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An Approach to the Analysis of Xenon Escape  
from  $\text{UO}_2$  during Postirradiation Heating

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**Abstract** The escape behavior of  $^{133}\text{Xe}$  from irradiated  $\text{UO}_2$  particle was studied by linear-temperature-rise and isothermal heating methods at temperatures below  $1700^\circ\text{C}$ . The results obtained were interpreted by Primak's analysis, and the activation energy spectrum, distributing in a wide range of the activation energy, for the release of  $^{133}\text{Xe}$  from traps was obtained. A unspecified process, which plays an important role in the initial short period of the isothermal heating, is involved in the transfer of the  $^{133}\text{Xe}$  released from traps.

A simple method for estimating the escape fraction of  $^{133}\text{Xe}$  in the isothermal heating is proposed, assuming an exponential distribution of the spectrum with activation energy. The method gives a fairly good estimation from only one datum of the escape.

照射後加熱時に  $\text{UO}_2$  から逃散する Xe の解析の一つの方法

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要旨 照射  $\text{UO}_2$  粒子からの  $^{135}\text{Xe}$  の逃散挙動を等速昇温加熱法と等温加熱法とによって  $1700^\circ\text{C}$  以下の温度で調べた。その結果を Primak の解析によって解釈したところ、 $^{135}\text{Xe}$  のトラップからの放出の活性化エネルギーは広い範囲に分布していることがわかった。また、トラップから放出された  $^{135}\text{Xe}$  のマトリックス内の移動には、等温加熱の初期にのみ影響を及ぼす過程が含まれていることがわかった。さらに、活性化エネルギーが指数関数的に分布するという仮定をおいて、等温加熱時の  $^{135}\text{Xe}$  の逃散率を推定する簡単な方法を導き、この方法により1ヶのデータによってもかなり精度のよい推定ができることを示した。

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## 1. Introduction

The escape of fission-gas from fuel material has been extensively investigated by many workers, but these studies have not explained the detailed mechanism of the escape process. It has been established that fission-gas escape is not a simple diffusion process following well-known mathematical laws. The model commonly accepted at present for fission-gas migration through a fuel involves the trapping of gas at imperfections and the release from them. Yajima et al.<sup>1)-3)</sup> made a study on the escape of  $^{133}\text{Xe}$  and  $^{131}\text{I}$  from graphite into which fission products had recoiled, and their experimental results were explained qualitatively by their trap model in which the  $^{133}\text{Xe}$  and  $^{131}\text{I}$  were assumed to be trapped before annealing. Carroll and Sisman<sup>4)</sup> made in-pile measurements on single-crystal  $\text{UO}_2$  and proposed a defect-trap model in which the fission-gas atoms were assumed to be diffusible at first and then trapped on way of diffusion. MacEwan and Stevens<sup>5)</sup> suggested a trap mechanism, and MacEwan and Morel<sup>6)</sup> assumed that up to 70 % of the xenon was in traps at the start of an anneal and used Hurst's analysis<sup>7)</sup> to explain the results. Auskern<sup>8)</sup> tried to interpret experimental results on the escape of xenon from UC at temperatures below about  $1000^\circ\text{C}$  by the analysis of Primak<sup>9)</sup> for a process occurring over a range of activation energies. Gautsch et al.<sup>10)</sup> also interpreted the results of krypton escape from thin layers of  $\text{UO}_2$  by the Primak's analysis. For the escape of fission products introduced into natural graphite by recoil, one of the present authors proposed an escape mechanism, consisting of three successive processes; release from trapping sites, diffusion of the released fission products, and desorption from the surface. The validity of this mechanism was substantiated by analysing the observed data for  $^{133}\text{Xe}$ <sup>11),12)</sup> and other non-gaseous fission products<sup>13)</sup>, in which the release from the trapping sites was treated kinetically by Primak's analysis. Furthermore, the escape rate of fission gas from fuel during irradiation was similarly analysed, and experimental data were explained satisfactorily, including the dependence of escape rate on decay constant, the contribution of precursor, and the change of escape rate with irradiation time<sup>14)</sup>. Yajima et al.<sup>15)</sup> measured a  $^{133}\text{Xe}$  escape from  $\text{UO}_2$  powder or pellet at temperatures up to  $2025^\circ\text{C}$ , and concluded that an annealing of trap might control the escape process below the temperature at which grain growth in  $\text{UO}_2$  commenced, and diffusion might control above that temperature.

In the present study, the escape of  $^{133}\text{Xe}$  from  $\text{UO}_2$  particles below  $1700^\circ\text{C}$  is measured and the results are discussed in relation to a mechanism

controlling the escape.

## 2. Experimental

### 2.1. Experimental Method

Two kinds of fused  $\text{UO}_2$ , each of which was prepared by a different manufacturer, were ground manually into -325/+400 mesh particles. These two powdered  $\text{UO}_2$ 's, each with an O/U ratio of about 2.008, are designated  $\text{UO}_2(\text{I})$  and  $\text{UO}_2(\text{II})$ . They were irradiated in JRR-2 for 10 min. at a neutron flux of about  $3 \times 10^{13}$  n/cm<sup>2</sup>.sec. After cooling 1 to 2 weeks, the irradiated  $\text{UO}_2$  was heated in a furnace (see Fig. 1). In the isothermal heating, the irradiated  $\text{UO}_2$  sample in a small boat (an alumina boat if the temperature were to reach 1700°C, otherwise a platinum boat) was first placed in the top portion of a furnace tube, and the interior of the tube was evacuated to below  $10^{-4}$  mm Hg pressure. Then, continuing the evacuation the furnace temperature was raised to any desired value in the range from 300° to 1400°C. Purified He was then passed and the boat was lowered till it touched the top end of the thermocouple. In the linear-temperature-rise heating, a  $\text{UO}_2$  sample was placed in the same thermocouple position, and the tube evacuated. Then He gas was passed and the temperature was raised at 5°C/min from room temperature to 1100°C for  $\text{UO}_2(\text{I})$  and to 1700°C for  $\text{UO}_2(\text{II})$ . For  $\text{UO}_2(\text{II})$ , the heating was done twice. That is, after reaching 1675°C in the first heating the sample was cooled down to room temperature (the first cooling), and then heated again in the same manner to 1690°C (the second heating), followed by cooling down to room temperature (the second cooling).

