are further complicated by the fact that resonance parameters are completely unknown at the higher energies; that is to say, the resonances are experimentally unresolved. The analyst is thus faced with the task of estimating the higher energy resonance parameters, a process which normally is accomplished by an extrapolation of a statistical description of the resolved (lower energy) resonances.

The fissile elements introduce the further complication of having small level spacings between resonances. This leads to interference between resonances, that is, an overlapping effect which tends to smear or wash out individual resonances. Because the fissile elements may give rise to a positive component in the prompt temperature coefficient (in highly enriched systems), the evaluation of the doppler coefficient for the fissile material takes on added importance.

There are other complicating intricacies involved with doppler coefficient calculations. To mention just a few, the actual heterogeneous arrangement of fuel, cladding, and coolant can lead to self-shielding of the resonances.

Interference between resonances of different isotopes, particularly between a fissile and non-fissile isotope, can cause a decrease in the doppler coefficients from the values calculated for each isotope separately. Large temperature gradients, such as are experienced in oxide fuels under high density conditions, can lead to coefficients different from those calculated, assuming some space-average temperature. Fortunately, these effects have been calculated for reactors with softer spectra (which enhances the effects), and all have been found to be small (see, for example, Ref. 5).

For compact in-core thermionic reactor concepts, the neutron spectrum is typically characterized by median fission energies of the order of 100 kev or higher. Even so, it is the resonances in the lower kev range (1–50 kev) which contribute most to the doppler effect (Ref. 2). Consequently, the magnitude and to a lesser extent the shape of the low energy flux spectrum tail is much more important for our purposes than is the overall spectral behavior. Both magnitude and shape of the tail may vary considerably from one thermionic reactor concept to another, depending on the amount of moderating material present in the core or reflector. Thus, the complication of being able to specify the reactor spectrum accurately and in considerable detail within the resonance region can not be ignored in any attempt to calculate by analytical methods the doppler coefficients.

For thermionic reactor stability analyses of a general nature, such detailed calculations of the doppler coefficients are neither warranted nor desired. They are unwarranted if the intention of the study is general; that is, a typical reactor concept, as opposed to a detailed specific concept, is to be represented. In addition, the coefficients often need to be specified only within some error bounds; i.e., a high degree of accuracy is usually unnecessary. Finally, we should remark that the general status of calculational methodology is still under investigation and in the case of doppler coefficient calculations, this factor can lead to relatively large disparities (Refs. 6, 7, 8). In some cases, good agreement with experiment is achieved while in others serious discrepancies arise, all experiments being analyzed by the same methodology and by highly competent investigators.

For these reasons a rapid method of estimation, given the particular reactor concept, is desired. The goal of this report is to provide such estimates for typical materials which may be employed within in-core thermionic reactors.

II. Analysis

A. Basis

The calculational difficulties outlined in the previous section can be circumvented by using recent experimental measurements of doppler coefficients. These measurements have been made, on small samples of material, in fast critical assemblies possessing reasonably "hard" spectra in their test sections (Refs. 9, 10). Typically, the median fission energy and the peak of the neutron flux in the test section are in the 100–300 kev range, an energy range characteristic also of the thermionic reactors (Ref. 11). The strong spectral dependence is effectively normalized out simply by dividing the doppler coefficients by the reactivity worth of the sample as measured in the same assembly. The validity of this procedure will be demonstrated subsequently. The doppler coefficients applicable to a given thermionic reactor concept are then obtained simply by multiplying this doppler-coefficient-per-unit-reactivity-worth ratio by the average reactivity worth of the material of interest in the concept of interest. This latter value may be calculated or may be estimated from experiment as will be demonstrated.

In addition to the ease and simplicity of this correlation technique, there is the added benefit of having incorporated the actual measured temperature-dependence of the doppler coefficient rather than having to rely on the theoretical values. The latter have now been shown to be in error at least for U-238, U-235, tungsten, and tantalum. The measurements on molybdenum and niobium are at present inconclusive.

The following qualitative arguments may be presented as a basis for the correlation technique. The vindication of these arguments actually rests on the results of several experiments, however.

Doppler experiments are conducted on small samples of material placed within a test section of a large critical assembly and the samples are subsequently heated. Two reactivity measurements are involved: the first is the reactivity worth of the sample in the critical assembly at room temperature relative to a void; the second is the reactivity change associated with heating the sample. The environment that is the test section neutron flux spectrum remains the same for both measurements.

If the reactivity worth of the sample is denoted by ρ, then

$$\rho = \int \sigma_x(u)\, w(u)\, du \qquad (1)$$

where $\sigma_x(u)$ is the interaction cross section (absorption or fission) of the sample and $w(u)$ is an appropriate weighting function proportional to the neutron flux expressed in lethargy units u. The doppler coefficient is simply the temperature derivative of ρ, all other variables, such as density, remaining constant.

$$\left(\frac{\partial \rho}{\partial T}\right)_D = \frac{\partial}{\partial T}\int \sigma_x(u)\, w(u)\, du \tag{2}$$

The ratio of the doppler coefficient to the total sample reactivity worth is

$$\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_D = \frac{\dfrac{\partial}{\partial T}\int \sigma_x(u)\, w(u)\, du}{\int \sigma_x(u)\, w(u)\, du} = \frac{\partial}{\partial T}\left[\ln \int \sigma_x(u)\, w(u)\, du\right] \tag{3}$$

which will in most cases be a slowly varying function because of the logarithmic argument. As an example, suppose that in a hard spectrum reactor, all of the doppler effect arises from low-lying resonances (low kev range) so that only the low energy tail of the neutron spectrum is important. Over a lethargy width encompassing all the important resonances, but which is still small in comparison with the total, the flux will vary as the reciprocal of the total cross section (Ref. 3). Taking

$$w(u) = A\phi(u) = c/\sigma_t \tag{4}$$

then

$$\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_D = \frac{\dfrac{\partial}{\partial T}\displaystyle\int_{\Delta u} (\sigma_x/\sigma_t)\, du}{\int (\sigma_x/\sigma_t)\, du} = \frac{\partial}{\partial T}\left[\ln \int_{\Delta u} (\sigma_x/\sigma_t)\, du\right] \tag{5}$$

which is independent of the spectrum. Obviously, the above example is a gross oversimplification of the actual situation. In reality the weighting function is not simply separable, as indicated, because of contributions from nonresonant isotopes which also may be present. Furthermore, consideration of more than a single lethargy group will usually be necessary so that the gross spectrum shape, reflecting the effects of leakage, inelastic slowing down, etc., will be incorporated. Nonetheless, because of the logarithmic nature of the reactivity ratio, the consequences of these effects should be small.

There are at least two different pieces of experimental evidence to support this hypothesis. Perhaps the most convincing is the direct comparison of U-238 doppler coefficients as measured in two different fast critical assemblies, the AETR or ECEL assembly 13-13 (Refs. 12, 13) and the ZPR-6 assembly 4Z (Ref. 14). The pertinent characteristics of these two assemblies are tabulated in Table 1. We note

Table 1. Characteristics of the ECEL 13-13 and ZPR-6-4Z assemblies

Energy, Composition, and Dimensions	ECEL 13-13	ZPR-6-4Z
Median fission energy, kev	62	167
Median absorption energy, kev	40	73
Median flux energy, kev	227	212
Composition and Dimensions:		
1. Test Region:		
Boundaries, cm	0–30.6	0–41.0
Volume, cm^3	1.2×10^5	2.89×10^5
U-235 mass, kg; (atom density) · $cm^{-3} \cdot 10^{22}$	79.5, 0.17	172, 0.15
U-238	5.9, 0.012	1208, 1.06
Thorium	200, 0.43	—
Carbon	58.7, 2.45	71, 1.24
Aluminum	81.5, 1.51	—
Iron	144, 1.29	337, 1.26
Sodium	—	117.8, 1.07
2. Buffer, Decoupler:		
Boundaries, cm	30.6–35.1	41–47
Volume, cm^3	6.1×10^4	1.46×10^5
U-235 mass, kg; (atom density) · $cm^{-3} \cdot 10^{22}$	—	$2.6, 4.54 \times 10^{-3}$
U-238	—	1255, 2.17
Carbon	—	84, 2.89
Iron	—	81, 0.60
Thorium	625, 2.67	—
Aluminum	13.2, 0.48	—
3. Driver:		
Boundaries, cm	35.1–46.0	47.0–54.0
Volume, cm^3	2.27×10^5	2.25×10^5
U-235 mass, kg; (atom density) · $cm^{-3} \cdot 10^{22}$	16.3, 0.018	599, 0.683
U-238	1.2, 0.0013	475, 0.525
Hydrogen	18.8, 4.95	—
Carbon	112.1, 2.48	55, 1.24
Aluminum	152.9, 1.50	—
Iron	77, 0.37	262, 1.26
Sodium	—	91.7, 1.07
4. Reflector:		
Boundaries, cm	46.0–51.4	54.0–84.0
Volume, cm^3	1.59×10^5	1.82×10^6
U-235 mass, kg; (atom density) · $cm^{-3} \cdot 10^{22}$	—	57.5, 0.0081
U-238	—	28, 680, 3.98
Hydrogen	17.4, 6.53	—
Carbon	103, 3.26	—
Aluminum	43, 0.60	—
Iron	44, 0.30	1014, 0.60

