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**EFFECTS OF UO_2 CRYSTALLINE BINDING
ON DOPPLER COEFFICIENTS CALCULATIONS
FOR FAST REACTOR SYSTEMS**

R. E. SCHENTER

JUNE 1967

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FOR FAST REACTOR SYSTEMS

By

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June 1967

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ABSTRACT

The change in the Doppler coefficient due to crystalline binding effects has been studied for ^{238}U in a UO_2 lattice for fast reactor systems. Use has been made of the "weak binding," or the short compound nucleus lifetime (SCNL) approximation, where the ^{238}U atoms in the UO_2 lattice are treated as a free gas with an effective temperature. Calculations were made to determine the dependence of effective temperature versus temperature using the experimental phonon distribution of Dolling, et al. Comparisons were also made with effective temperatures obtained assuming a Debye model for the crystal. It was found that for temperatures above 300 °K the effective temperature dependence was well represented by the Debye model ($\theta_D \sim 620$ °K). The temperature dependence of the reactivity was calculated for the ANL critical assembly, ZPR-III, 47 with the result that incorporation of the crystalline binding effects caused the calculated Doppler coefficient ($\frac{dk}{dT}$) to diverge from the " $\frac{1}{T}$ " rule in the low temperature region, in qualitative agreement with the experimental results of Reynolds and Stewart.

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PART I. INTRODUCTION

Crystalline binding effects in uranium oxide have an important effect on the low temperature Doppler coefficient in fast spectrum oxide reactors.⁽¹⁾ The Doppler effect, coming primarily from the temperature broadening of resonances in the ^{238}U capture cross section, is reduced by crystalline binding effects. This reduction occurs because the ^{238}U atoms in the UO_2 crystal vibrate with the average kinetic energy larger than that which would occur in either the metallic or the free gas state. The effective temperature of the ^{238}U atoms is thus higher than the temperature of the medium, with the result that the broadening of the resonances with increased temperature is smaller than that given by a noncrystalline state.

This paper deals with this reduction in the Doppler coefficient, where the "weak binding," or the short compound nucleus lifetime (SCNL) approximation⁽²⁾ has been assumed. In this approximation the ^{238}U atoms in the UO_2 lattice, for the process of capturing fast neutrons, behave like a free gas with an effective temperature.[†] Calculations of the effective

temperature versus temperature have been performed by using the experimental phonon distribution of Dolling et al.,⁽⁴⁾ and comparisons were made with results obtained assuming a Debye model for the crystal. Part II gives the graphical results and the equations used for the above analyses. In Part III an expression for the "bound" Doppler coefficient is derived in terms of the "free-gas" Doppler coefficient^{††} assuming the validity of the effective temperature model.

This report also presents the calculation of the reactivity change versus temperature for the ZPR-III, Assembly 47, performed by means of using the effective temperature model.

PART II. EFFECTIVE TEMPERATURE MODEL

The effective temperature T_{eff} in the SCNL approximation is given in terms of $\bar{E}$ the average kinetic energy per mode of oscillation as⁽³⁾

$$KT_{\text{eff}} = \bar{E} = \int_0^\infty d\nu \, g(\nu) \left(\frac{h\nu}{2} \right) \coth \left(\frac{h\nu}{2KT} \right), \quad (1)$$

[†] This should be a good approximation for neutrons with energies that are important for Doppler coefficient calculations in fast reactors (~ 2 Kev). See Reference 3 for a discussion of this point.

^{††} Temperature dependent cross sections used as input in Doppler coefficient calculations are usually obtained assuming the targets to be in a free gas state.

where ν is the phonon frequency and $g(\nu)$ is the phonon frequency distribution, number per unit frequency interval. Figure 1 gives the results of using Equation (1) for the Dolling⁽⁴⁾ phonon distribution (Figure 2) and for the Debye distribution, where $g(\nu)$ is given as⁽⁵⁾

$$g_D(\nu) = 3 \frac{\nu^2}{v_D^3} \text{ for } \nu \leq v_D \quad (2a)$$

$$= 0 \text{ for } \nu > v_D, \quad (2b)$$

and the Debye temperature θ_D is defined as

$$k\theta_D = h\nu_D. \quad (3)$$

The description of a crystalline substance is frequently given in terms of its Debye temperature so that crystalline effects in a fast reactor, for example, are sometimes characterized by a single number θ_D . This description is approximate, because in general the crystal does not have a Debye phonon distribution. One can, however, define an effective Debye temperature which varies with the medium temperature and gives the correct answer for a given physical property. Figure 3, shows the results of this type of prescription, where the effective Debye temperatures⁽⁵⁾ $\theta_D(T)$ are given by the following equations:

$$\bar{E}(T) = \int_0^{k\theta_{D,\bar{E}}(T)} g_D[\theta_{D,\bar{E}}(T), \nu] \left(\frac{h\nu}{2kT}\right) \coth\left(\frac{h\nu}{2kT}\right) d\nu, \quad (4)$$

$$C_V(T) = k \int_0^{k\theta_{D,C_V}(T)} g_D[\theta_{D,C_V}(T), \nu] \left(\frac{h\nu}{2kT}\right)^2 \text{csch}^2\left(\frac{h\nu}{2kT}\right) d\nu, \quad (5)$$

where $\bar{E}(T)$ and $C_V(T)^*$ are calculated from the "true" phonon distribution. The results show (Figures 1 and 3), that to a good approximation, for temperatures above 300 °K, ²³⁸U in a UO₂ lattice is described by a Debye model with a Debye temperature of 640 °K.

PART III. DOPPLER COEFFICIENT DEPENDENCE

When the SCNL approximation is assumed, the bound capture cross section of the ²³⁸U atoms varies with temperature as

$$\sigma_{CB}(T) = \sigma_{CFG}(T_{\text{eff}}), \quad (6)$$

where $\sigma_{CFG}(T_{\text{eff}})$ is the capture cross section which gives the correct reaction rate for a free gas system at a temperature T_{eff} . The temperature variation in the criticality factor $k_B(T)$ for a crystalline system then becomes

$$\begin{aligned} k_B(T) &= k[\sigma_{CB}(T)] \\ &= k_{FG}(T_{\text{eff}}), \end{aligned} \quad (7)$$

* The specific heat curve $C_V(T)$ is given to indicate how the effective Debye temperature varies with a physical property. Also, the specific heat dependence is the experimental quantity which is usually used to determine the Debye temperature of a given substance (see Reference 3).

where $k_{FG}(T_{eff})$ is the criticality factor for a free gas system at the temperature T_{eff} . Differentiating Equation (7) relates the bound and free gas Doppler coefficients

$$\frac{dk_B}{dT} = \frac{dk_{FG}}{dT_{eff}} \frac{dT_{eff}}{dT} . \quad (8)$$

In order to explicitly demonstrate the crystalline binding effects on the Doppler coefficient, a $1/T$ dependence is assumed for the free gas Doppler coefficient.[†] Equation (8) then becomes

$$\frac{dk_B}{dT} = \left\{ \frac{T}{T_{eff}} \frac{dT_{eff}}{dT} \right\} \frac{dk_{FG}}{dT} . \quad (9)$$

The quantity in the brackets in Equation (9) then represents the deviation of the Doppler coefficient from the free gas case. As can be seen in Figure 1, both factors T/T_{eff} and

$\frac{dT_{eff}}{dT}$ tend to reduce the value of

the bound Doppler coefficient.

Figure 4 shows the temperature dependence of this reduction, where the ratio of the bound to free gas coefficient is plotted by using the effective temperature results given in Part II. Figure 5 shows the temperature dependence of the reactivity with and without binding effects^{††} obtained from a calculation on the ZPR-III, Assembly 47

[†] This is a good approximation for most fast reactor systems (Reference 1).

^{††} The curves have been normalized to pass through the 1100 °K experimental points of Reynolds and Stewart (Reference 8).

by means of the FCC fundamental mode code⁽⁶⁾ with a modified version of the Russian 26-group data compilation as input.⁽⁷⁾

PART IV. DISCUSSION OF THE RESULTS

The effect on the ^{238}U Doppler coefficient due to crystalline binding (Figure 4) is quite significant in the low temperature range, where at room temperature (300 °K), for example, the reduction of the free gas value is 30%. In interpreting a low temperature critical experiment the calculated change in criticality should be reduced (by 15%, for example, for a temperature change of 300 °K to 1100 °K) to allow for binding effects.

An interesting result is seen in Figure 5, where the crystalline binding curve for reactivity diverges from the usual "ln T" rule in the low temperature region. This effect agrees with the experimental findings of Reynolds and Stewart⁽⁸⁾ for critical experiments on ZPR-III, Assembly 47.

For this study the Dolling and Debye phonon distributions only were used even though other distributions are available (see Reference 3). The essential results given in this paper, however, probably will not be changed by using the other distributions except for the detailed structure in the effective Debye temperature curves (Figure 3).

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FIGURE 1. Effective Temperature Versus Temperature for the Dolling Phonon Distribution and the Debye Distribution ($\theta_D = 620$ °K)

FIGURE 2. Dolling Phonon Distribution for ^{238}U in a UO_2 Lattice

FIGURE 3. Effective Debye Temperature Versus Temperature for the Dolling Phonon Distribution

FIGURE 4. Crystalline Binding Effects on the ^{238}U Doppler Coefficient for the Dolling Phonon Distribution

FIGURE 5. Reactivity Change Versus Temperature for the ZPR-III, Assembly 47 Including Binding Effects

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