The He gas was purified by passing through a cold trap cooled with dry ice, and through a bed of Ti heated at 800°C and another of Ca at 700°C, in that order. The gas flow rate was set at 100 ml/min. Escaped  $^{133}\text{Xe}$  was collected by a charcoal trap cooled with dry ice, after passage through a KOH bed and a cold trap cooled with dry ice. The charcoal trap was replaced by a new one at appropriate intervals in order to follow the amount of  $^{133}\text{Xe}$  escape as a function of heating time. After completion of the heating, the  $\text{UO}_2$  sample was dissolved in concentrated  $\text{HNO}_3$  and the  $^{133}\text{Xe}$  evolved was collected on charcoal to obtain the residual amount of  $^{133}\text{Xe}$  in the heated  $\text{UO}_2$ . The gamma activity of  $^{133}\text{Xe}$  in the charcoal traps was measured.

## 2.2. Experimental Results

### (1) Linear-Temperature-Rise Heating

The experimental results for the linear-temperature-rise heating are shown in Fig. 2; the change in the fractional escape rate is plotted against the average temperature during the sampling interval. The fraction of  $^{133}\text{Xe}$  escaped from  $\text{UO}_2(\text{I})$  after the heating was 0.00133. The fractions escaped from  $\text{UO}_2(\text{II})$  were 0.0778 in the first heating, 0.0112 in the first cooling, 0.0529 in the second heating, and 0.0262 in the second cooling. Figure 2 shows that: 1) The escape rate for  $\text{UO}_2(\text{II})$  was about five times larger than that for  $\text{UO}_2(\text{I})$ , 2) The rate increased remarkably with increase of the temperature, 3) The rate passes through maxima at certain temperatures, at Yajima et al.<sup>15)</sup> observed, 4) For  $\text{UO}_2(\text{II})$ , the rate in the second heating was negligible below about  $1300^\circ\text{C}$ ; while, the rate below  $1300^\circ\text{C}$  during the first heating was considerably large. The rate in the second heating increased exponentially with temperature above  $1300^\circ\text{C}$ , the rate at  $1650^\circ\text{C}$  being nearly equal to that in the first heating, 5) In both first and second coolings of the  $\text{UO}_2(\text{II})$  sample, the escape rate decreased rapidly, and the phenomenon of a cooling burst did not occur.

### (2) Isothermal Heating

The results for  $\text{UO}_2(\text{I})$  in the temperature range of  $400^\circ$  to  $1022^\circ\text{C}$  and for  $\text{UO}_2(\text{II})$  at  $300^\circ\text{C}$  are shown in Fig. 3, where the fraction of  $^{133}\text{Xe}$  remaining in the  $\text{UO}_2$  is plotted against the logarithm of annealing time. The point of origin in time marking the practical initiation of the isothermal annealing was determined conventionally in a manner similar to that for  $^{133}\text{Xe}$  escape from graphite<sup>12)</sup>, as described below. When the sample is inserted into the furnace center, the temperature measured just below the sample boat registers a small dip followed by a rise to regain the preset value, and in the meantime the  $^{133}\text{Xe}$  fraction remaining starts to fall off gradually at first, then more steeply during the period of temperature rise. This sequence of events is shown schematically in Fig. 4. Thus neither the instant of the sample insertion nor that of any referable point in the temperature record can be regarded as the point of annealing initiation. This point was determined as a conventional method by plotting the remnant  $^{133}\text{Xe}$  fraction on a logarithmic scale against time and extrapolating the initial portion of the decrease branch to the ordinate value of unity (see Fig. 4). Thus the point of annealing initiation may be

determined by giving consideration to the temperature increase of a sample after insertion and to the time lag from the escape of  $^{133}\text{Xe}$  from the sample to the collection by charcoal trap. The fraction of  $^{133}\text{Xe}$  remaining in  $\text{UO}_2(\text{I})$  after heating at  $1400^\circ\text{C}$  for 3 hrs was 0.9825. In this case, time dependence of the escape was not followed and the annealing was regarded to start from the instant of the sample insertion into the furnace, since the heating time was long enough not to be influenced markedly by the correction of the annealing initiation described above.

It is seen from Fig. 3 that a heating burst occupies a short initial period, followed by slower escape rate. The fraction remaining during the slower step seems to be related linearly to the logarithm of annealing time except for heating at  $1022^\circ\text{C}$ . The temperature-dependence of  $^{133}\text{Xe}$  escape does not appear simple, and at present it can only be qualitatively stated that it shows an increasing tendency with increasing temperature. These results are quite similar to those of fission product escape from graphite<sup>12), 13)</sup>.