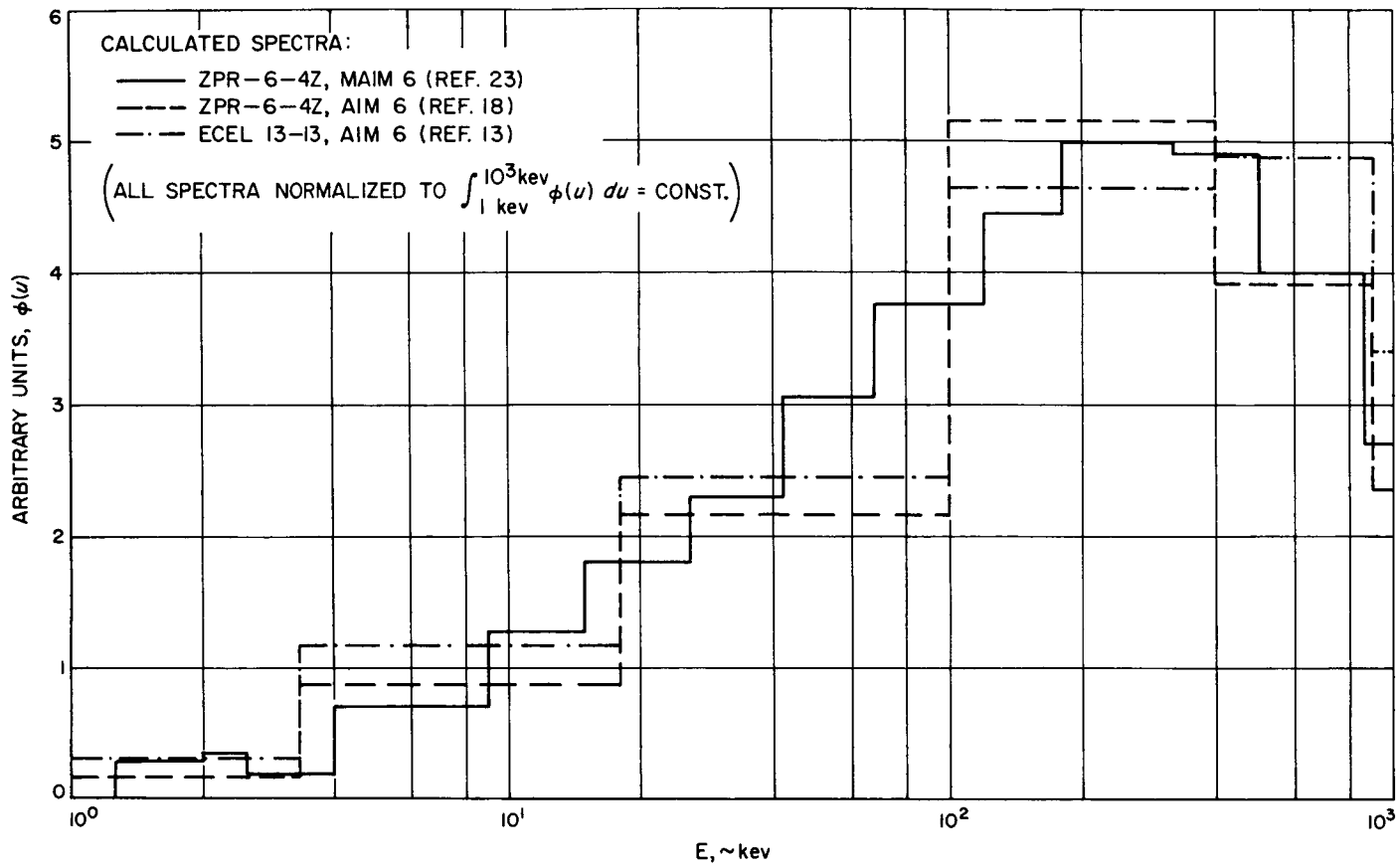

Fig. 1. Calculated spectra for ZPR-6-4Z and ECEL 13-13

that they are quite dissimilar in size, composition, and spectral character. The calculated spectra for the two assemblies are given in Fig. 1. Comparison of the two dashed curves shows the ECEL assembly to have a harder spectrum and a higher magnitude low energy tail. The solid curve is a more detailed energy group calculation of the ZPR-6 assembly and consequently gives a better representation of the actual flux distribution. Figure 2 presents a comparison between two different measurements of the flux spectrum of ZPR-6-4Z and the detailed energy group calculation. The overall agreement is seen to be excellent.

This particular comparison of reactivity ratios should provide a rather stringent test of the hypothesis since the significant U-238 resonances extend up to around 200 kev and consequently encompass a considerable portion of the neutron spectrum. The results of these measurements, which will be presented subsequently, indicate that the magnitudes of the reactivity ratios for U-238 agree to within 25%. Furthermore, there is no detectable difference in the temperature dependence between the two measurements.

A similar set of measurements has been made for U-235 in both assemblies. However, because of the presence of a thermal expansion reactivity coefficient which is the same order of magnitude as the doppler coefficient, the AETR data are not quite as reliable as the ZPR-6 data. Even so, the magnitudes of the reactivity ratios appear to agree to within 30%, based on the smaller but more reliable of the two values. A temperature dependence comparison comparable to that given for U-238 is not possible because of the thermal expansion effect in the AETR measurements. At present, these are the only two direct comparisons of doppler coefficients available.

The second piece of experimental evidence is the observation that the ratio of the reactivity worth of one material to another does not vary significantly between several different fast critical assemblies. Again, the concept of normalizing out the spectral dependence is employed by simply dividing the reactivity worth of one material by the reactivity worth of a second material.

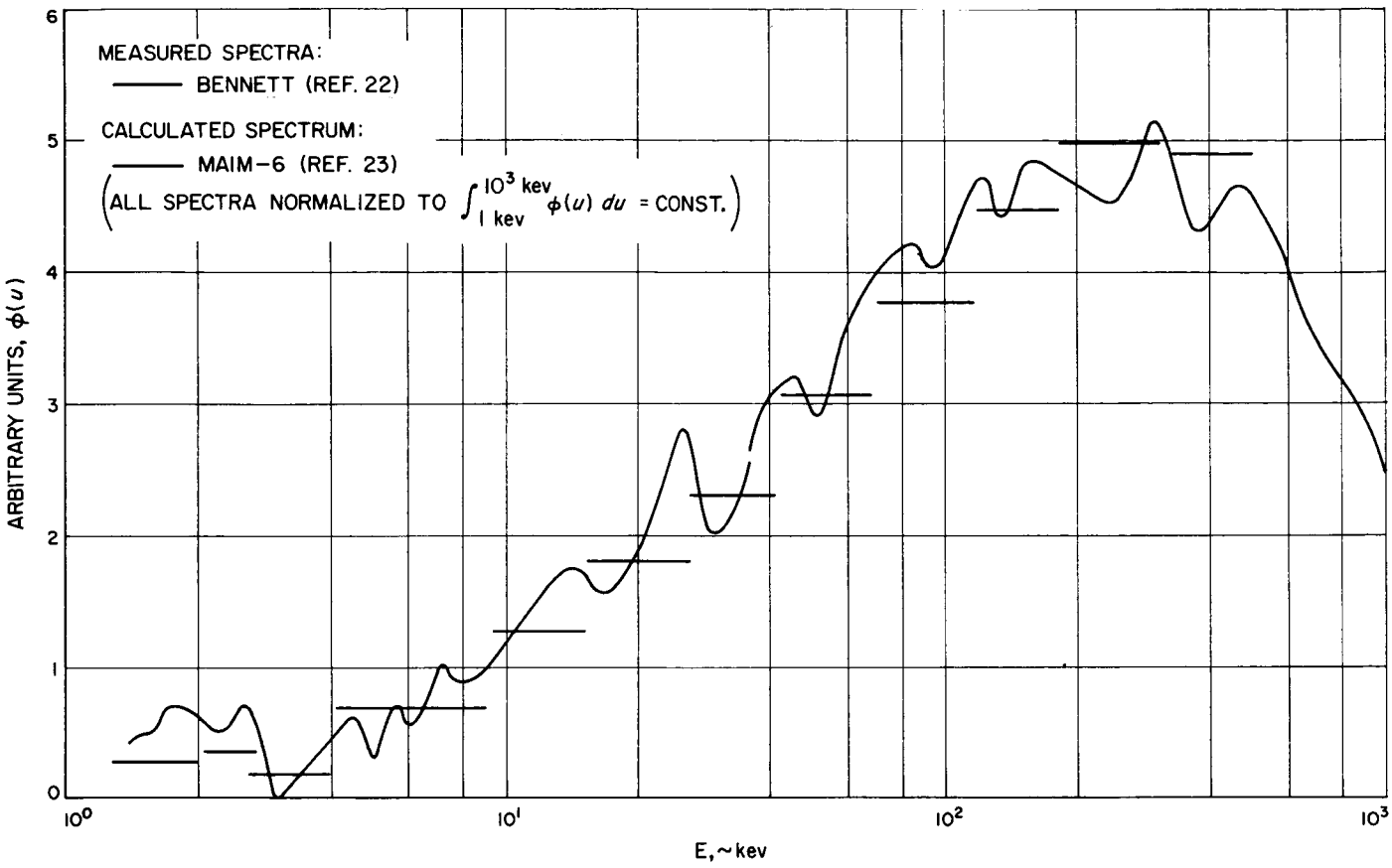

Fig. 2. Comparison of calculated and measured spectra in ZPR-6-4Z

For this comparison, the worths of U-238, W, Ta, Mo, and Nb were compared to U-235 in four hard spectrum assemblies. The measurements were made in the previously mentioned AETR (Ref. 9) and ZPR-6-4Z (Ref. 10) assemblies, and in the Swedish fast reactor FRO assemblies 1 and 3 (Ref. 15). These data are summarized in Tables 2 and 3, column 4, respectively. With but a single exception, the relative measured worths in the four assemblies are consistent between assemblies within less than 40%. The exception is the low U-238 worth in the FRO assemblies caused undoubtedly by resonance self-shielding due to their high U-238 content (20% enriched). It is entirely consistent to postulate that the relative worth ratios of the various materials in the FRO assemblies should be less than in the AETR or ZPR-6 assemblies because of the high U-238 resonance shielding effect. Without exception, this is the case. It is also consistent to postulate that the relative worth ratios in the ZPR-6 assembly should be smaller than the corresponding values measured in the AETR because of the larger sample size used and the U-238 content in the ZPR-6 measurements (see Table 2, column 1). Again, without exception, this is the case.

The experimental evidence may be summarized as follows: the two measured values of the U-238 doppler coefficients, when normalized by the room temperature reactivity worth of the sample, agree well in both magnitude and temperature dependence. The small difference in magnitude (25%) is thought to be primarily attributable to U-238 resonance self-shielding in the ZPR-6. The doppler coefficient comparison of U-235 is not as definitive as for U-238 because of a thermal expansion effect which tends to mask the doppler effect. Nevertheless, the magnitudes over a limited temperature range compare within 30%, based on the more reliable of the two measurements. Finally, ratios of reactivity worths in quite different hard-spectrum cores agree well among the four assemblies investigated. The differences, which for our purposes may be considered small, are all consistent with the qualitative argument of resonance shielding, either because of relatively larger sample sizes or (in the case of the FRO data)

Table 2. Experimental reactivity worths and doppler coefficients for the temperature range 20–600°C, AI and ANL data

Sample material	1 Sample mass, grams	2 Reactivity δ (¢) $\beta = 0.007$ $T = 20°C$	3 Reactivity sample mass $(\delta k/k)$ kg $(\times 10^4)$	4 Worth relative to U-235	5 Doppler reactivity $(\Delta\rho)_D$ (¢) 20°C–600°C	6 $(\Delta\rho)_D$ Sample mass $(\delta k/k)$ kg $(\times 10^6)$	7 Experimental error of value in column 6 $(\pm 1 \times 10^6)$	8 $\frac{(\Delta\rho)_D}{\rho}$
A I data (Ref. 9):								
U-235	183.81	2.091	7.97	1.0	0.00623	2.37	0.23	0.00297
U-235	59.11	0.645	7.63		0.00196	2.32	0.71	0.00304
33% U-235, U-238	176.98	0.543	2.15		−0.0072	−2.85	0.24	−0.0133
22% U-235, U-238	181.46	0.312	1.20		−0.01328	−5.12	0.23	−0.0427
10% U-235, U-238	180.08	0.0234	0.091		−0.0130	−5.05	0.23	−0.5549
U-238	179.17	−0.1827	−0.714	−0.09	−0.0152	−5.94	0.23	0.0832
U-238	117.87	−0.1198	−0.711		−0.00998	−5.93	0.38	0.0833
W	187.97	−0.286	−1.065 (−1.25)	−0.134 (−0.157)	−0.0109	−4.06		0.0381
Mo	100.10	−0.170	−1.189 (−1.25)	−0.149 (−0.157)	−0.0063	−4.41		0.0371
Ta	163.78	−0.795	−3.398 (−3.9)	−0.426 (−0.489)	−0.0513	−21.9		0.0645
Nb	84.94	−0.220	−1.814 (−1.9)	−0.228 (−0.238)	−0.0153	−12.61		0.0695
Argonne data (Refs. 10, 14, 16):								
U-235	13.869	0.186	9.41	1.0	0.000425	2.16		0.00230
U-238	1153.8	−1.176	−0.714	−0.076	−0.0776	−4.72		0.066
W	1108	−1.75	−1.107	−0.118				
Mo	599	−1.042	−1.219	−0.130				
Ta	924.7	−3.63	−2.754	−0.293				
Nb	481.2	−1.155	−1.684	−0.179				

Values in parenthesis represent infinite dilution values.

Table 3. Reactivity worths in the FRO reactor

Sample material	1 Sample mass, grams	2 Reactivity δ (¢) $\beta = 0.007$	3 Reactivity sample mass $(\delta k/k)$ kg $(\times 10^4)$	4 Worth relative to U-235
Assembly 1:				
U-235[a]	16.2	2.91	125.0	1.0
U-238	82.0	−0.316	−2.7	−0.022
W	191.0	−3.00	−11.0	−0.088
Mo	237.0	−4.06	−12.0	−0.096
Ta	215.0	−7.10	−23.1	−0.185
Nb	40.6	−1.05	−18.1	−0.148
Assembly 3:				
U-235[a]	16.2	1.90	88.2	1.0
U-238	82.0	−0.387	−3.3	−0.037
W	191.0	−2.37	−8.7	−0.099
Mo	237.0	−3.15	−9.3	−0.105
Ta	215.0	−6.14	−20.0	−0.226

[a]Data corrected from 20% U-235 sample.

because of the presence of large quantities of U-238. This normalization procedure has a rather obvious shortcoming which perhaps should nonetheless be emphasized. For U-235, the doppler effect changes from negative to positive as the median fission energy of the system is increased (see Fig. 3). This method cannot accommodate such a sign change since the reactivity worth of U-235 is always positive. Consequently, only assemblies characterized by median fission energies above approximately 50 kev or so, where the doppler effect is relatively constant, can be handled by this method.