### 3. Discussions

#### 3.1. Interpretation of $^{133}\text{Xe}$ Escape

Although it is established that fission-gas escape is not a simple diffusion process, this is clearly demonstrated by the experimental results indicating that maxima in the escape rate appeared in the first linear-temperature-rise heating and they disappeared in the second heating. In the escape of  $^{133}\text{Xe}$  from graphite into which fission products had been introduced the maxima in the escape rate were observed, and the escape was explained as being controlled by the release from traps<sup>11), 12)</sup>. Therefore, as one of the methods to interpret the experimental results, it may be permitted to assume that the escape is controlled by the mechanism similar to that for the escape from the graphite. Then, for the release process from traps, the analysis of Primak, which will be briefly described in Appendix I, may be used in order to obtain the activation energy spectrum. The spectra obtained is shown in Fig. 5. The value of  $\beta_0$  was determined as  $10^{10} \text{ min}^{-1}$  by applying the relation given by Eq. (A2) both to the observed isothermal data and the step-heating data at the end of the heating run for  $\text{UO}_2(\text{I})$ , and the same value was assumed for  $\text{UO}_2(\text{II})$ . It is seen that the spectra for  $\text{UO}_2(\text{I})$  obtained from both the linear-temperature-rise and



isothermal heatings show a fair agreement, except the portions of the spectrum shown by dotted lines which were obtained from the data for the initial short period of the isothermal heating. This agreement shows clearly that the Primak's analysis can be applied to the  $^{133}\text{Xe}$  escape during the postirradiation heating.

The existence of dotted portions in the spectrum suggests that a certain kind of resistance to the transport of  $^{133}\text{Xe}$  in  $\text{UO}_2$  is involved, and this resistance played an unimportant role in the slower step. This kind of resistance was also observed in the escape of  $^{133}\text{Xe}$  from graphite, and it was regarded that the resistance was caused by the diffusion of the xenon released from traps through interlaminar spaces of the graphite crystal. One of the interpretations for the above resistance is shown in Appendix II, where the diffusion of the  $^{133}\text{Xe}$  released from traps in a spherical  $\text{UO}_2$  matrix is assumed. The activation energy for diffusion was obtained as 0.047 eV/atom, which is, however, too small to depict a volume diffusion in the  $\text{UO}_2$  matrix, unless a transport in an interconnected pore or a grain boundary occurs. Another interpretation arises from the assumption that all the  $^{133}\text{Xe}$  escaped during the heating originated on or near the surface of the  $\text{UO}_2$  and the resistance stated above was caused by a diffusion through the matrix near the surface. The analysis of the data by this assumption would give also a small value of activation energy for diffusion, probably close to the value given above. Therefore, in order to interpret the observed resistance properly, further work will be necessary on the nature of the traps and the  $\text{UO}_2$  surface in connection with radiation damage of the  $\text{UO}_2$  matrix.

### 3.2. Simple Method for Estimating the Escape Fraction of $^{133}\text{Xe}$ during Postirradiation Heating

The results shown in Fig. 2 indicate that the escape rate can be regarded to increase exponentially with temperature if the maxima are ignored. The same tendency will be found in the reported data shown in Fig. 6, in which results of Yajima et al.<sup>15)</sup>, Booth and Rymer<sup>16)</sup>, and Parker et al.<sup>17)</sup> are reproduced. The temperature dependence of the escape rate by Yajima et al. may be taken as nearly the same as that in Fig. 2, if the maxima are ignored. The results, obtained by the isothermal heating for constant period, of the latter two researchers indicate that the escape fraction is considered to increase exponentially with temperature. Since

the differentiation of the escape fraction with temperature at constant time of the heating may be regarded, as a first approximation, to correspond to the escape rate in the linear-temperature-rise heating, the escape rate expected from the above two results may be regarded to increase exponentially with temperature holding the same slope as appeared in the plots of the escape fraction against temperature. Here, it is to be noted that the slope in the plots of the escape fraction against temperature is regarded to be nearly same to that of Yajima et al. (see Fig. 6) or of the present study (see Fig. 2).

Since the escape of  $^{133}\text{Xe}$  from  $\text{UO}_2$  at temperatures below about  $1500^\circ\text{C}$  could be mathematically described by Primak's analysis, as in the preceding section, except the initial burst, the above consideration may permit us to assume that the escape of  $^{133}\text{Xe}$  from other  $\text{UO}_2$  is also described by Primak's analysis. Then the activation energy spectrum may be regarded to be exponentially increasing with activation energy  $\epsilon$  if the value of  $\beta_0$  is taken as constant and the maxima in the spectrum are ignored. Then the distribution of the spectrum,  $p(\epsilon, 0)$ , is expressed by

$$p(\epsilon, 0) = A e^{B\epsilon}, \quad (1)$$

where  $A$  and  $B$  are constants inherent to a kind of  $\text{UO}_2$  sample, and  $\epsilon$  is defined between the minimum  $\epsilon_1$  and the maximum  $\epsilon_2$ . When the escape of  $^{133}\text{Xe}$  is limited only by the release from the traps, the fraction,  $F(\tau, t)$ , of  $^{133}\text{Xe}$  remaining in  $\text{UO}_2$  at  $\tau$  and  $t$  (definition is given in Appendix I) is expressed by

$$F(\tau, t) = \int_{\epsilon_0}^{\epsilon_2} p(\epsilon, 0) d\epsilon = \frac{A}{B} (e^{B\epsilon_2} - e^{B\epsilon_0}). \quad (2)$$

Putting Eq. (A2) into this equation,

$$F(\tau, t) = \frac{A}{B} (e^{B\epsilon_2} - (\beta_0 t)^{B\tau}) \quad (3)$$

is obtained. Since  $F(\tau, 0) = 1$ , Eq. (3) is written finally as

$$F(\tau, t) = 1 - \frac{A}{B} (\beta_0 t)^{B\tau} \quad (4)$$

This equation enables us to estimate  $F(\tau, t)$  if the values of  $A$ ,  $B$ , and  $\beta_0$  are known from, at least, three values of  $F(\tau, t)$ .