B. Data Sources

Only two laboratories have reported doppler effect reactivity measurements pertinent to this study. These measurements have been performed on the ECEL (formerly the AETR) critical assembly 13-13 at Atomics International (AI), and on the ZPR-6 assembly 4Z at Argonne National Laboratory (ANL). Both General Electric and

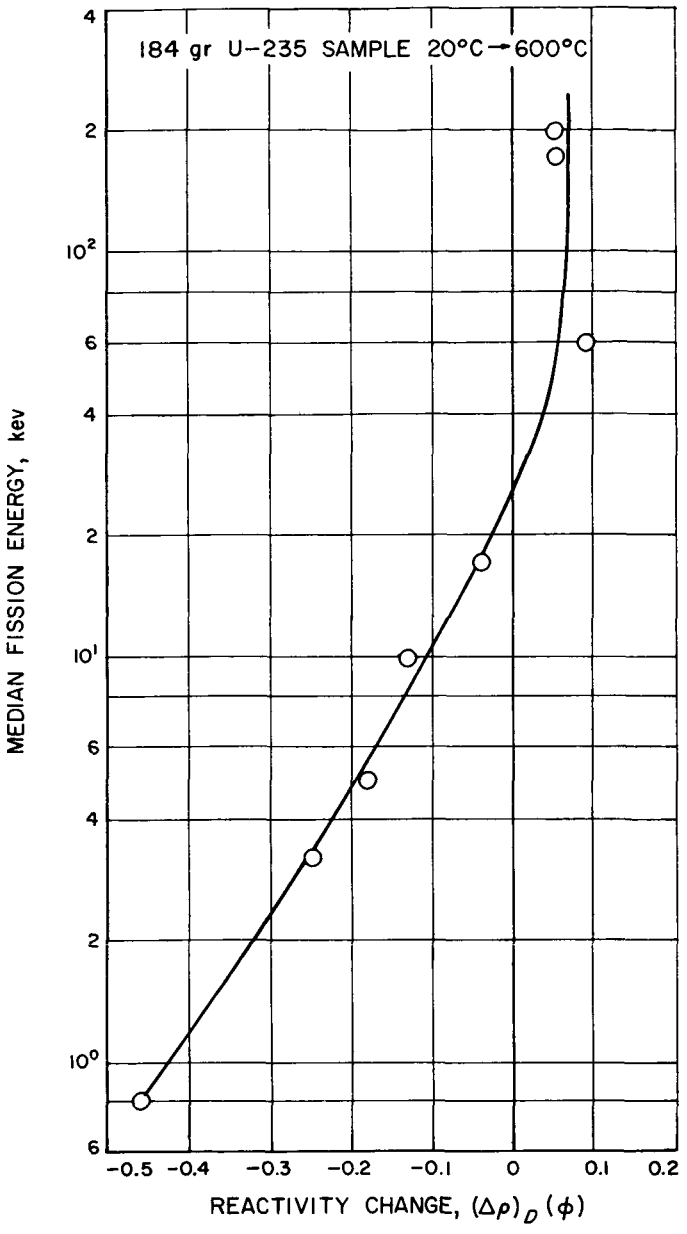

Fig. 3. Variation of doppler effect reactivity vs median fission energy for U-235

Argonne personnel are conducting doppler effect measurements by the foil activation method (Ref. 17), but this type of experiment is fundamentally different from the reactivity method and cannot be utilized here.

The correlations to be presented are based primarily on data reported in Report ANL-7120, Ref. 10, and private conversations with both AI and ANL investigators (Refs. 9, 16, 18).

C. Temperature and Isotopic Density Dependence

The temperature dependence of the doppler coefficient of reactivity is normally expressed in the form

$$\left(\frac{\partial\rho}{\partial T}\right)_D = AT^{-\gamma} \tag{6}$$

A theoretical value of $\gamma = 3/2$ is, as Nicholson points out (Ref. 2), only an approximation which appears to give an upper limit to the temperature dependence. Fortunately, experimental data for most of the materials of interest have now been obtained. The values of γ given in this report are all based upon experimental data with the exceptions of Mo and Nb for which the experimental data are inconclusive. In these latter cases, the theoretical value is assumed.

To obtain the magnitude A and the exponent γ experimentally, measurements are made of the reactivity change of a sample associated with a known change of temperature from some reference temperature T_0 to a temperature T. Integration of Eq. (6) gives for this reactivity change:

$$(\Delta\rho)_D = \frac{A}{1-\gamma}\left[T^{1-\gamma} - T_0^{1-\gamma}\right]; \qquad \gamma \neq 1 \tag{7a}$$

$$(\Delta\rho)_D = A \ln \frac{T}{T_0}; \qquad \gamma = 1 \tag{7b}$$

The reactivity data are analyzed by the method of least squares to an equation of the form (7a) or (7b), with values of γ ranging from 0.5 to 1.5. The value of γ leading to the minimum mean-square deviation is selected as the best value. Knowing γ, the constant A is easily evaluated.

For a mixture of the isotopes U-235 and U-238, the doppler coefficient theoretically depends on the square of the constituent densities, as opposed to a simple linear dependency (Refs. 2, 3). That is, if I is taken as the enrichment of the fuel (atomic fraction of U-235 to total uranium), then the reactivity change associated with the doppler effect in an isotopic mixture of U-235 and U-238 in proceeding from T_0 to T is:

$$(\Delta\rho)_D = I^2 (\Delta\rho)_D^{235} + (1-I)^2 (\Delta\rho)_D^{238} \tag{8}$$

where $(\Delta\rho)^i/D$ is the reactivity change associated with pure isotope i. A verification of this density dependence utilizing limited data from AI (Refs. 9, 18) was attempted here, but the results are inconclusive because of data

scatter. Additional data on isotopic mixtures, reported in Ref. 14, cannot be utilized at this time because these data have not been corrected for an important enrichment-dependent thermal-expansion effect. Actually, the density dependence, whether linear or squared, has only a small importance for our purposes, since normally the reactor concepts of interest use a high enrichment.

D. Results of Data Correlation

U-235

As has been mentioned, the measurement of the U-235 doppler coefficient is complicated by the presence of a thermal expansion surface-to-mass-ratio effect of the same magnitude but opposite in sign to the doppler effect. The overall reactivity change on an unrestrained U-235 sample is consequently about zero.[1]

The AI data on U-235 have been obtained on small unrestrained samples. An analytical correction of the data to account for the expansion effect results in data scatter too large to permit any interference of temperature dependence, but overall reactivity changes from 20°C to 600°C on two different sample sizes are reasonably consistent. In addition to the very hard spectrum cores, these measurements have been carried out in test sections in which the spectrum has been purposely softened. The results of these measurements are presented in Figure 3. The spectrum is characterized by the median fission energy. The total reactivity change, not the doppler coefficient, associated with the temperature change from 20°C to 600°C is plotted. This curve is particularly interesting since it indicates that the doppler effect approaches some maximum positive value and then either levels off or decreases slightly with increasing hardness of the spectrum. Theoretically, this is an anticipated behavior because of the resonance overlap at the higher energies (Refs. 2, 3).

The most recent Argonne data[2] (Ref. 16) have been obtained through a combination of experiments designed specifically to isolate the doppler effect. In essence, the cylindrical samples are physically restrained both axially and radially. Normalization of the expansion reactivity effect is obtained by observing the reactivity change associated with the phase change of uranium metal at about 660°C.

The Argonne results are shown in Fig. 4 where the total reactivity change from 20°C is plotted vs the logarithm of the temperature. The deviation of the data at the lower temperatures is thought to be due to an expansion correction which was neglected in this temperature range. The logarithmic dependence indicates a γ value of unity (as opposed to the more commonly assumed value of 3/2).

Using the Argonne data, the resulting reactivity ratio is:

$$\frac{1}{\rho^{235}}\left(\frac{\partial \rho^{235}}{\partial T}\right)_D = 2.1 \times 10^{-3}/T; \qquad {}^{\circ}K^{-1} \qquad (9)$$

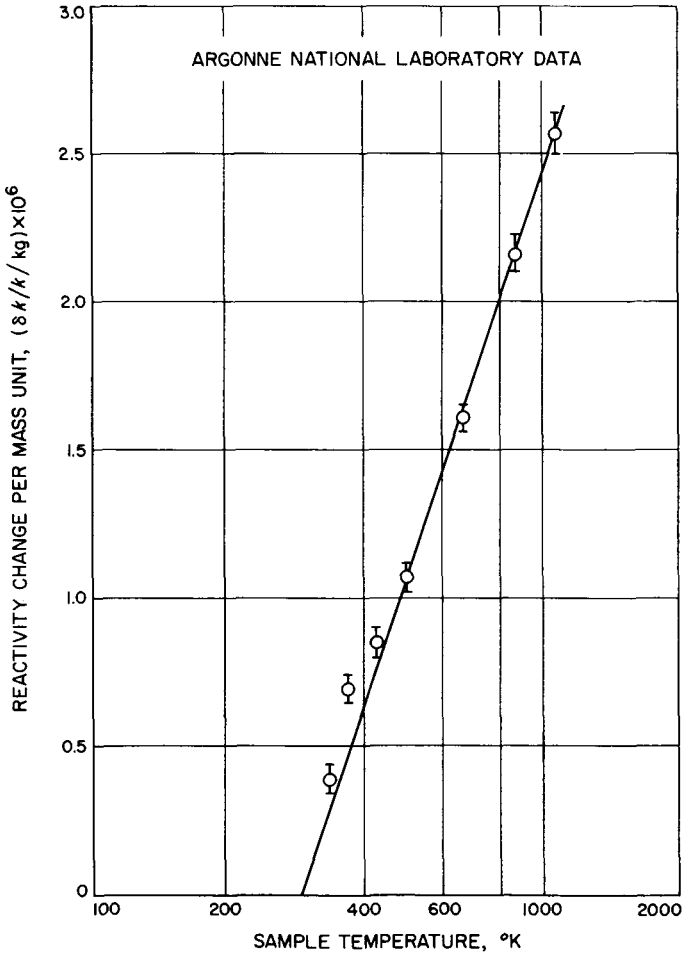

Fig. 4. Reactivity change due to the doppler effect for U-235 vs temperature

[1]This same effect will be present in thermionic space reactors to a degree which will depend upon the specific design. The expansion reactivity coefficient may be made small by physically restraining the expansion as, for example, in the case of a tungsten clad (emitter surface) fuel element. Since the expansion effect is negative, it will usually be conservative, for purposes of stability analysis, to ignore it until detailed studies for a specific concept are to be performed.

[2]Earlier values of doppler reactivities reported in Ref. 14 are in error due to the neglect of a radial expansion component. These samples were restrained axially only. The radial component was subsequently discovered to be significant.

U-238

For the non-fissile materials, the data interpretation is considerably simplified because of the absence of any strong thermal expansion reactivity component. A direct comparison of the Argonne and AI data is given in Fig. 5. Again, the total reactivity change is plotted, not the doppler coefficient. Here, the data are plotted vs $T^{0.2}$ indicating a γ-value of 0.8. This "best" value was obtained through a least-squares data analysis performed by both AI and Argonne investigators as mentioned previously. The difference in the slope of the two straight lines reflects the 25% difference in the magnitude of the reactivity ratio and is thought to be due to resonance self-shielding introduced by the larger sample size used in the Argonne measurements. In any event, an error of this order in the magnitude is of no consequence for purposes of this report.

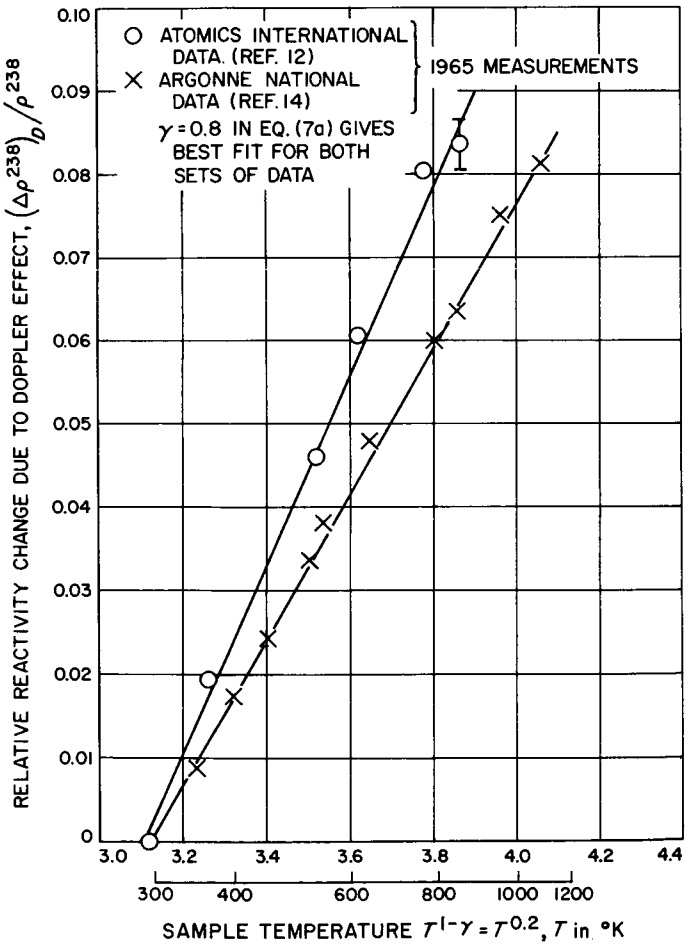

Fig. 5. Reactivity change due to the doppler effect for U-238 vs temperature; $\gamma = 0.8$

Using the AI data, the resulting reactivity ratio is:

$$\left(\frac{1}{\rho^{238}}\frac{\partial\rho^{238}}{\partial T}\right)_D = 2.2\times 10^{-2}/T^{0.8}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (10)$$

U-235 and U-238 isotopic mixtures

The results of an attempt to verify the squared dependence of the isotopic density are shown in Fig. 6. The data scatter is obviously too large to draw any conclusions. Consequently for mixtures, the theoretical density squared dependency is recommended.

$$\left(\frac{\partial\rho^{\mathrm{mix}}}{\partial T}\right)_D = I^2\left(\frac{\partial\rho^{235}}{\partial T}\right)_D + (1-I)^2\left(\frac{\partial\rho^{238}}{\partial T}\right)_D \qquad (11)$$

W, Mo, Ta, Nb

The only data available on these refractory metals are those from AI (Refs. 9, 18). The γ values are at best very approximate, having been obtained only over the temperature range 20 to 600°C. In the cases of Mo and Nb, the data were inconclusive but appeared to be tending toward the theoretically limiting value of 3/2. The correlation results for these elements are:

W:

$$\frac{1}{\rho^{\mathrm{W}}}\left(\frac{\partial\rho^{\mathrm{W}}}{\partial T}\right)_D = 8.5\times 10^{-3}/T^{0.8}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (12)$$

Mo:

$$\frac{1}{\rho^{\mathrm{Mo}}}\left(\frac{\partial\rho^{\mathrm{Mo}}}{\partial T}\right)_D = 0.72/T^{1.5}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (13)$$

Ta:

$$\frac{1}{\rho^{\mathrm{Ta}}}\left(\frac{\partial\rho^{\mathrm{Ta}}}{\partial T}\right)_D = 1.5\times 10^{-2}/T^{0.8}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (14)$$

Nb:

$$\frac{1}{\rho^{\mathrm{Nb}}}\left(\frac{\partial\rho^{\mathrm{Nb}}}{\partial T}\right)_D = 1.36/T^{3/2}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (15)$$

III. Applications

A. Thermionic Reactor Applications

To apply Eqs. (9–15) the average reactivity worth, attributable to the resonance absorption in each given material in the reactor concept of interest, is required.

These values can be calculated or inferred from measurements. In either case the change in leakage reactivity effect, attributable to the scattering cross section of the resonance material, must be subtracted. By calculation, this can be accomplished by deleting the scattering cross section of the resonance absorber and then performing a perturbation calculation relative to the basic reactor (for U-235, only the scattering cross section of the perturbing increments are deleted). Experimentally, in the test assemblies having approximately flat flux and importance distributions through the test regions (such as the assemblies used in this correlation), this scattering correction is small and may be neglected.

Alternatively, for estimation purposes, the following scheme may be employed. A reasonable rule-of-thumb estimate for U-235 worth in a fast assembly is 0.5% $\delta k/k$ per % $\delta M/M$, where M is the critical mass of U-235; i.e., a 1% change of U-235 mass in a fast reactor leads approximately to a 0.5% reactivity change. Some examples of this approximation are included in the Douglas concept (Ref. 11) where the calculations indicate a value of 0.54%/%. In the two FRO assemblies, which are only 20% enriched, the measured central reactivity worth is 0.91%/% for assembly 1 and 0.97%/% for assembly 3. Using a peak to average importance weighting of 1.8, the average worths are about 0.5%/% (Ref. 15). For the Fermi reactor mockup conducted in ZPR-3, this ratio is 0.474 (Ref. 19). As with all rules of thumb, there are exceptions so that reasonable care should be exercised in using this value. Note that this constant value is completely consistent with the doppler effect (above ~60 kev median fission energy) shown in Fig. 3.

Knowing the U-235 reactivity worth, the remaining materials can all simply be scaled by experimental worth ratios, such as those given in column 4 of Table 2. It is recommended that the infinite dilution values indicated in parenthesis be used. Obviously, if significant self-shielding is involved, this method will result in too large an estimation of the doppler effect for any given material. The doppler coefficient is simply:

$$\left(\frac{\partial \rho^i}{\partial T}\right)_D = A^i T^{-\gamma i} \times \left(\frac{M^i}{M^{235}}\right) \times \left(\frac{\% \delta k/k}{\% \delta M/M}\right)^{235} \times \left(\frac{(\rho/\mathrm{gm})^i}{(\rho/\mathrm{gm})^{235}}\right)$$

where A^i and γ^i are the appropriate coefficients from Eqs. (9–15).