When the values of  $B$  and  $\beta_0$  are known, only one value of  $F(\tau, t)$  is enough to estimate  $F(\tau, t)$ . The value of  $B$  may be obtained from the spectrum shown in Fig. 5 and from the results given in Fig. 6 by assuming  $\beta_0$  as  $10^{10} \text{ min}^{-1}$  as in the present study. Then the value of  $B$  is determined as  $1.90 \text{ eV}^{-1}$  for the  $\text{UO}_2$  samples discussed. Thus Eq. (4) can be written as

$$F(\tau, t) = 1 - \frac{A}{1.90} (10^{10} t)^{1.90\tau} \quad (5)$$

Examples of the calculation are given below. From Fig. 3,  $F(\tau, t) = 0.99867$  at  $850^\circ\text{C}$  and  $t = 140 \text{ min}$  was chosen, and estimated values of  $F(\tau, t)$  are shown in Fig. 7. It is seen that the estimated and observed values agree fairly well at temperatures from  $400^\circ\text{C}$  to  $1400^\circ\text{C}$ , except the observed values at  $1022^\circ\text{C}$ . Another example of the estimation was done for the data of Booth and Rymer. The observed data and estimated values using the data at  $1200^\circ\text{C}$  are compared in Fig. 6, in which an agreement is seen to be fairly well. These results suggest that the assumptions involved in deriving Eq. (5) are adequate and the estimation of  $F(\tau, t)$  in the slower escape step of the isothermal heating can be done with a fairly good approximation, without complicated calculation, by the above method. The result that the Primak's analysis could be applied to the escape process of  $^{133}\text{Xe}$  from  $\text{UO}_2$  during the postirradiation heating will be considered as a proof to the assumption taken into the mathematical treatment for fission gas escape during the isothermal irradiation reported before<sup>14)</sup>.

#### 4. Conclusion

Data obtained for  $^{133}\text{Xe}$  escape from  $\text{UO}_2$  during a linear-temperature-rise and an isothermal heating indicated definitely that the escape was not controlled by a simple diffusion below temperature at which grain growth in  $\text{UO}_2$  might occur. Experimental results on the linear-temperature-rise and isothermal heating were interpreted by the Primak's analysis, and the activation energy spectrum, distributing in a wide range of the activation energy, for the release of  $^{133}\text{Xe}$  from traps could be obtained. It was pointed out that a certain process which played an important role in the initial burst process was involved in the transfer of the  $^{133}\text{Xe}$  released from traps. Interpretation for this process suggested that the transfer resistance of the released  $^{133}\text{Xe}$  in the  $\text{UO}_2$  was not large.

A simple method for estimating the escaped fraction of  $^{133}\text{Xe}$  in the isothermal heating was proposed, assuming an exponential distribution of the spectrum with activation energy. It was made clear that the escape fraction could be roughly estimated at temperatures below about  $1500^{\circ}\text{C}$  when a single datum of the isothermal heating was given.

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## Appendix I. Release from Trapping Sites

The mathematical treatment for the release process, based upon Primak's analysis, was reported previously<sup>11)-13)</sup>. Here, only the result for isothermal annealing is briefly reproduced. The release rate from trapping sites,  $-dP(t)/dt$ , is given by

$$-\frac{dP(t)}{dt} = \frac{\tau}{t} p_0(\epsilon_0) \quad (\text{A1})$$

where  $P(t)$  is the quantity of  $^{133}\text{Xe}$  trapped at an annealing time  $t$  in the trapping sites,  $\tau$  is the product of the Boltzmann's constant and the absolute temperature, and  $p_0(\epsilon)d\epsilon$  is the concentration of  $^{133}\text{Xe}$  trapped in the sites with activation energy between  $\epsilon$  and  $\epsilon+d\epsilon$  before annealing occurs. The characteristic activation energy  $\epsilon_0$  is expressed by

$$\epsilon_0 = \tau \ln(\beta_0 t) \quad (\text{A2})$$

where  $\beta_0$  is a constant having the dimension of frequency. A plot of  $p_0(\epsilon)$  against  $\epsilon$  is termed an initial activation energy spectrum, and is given with good approximation by a plot of  $p_0(\epsilon_0)$  against  $\epsilon_0$ . The method for determining  $\beta_0$  is written in the previous report<sup>12)</sup>.

Appendix II. Analysis of Diffusion of the Released  $^{133}\text{Xe}$ 

The basic diffusion equation for the  $^{133}\text{Xe}$  atoms released from trapping sites \_\_\_\_\_ granted that the diffusion occurs radially in sphere of radius  $a$  \_\_\_\_\_ is

$$\frac{\partial c}{\partial t} = D \left( \frac{\partial^2 c}{\partial r^2} + \frac{2}{r} \frac{\partial c}{\partial r} \right) + \left( -\frac{dP(t)}{dt} \right), \quad (\text{A3})$$

where  $c$  is the concentration of diffusing  $^{133}\text{Xe}$ ,  $r$  the radial distance and  $D$  the diffusion coefficient. The initial and boundary conditions are:

$$\left. \begin{array}{l} t = 0; \quad c = 0 \quad \text{at} \quad 0 \leq r \leq a \\ t = 0; \quad c = 0 \quad \text{at} \quad r = a \end{array} \right\} \quad (\text{A4})$$

It is assumed that all the  $^{133}\text{Xe}$  atoms are trapped at  $t = 0$ , and there is no resistance to the transport of  $^{133}\text{Xe}$  at the surface.