Using the 0.5%/% value for U-235 and the infinite dilution worth ratios from the AI data results in the following expressions.

U-235:

$$\left(\frac{\partial \rho^{235}}{\partial T}\right)_D = 1 \times 10^{-3} I^2/T; \qquad {}^\circ\mathrm{K}^{-1} \qquad (16)$$

U-238:

$$\left(\frac{\partial \rho^{238}}{\partial T}\right)_D = -1 \times 10^{-3} (1 - I)^2/T^{0.8}; \qquad {}^\circ\mathrm{K}^{-1} \qquad (17)$$

W:

$$\left(\frac{\partial \rho^{W}}{\partial T}\right)_D = -\frac{6.9 \times 10^{-4}}{T^{0.8}} \left(\frac{M^W}{M^{235}}\right); \qquad {}^\circ\mathrm{K}^{-1} \qquad (18)$$

Mo:

$$\left(\frac{\partial \rho^{Mo}}{\partial T}\right)_D = -\frac{5.8 \times 10^{-2}}{T^{1.5}} \left(\frac{M^{Mo}}{M^{235}}\right); \qquad {}^\circ\mathrm{K}^{-1} \qquad (19)$$

Ta:

$$\left(\frac{\partial \rho^{Ta}}{\partial T}\right)_D = -\frac{3.7 \times 10^{-3}}{T^{0.8}} \left(\frac{M^{Ta}}{M^{235}}\right); \qquad {}^\circ\mathrm{K}^{-1} \qquad (20)$$

Nb:

$$\left(\frac{\partial \rho^{Nb}}{\partial T}\right)_D = -\frac{0.15}{T^{1.5}} \left(\frac{M^{Nb}}{M^{235}}\right); \qquad {}^\circ\mathrm{K}^{-1} \qquad (21)$$

All that remains to be specified is the enrichment I and the ratio of the mass of material i to the critical mass (loading) of U-235, (M^i/M^{235}).

B. Isotopic Concentration of U-235 for Zero Doppler Coefficient

For stability purposes, it may be desirable to ensure a zero or negative prompt fuel coefficient. Neglecting the expansion effect, this can be accomplished by reducing the enrichment to the point where the U-238 doppler coefficient cancels the positive U-235 coefficient.

For isotopic mixtures of uranium, the total doppler coefficient is given by Eq. (11). Using the values of the

previous section, Eq. (11) becomes

$$\left(\frac{\partial \rho^{mix}}{\partial T}\right)_D = \frac{10^{-3} I^2}{T} - \frac{10^{-3}(1-I)^2}{T^{0.2}}$$

Denoting I_0 as the enrichment for which

$$\frac{\partial \rho^{mix}}{\partial T_D}$$

is zero, the above equation results in

$$I_0 = \frac{1}{1 + T^{-0.1}} \qquad (22)$$

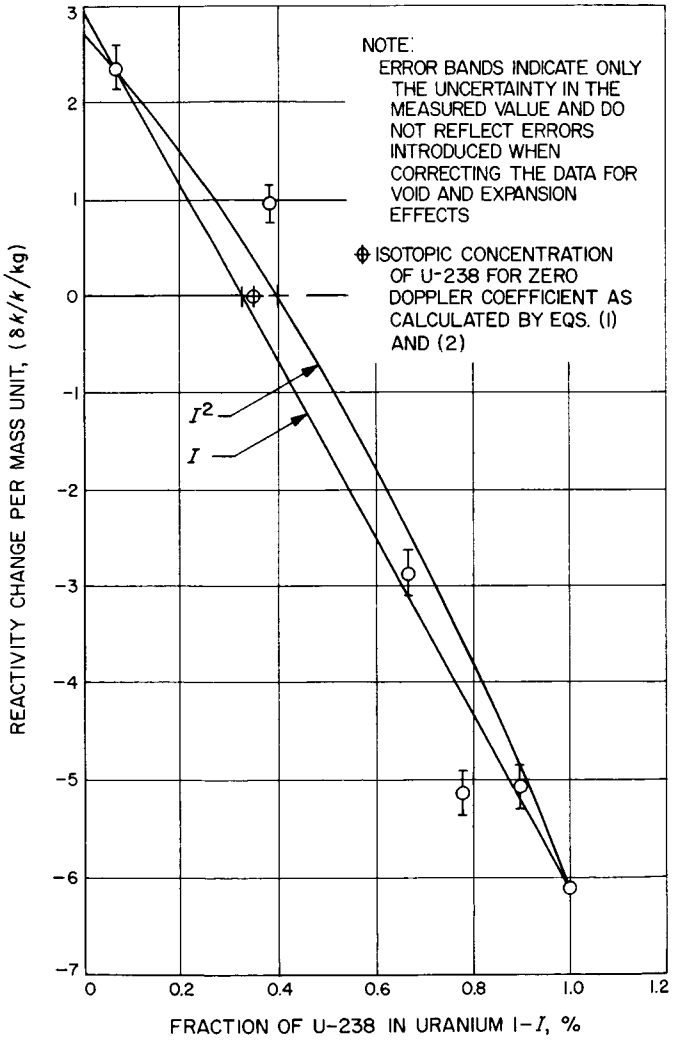

Fig. 6. Density dependence of doppler effect for isotopic mixtures of uranium

The temperature dependence of Eq. (22) is negligible. I_0 evaluated at 500°K and 2000°K changes from 65% to 68%. Because of the negligible temperature dependence, this value can also be read directly from Fig. 6.

If power flattening were accomplished by varying the enrichment radially and axially in a thermionic reactor, the above average enrichment would be increased somewhat due to importance-weighting considerations. In any event, this approximate value for an overall enrichment is not outside the realm of interest. In the General Electric concept for example (Ref. 20), power flattening is achieved by varying the U-235 content from 44% in the center of the reactor to fully enriched at the extremities. However, this concept substitutes void for fuel instead of U-238, as proposed here.

C. Estimation of Uncertainty Associated with Extrapolation

The Argonne U-235 data for the reactivity change associated with a temperature change were presented previously in Fig. 4 plotted as a function of log T. The resulting straight line indicated a doppler coefficient temperature dependence of T^{-1}. In Fig. 7, the same data have been re-plotted vs $T^{1-\gamma}$ for $\gamma = 0.8$ and $\gamma = 1.2$. A value of $\gamma = 1.2$ appears to be overestimating the temperature dependence while the value of $\gamma = 0.8$ actually still gives a reasonably good fit of the data. The consequent effect on the magnitude of the doppler coefficient as a function of temperature is shown in Fig. 8 together with similar estimates for U-238 and W. In the case of U-235, the error introduced is seen to be relatively small. For example, at 2000°K, the deviation in doppler coefficient from its $\gamma = 1$ value is only of the order of 20 to 30%. Even at 3000°K, the deviation is only 30 to 40%.

For U-238 the same type of error estimation can be made. As mentioned previously, the value of $\gamma = 0.8$ was determined from a minimized deviation least-squares fit of the data. Changes in deviation about the minimum value leading to the $\gamma = 0.8$ value are small, however. That is to say, a value of $\gamma = 1.0$, for example, will also fit the data well with only a small increase in the mean-square deviation. In Figs. 9 and 10, the U-238 data are re-plotted vs ln T ($\gamma = 1$) and $T^{0.3}$ ($\gamma = 0.7$), respectively. This range of values of γ still results in reasonably good fits to the data. At still larger or smaller values of γ, the fit becomes obviously poorer. The resultant temperature dependence of the doppler coefficient is shown in Fig. 8. At 2000°K, the magnitude of uncertainty about the $\gamma = 0.8$ value is seen to be only ±20%, similar to the U-235.