A general solution for Eq.(A3) can not be derived unless the release rate  $-dP(t)/dt$ , which depends upon the initial activation energy spectrum, is given explicitly. However, when the distribution of the spectrum is assumed to be uniform as shown in Fig. A1, that is, the spectrum distributes uniformly with the value  $\bar{p}_0$  over a range of activation energies from  $\epsilon_1$  to  $\epsilon_2$ , the solution of Eq.(A3) may be obtained as described below. The  $b$  and  $(b+t_2)$  are the heating times corresponding to  $\epsilon_1$  and  $\epsilon_2$ , respectively, and are given by

$$\epsilon_1 = \tau \ln(\beta_0 b), \quad (A5)$$

$$\epsilon_2 = \tau \ln[\beta_0 (b+t_2)]. \quad (A6)$$

The release rate in this case is given approximately by  $\bar{p}_0 \tau / (b+t)$  since  $b$  is regarded as much smaller than  $t^*$ . The solution of Eq.(A3) is

$$c = \frac{-2a\bar{p}_0\tau}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n\pi r}{a} e^{-Dn^2\pi^2(b+t)/a^2} \Delta \bar{E}_1\{n,b,t\}, \quad (A7)$$

where

$$\Delta \bar{E}_1\{n,b,t\} \equiv \bar{E}_1(Dn^2\pi^2(b+t)/a^2) - \bar{E}_1(Dn^2\pi^2b/a^2) \quad \left. \vphantom{\Delta \bar{E}_1\{n,b,t\}} \right\} \quad (A8)$$

$$\bar{E}_1(x) = \int_{-\infty}^x \frac{e^u}{u} du$$

Then the fraction  $F$  of  $^{133}\text{Xe}$  remaining in the  $\text{UO}_2$  is

$$F = \frac{\ln\left(\frac{b+t_2}{b+t}\right) + X}{\ln\left(\frac{b+t_2}{b}\right)}, \quad (A9)$$

where

$$X \equiv \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-Dn^2\pi^2(b+t)/a^2} \Delta \bar{E}_1\{n,b,t\}. \quad (A10)$$

It is seen from Eq.(A9) that  $F$  consists of a release reaction term  $F_1$  and a diffusion term  $F_2$ :

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\* The actual value of  $b$  was 0.032 min at 400°C and 0.24 min at 750°C in this study.

$$F_1 = \frac{\ln\left(\frac{b+t_2}{b+t}\right)}{\ln\left(\frac{b+t_2}{b}\right)}, \quad (\text{A11})$$

$$F_2 = \frac{X}{\ln\left(\frac{b+t_2}{b}\right)}. \quad (\text{A12})$$

The situation can be represented as in Fig. A2, in which  $F$  is shown against  $\ln(b+t)$ . The values of  $b$  and  $(b+t_2)$  are obtained from the linear relationship between  $F_1$  and  $\ln(b+t)$ . Then the value of  $D$  can be determined from the relationship between the observed value of  $F_2$  and  $\ln(b+t)$  if the escape mechanism is valid.

The experimental results shown in Fig. 3 indicate that the fraction remaining during the slower step is proportional to the logarithm of the annealing time, as a first approximation. This linear relationship was considered to be expressed by Eq.(A11) and the values of  $b$  and  $(b+t_2)$  were determined. Then, by application of Eq.(A12) to the data observed in the initial short period at a specified temperature the value of  $D/a^2$  was determined. The values of  $D/a^2$  obtained were found to be independent of the time, and its temperature dependence is shown in Fig. A3, which can be expressed by

$$\frac{D}{a^2} = 6 \times 10^{-2} e^{-0.047/\tau} \quad (\text{A13})$$

It is seen that  $D/a^2$  has a fairly large value, the activation energy for diffusion being 0.047 eV/atom. The value of  $D/a^2$  thus determined will be somewhat larger than the true value of  $D/a^2$ , since the actual distribution of the spectrum was not uniform but it increased rapidly with increasing activation energy.