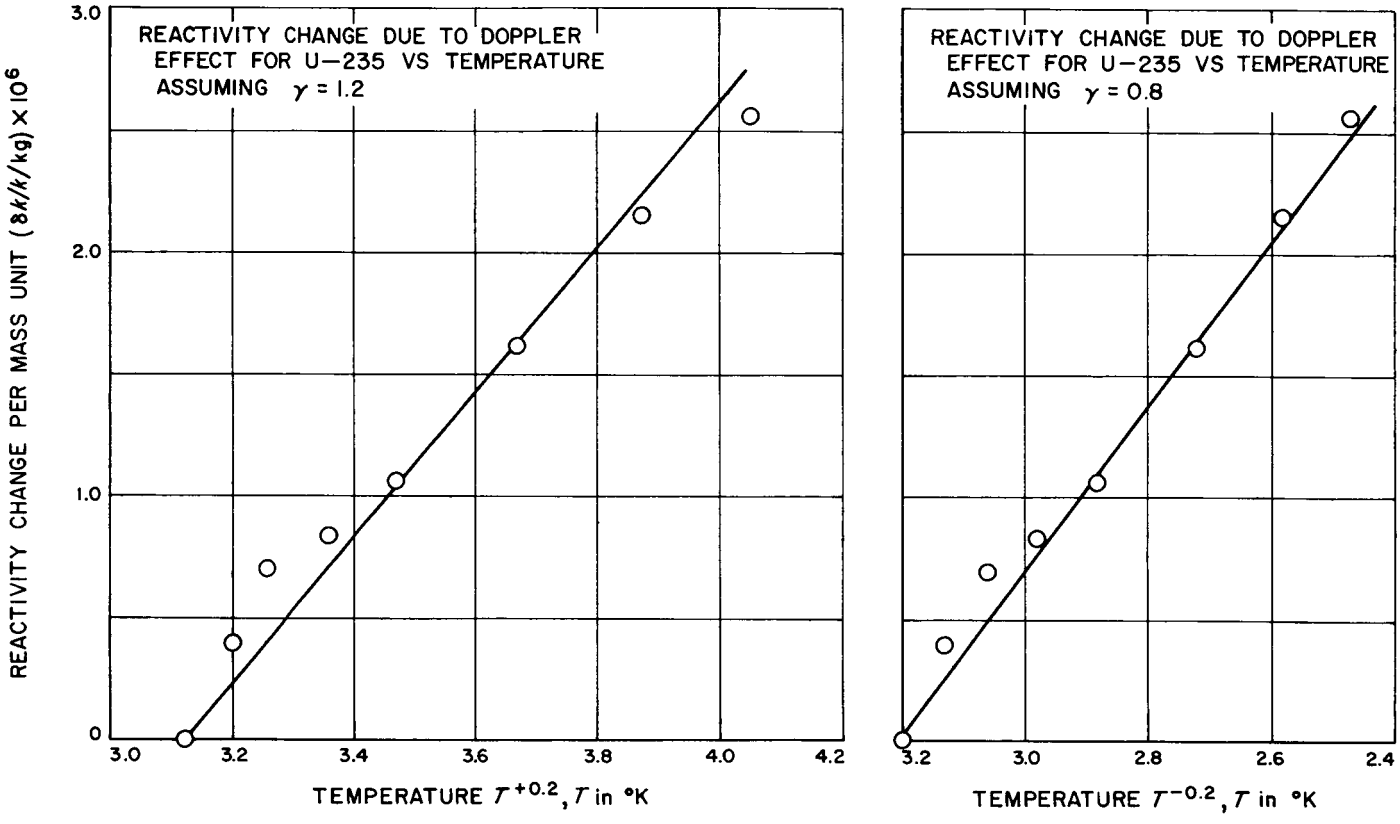

Fig. 7. Comparison of fit of U-235 doppler effect data to $T^{1-\gamma}$ for $\gamma = 0.8$ and $\gamma = 1.2$

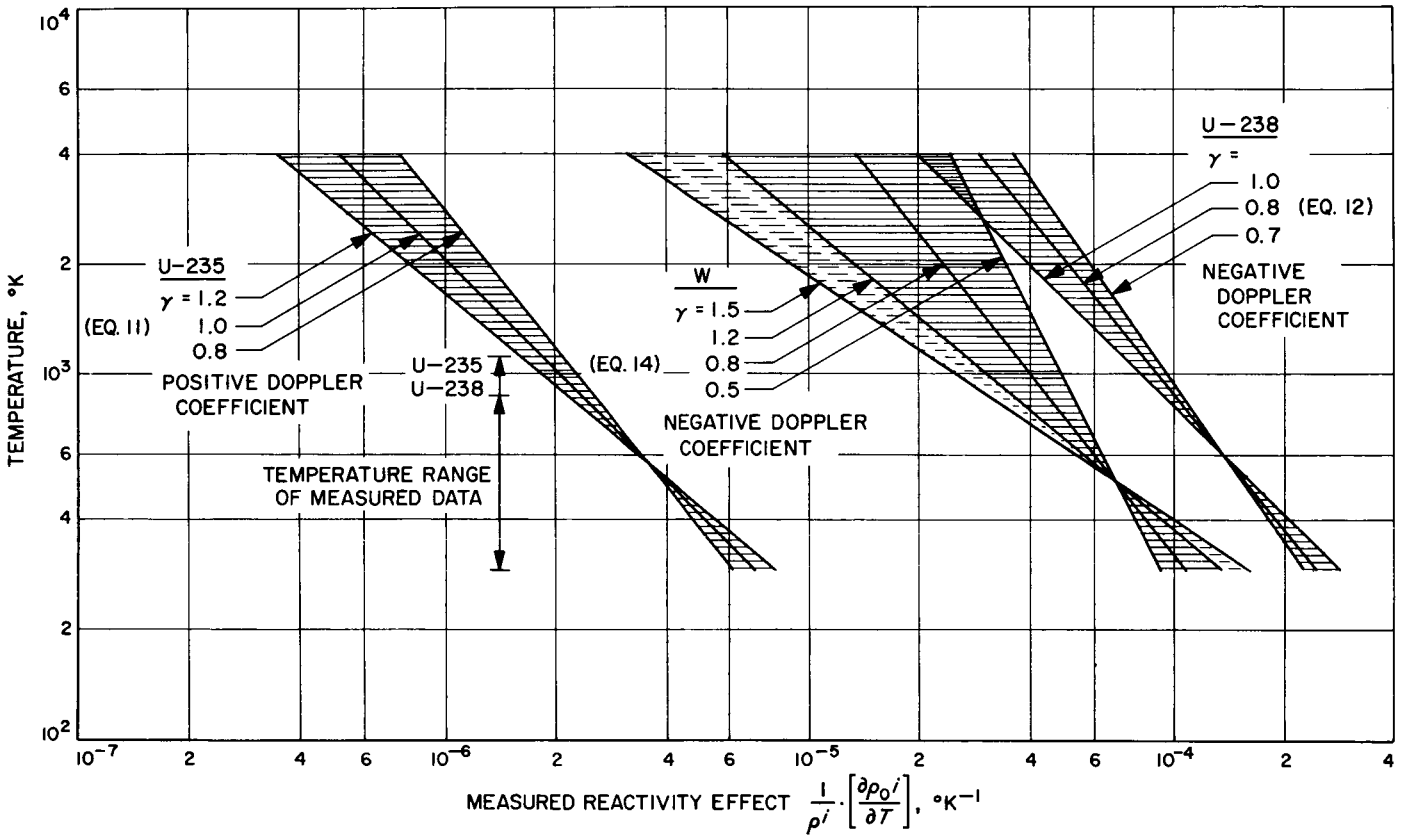

Fig. 8. Estimates of errors for the doppler coefficients of U-235, U-238, and tungsten