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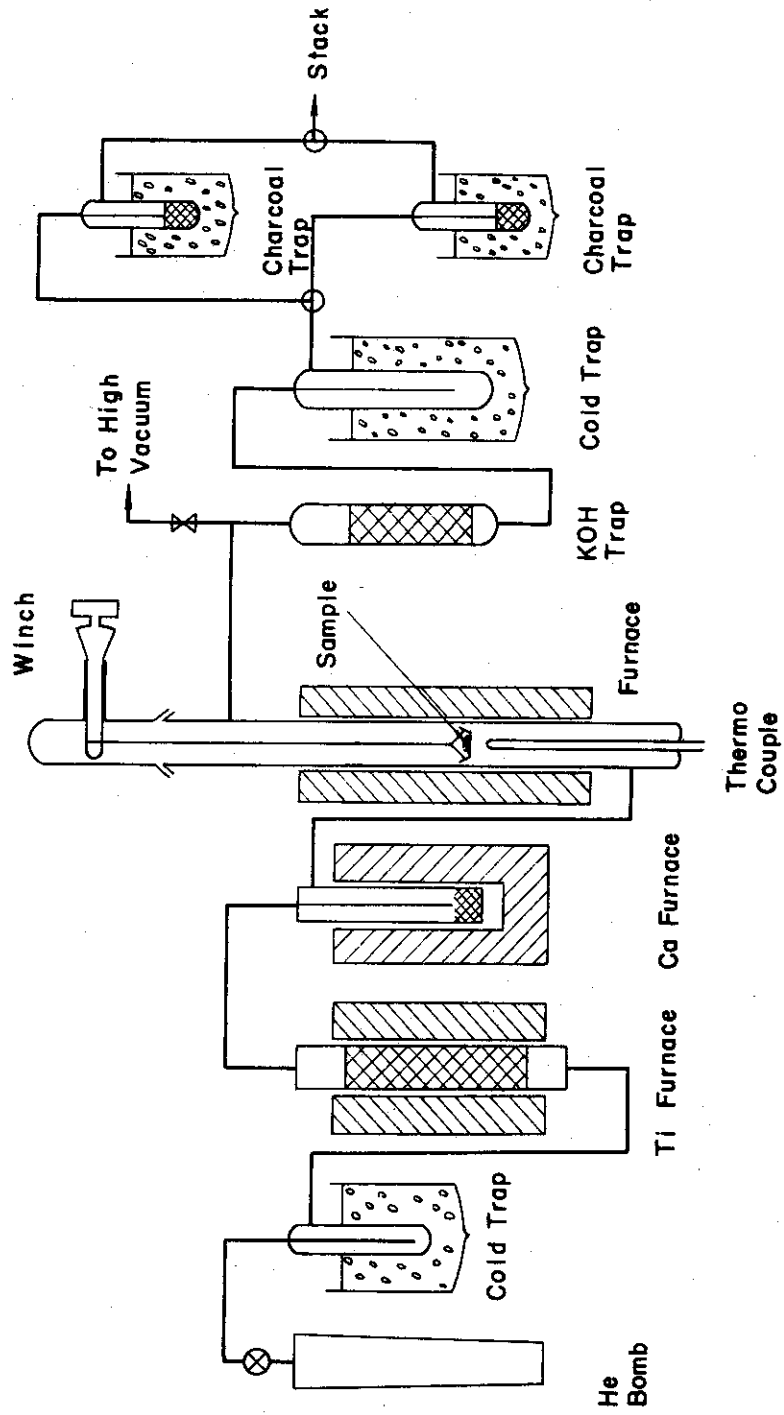


Fig. 1 Experimental Apparatus

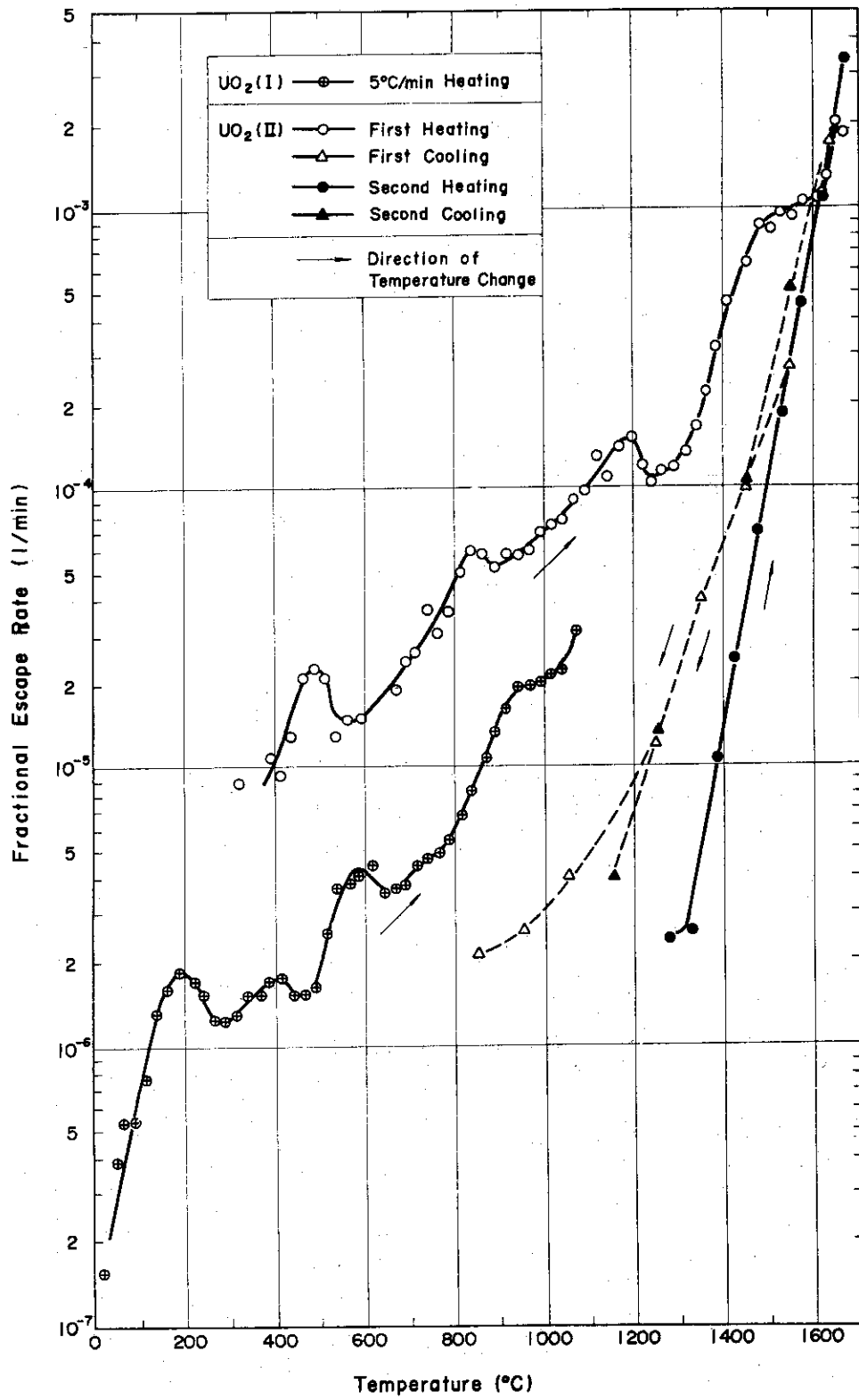


Fig. 2 Experimental Results for 5°C/min Heating

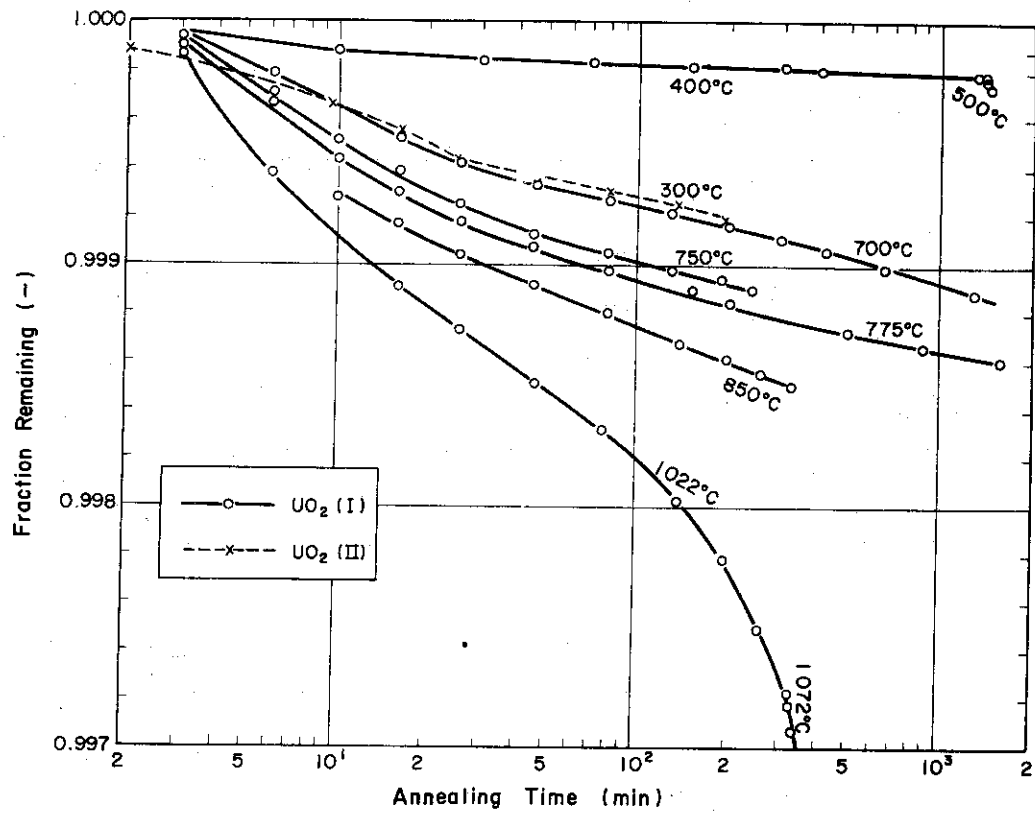


Fig. 3 Experimental Results for Isothermal Heating

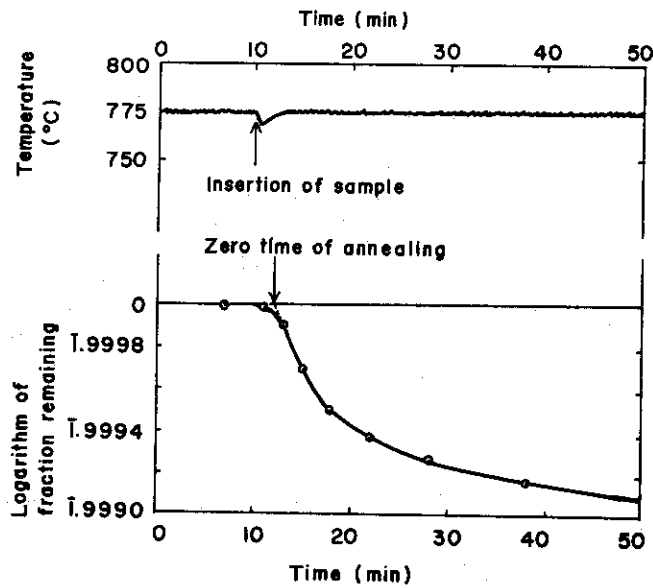


Fig. 4 Conventional Determination of the Zero Time of Isothermal Annealing