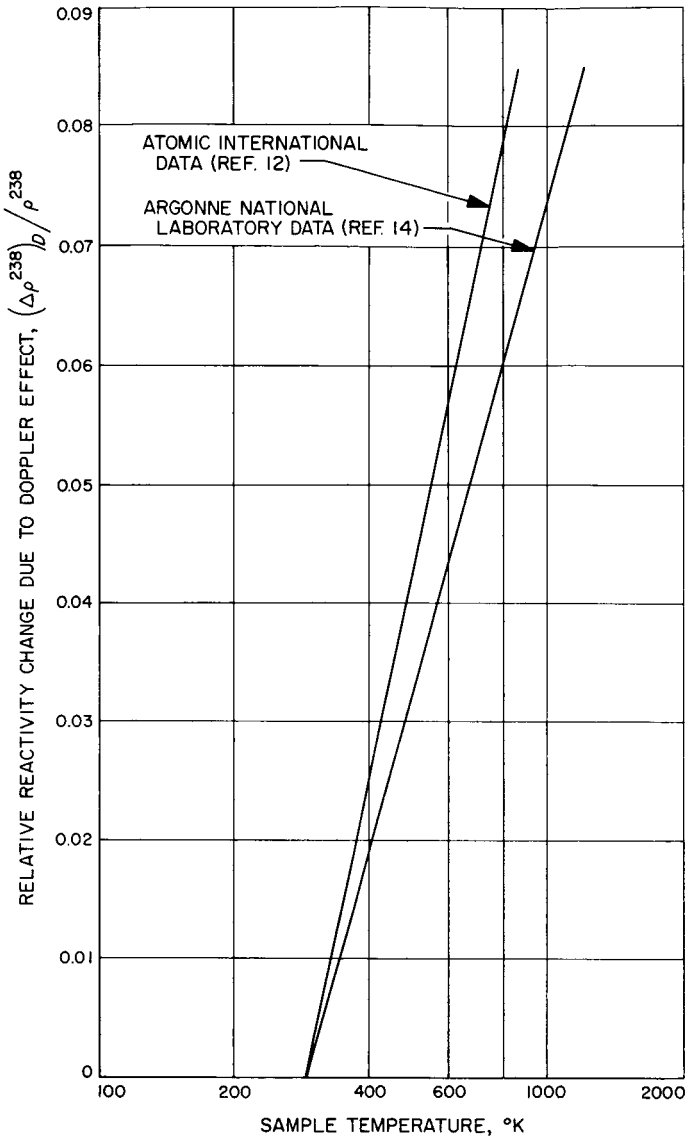

Fig. 9. Reactivity change due to the doppler effect for U-238 vs temperature; $\gamma = 1.0$

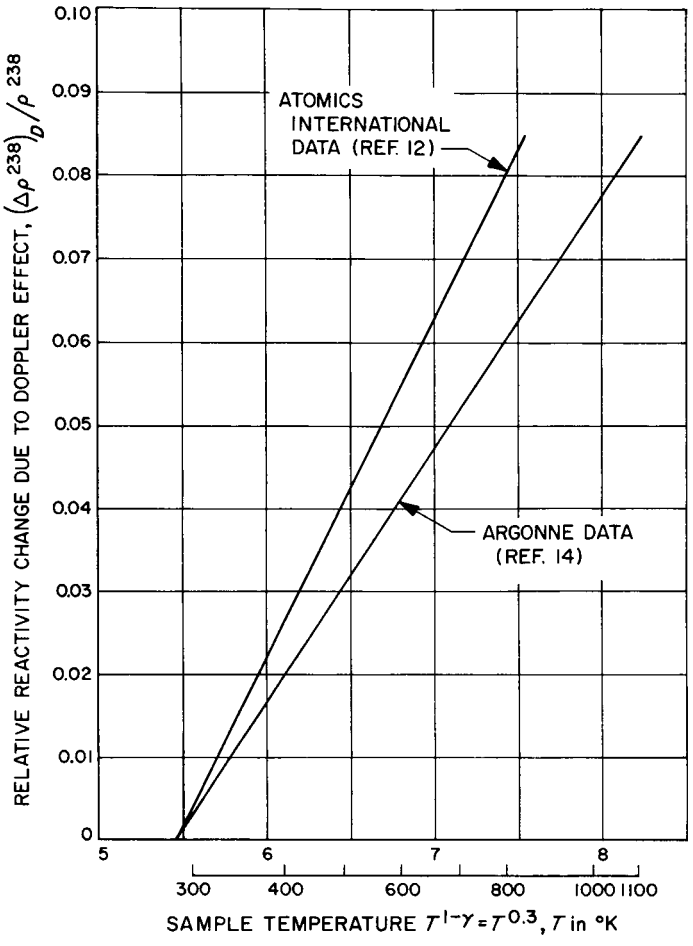

Fig. 10. Reactivity change due to the doppler effect for U-238 vs temperature; $\gamma = 0.7$

Following the same procedure as for uranium, estimates of the error for W, Mo, Ta, and Nb can also be made. In these cases, however, a larger uncertainty in the value of γ exists, due to the fact that fewer data have been taken and the data extend over a narrower temperature range (20–600°C). Figure 8 shows an estimation for tungsten. The limits of $\gamma = 0.5$ and $\gamma = 1.2$ about the "best" value of $\gamma = 0.8$ result in an uncertainty of the doppler coefficient at 2000°K of ±50%. The behavior for tantalum will be identical to that of tungsten since the "best" values of γ are the same.

Similarly the uncertainty for molybdenum and niobium will be obtained by considering variations about the $\gamma = 1.5$ value, plotted for convenience also in Fig. 8. The value of $\gamma = 1.5$ is probably an upper limit to the temperature dependence. The paucity of data prevents a better estimate of γ at this time and, consequently, a relatively large uncertainty band about the $\gamma = 1.5$ value should be assumed.

D. Comparison with Nicholson's Calculation for the Fermi Reactor

In Ref. 2, Nicholson calculates the following values for the Fermi reactor at 560°K:

$$I^2\left(\frac{\partial\rho^{235}}{\partial T}\right)_D = 0.25\times 10^{-6}; \qquad °K^{-1}$$

$$(1-I)^2\left(\frac{\partial\rho^{238}}{\partial T}\right)_D = -2\times 10^{-6}; \qquad °K^{-1}$$

From Ref. 2, $I = 0.27$, and from Ref. 19, $\rho^{235} = 0.474\%/\%$, $\rho^{238} = 0.021\%/\%$. The reactivity worth values are from the Fermi reactor mockup on ZPR-III (Assembly 20). Reference 19 reports an average U-235 worth of 51.6 Ih/kg $\cong$ 0.11¢/kgm for a critical mass loading of 431 kg, and also gives central reactivity worths for U-235 and U-238. The ρ^{238} value is obtained by a simple ratio of central worths times the average U-235 worth.

At 560°K, Eqs. (9) and (10) are:

$$I^2\left(\frac{\partial\rho^{235}}{\partial T}\right)_D = (0.27)^2 \times 2.1 \times 10^{-3} \times 0.474/560$$
$$= 0.13 \times 10^{-6}; \qquad °K^{-1}$$

$$(1-I)^2\left(\frac{\partial\rho^{238}}{\partial T}\right)_D = -(0.73)^2 \times 0.022 \times 0.0207/(560)^{0.8}$$
$$= -1.15 \times 10^{-6}; \qquad °K^{-1}$$

Thus, the experimentally derived values are roughly a factor of two smaller than those calculated by Nicholson.

IV. Conclusions and Recommendations

A method of estimating doppler coefficients for fast reactors characterized by a high median fission energy has been presented. Experimental data may be used directly to give good estimates of both the doppler coefficient and its temperature dependence. Experimentally evaluated correlations for the doppler coefficient of the uranium isotopes 235 and 238 and for the refractory metals, tungsten, tantalum, molybdenum, and niobium, are given by Eqs. (9–15). The application to a typical thermionic reactor concept, using additional experimental data and a rule-of-thumb reactivity worth for U-235 in this type of reactor, results in Eqs. (16–21). A zero doppler coefficient for 65% enriched fuel is estimated. The uncertainty associated with extrapolating present experimental data to higher temperatures is acceptably small for U-235 and U-238, but may be considerably larger for the refractory metals. Finally, a comparison of this method with the theoretical calculations of Nicholson on the Fermi reactor indicates that the latter calculations may be high by about a factor of two. Considering the complexity of the problem, this is not an unreasonable difference.

Further experimental work is certainly warranted (Refs. 21, 22, 23). It would be highly desirable to have a third independent measurement of the U-238 doppler coefficient, since this isotope provides a stringent test of the spectral normalization assumption. Assuming the validity of this method, however, the present doppler coefficient measurements on both U-238 and U-235 are sufficiently accurate and extend over a sufficiently large temperature range so that further experimentation, for our purposes, is unnecessary.

The refractory metal experiments are not as satisfactory. A second set of measurements by the Argonne investigators and a repetition of the AI experiments over a larger temperature range (to at least ~1100°K) are required. These measurements would serve to refine and verify the present data and would provide a sufficient temperature range basis for accurate extrapolation to the higher temperatures of interest for thermionic reactor concepts.

Nomenclature

A	proportionality constant
c	proportionality constant
D	doppler, subscript
I	U-235 enrichment in uranium
i	identification index for material i, superscript
M	mass
o	room temperature or zero net doppler effect reactivity, subscript
T	temperature
u	lethargy variable
$w(u)$	weighting function
γ	exponent for temperature dependence
ρ	reactivity
σ_x	microscopic interaction cross section
ϕ	neutron flux

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