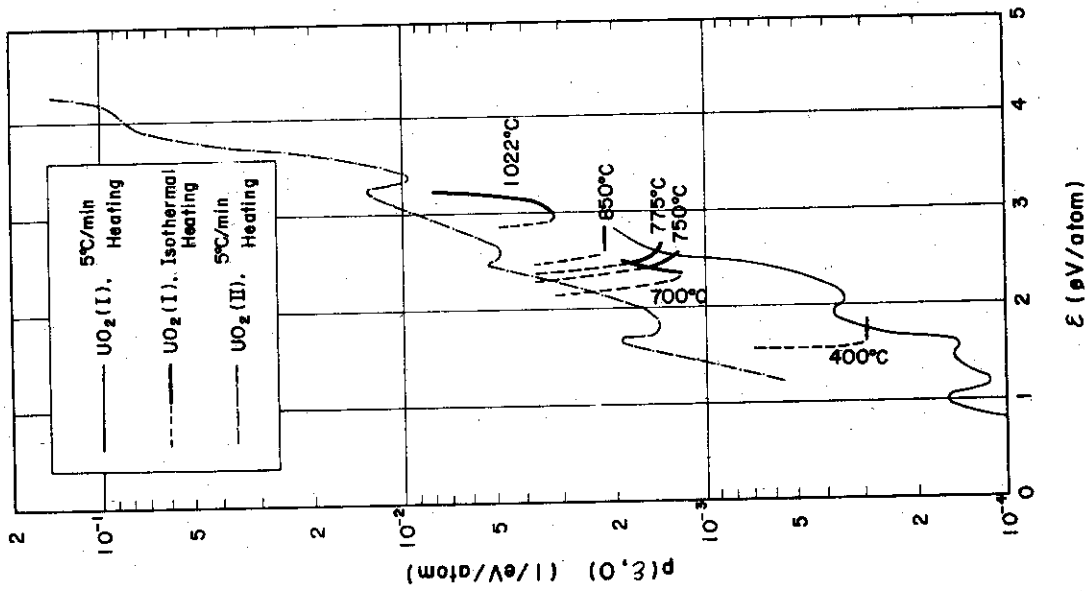


Fig. 5 Initial Activation Energy Spectrum for Trapped  $^{133}\text{Xe}$

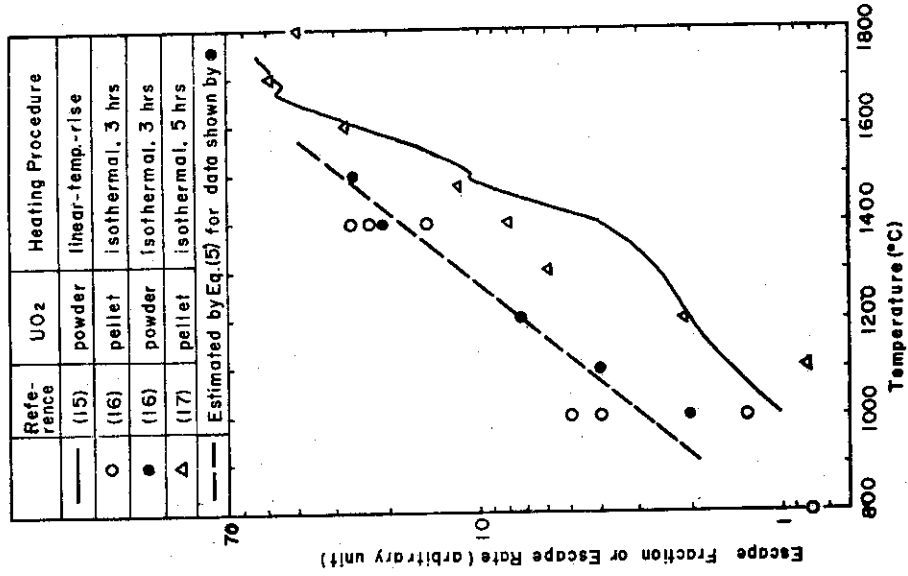


Fig. 6 Some Reported Data on  $^{133}\text{Xe}$  Escape

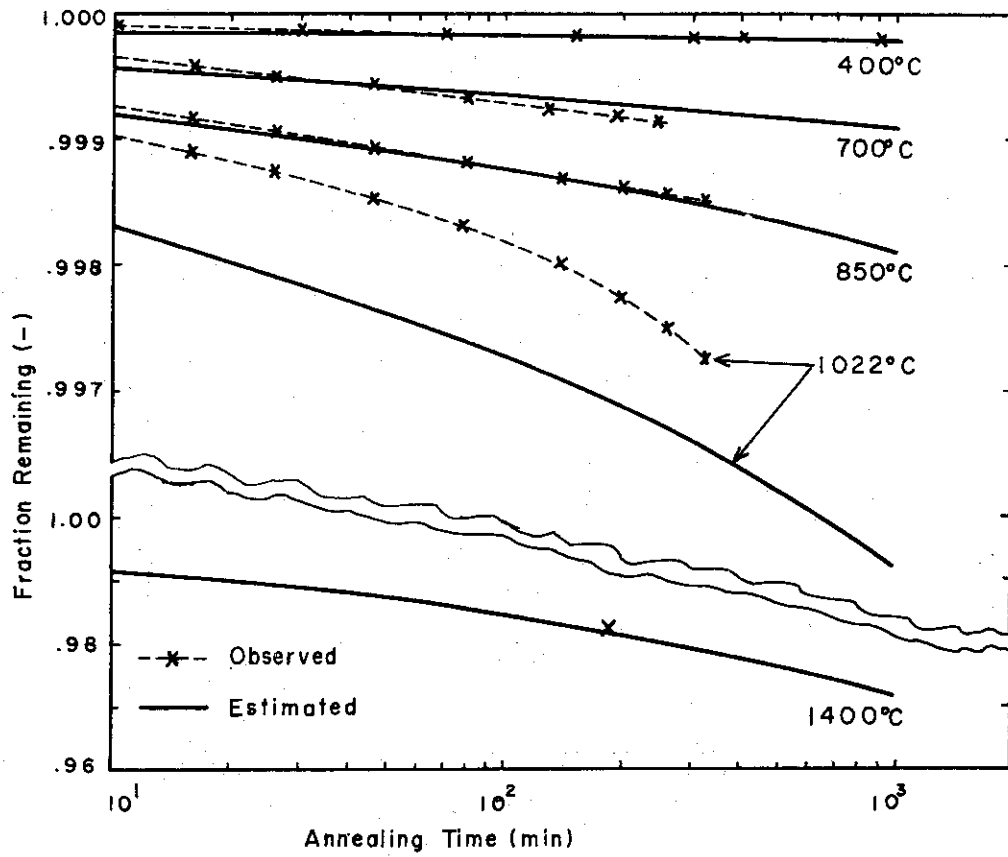


Fig. 7 Comparison of Estimated Data with Observed Ones

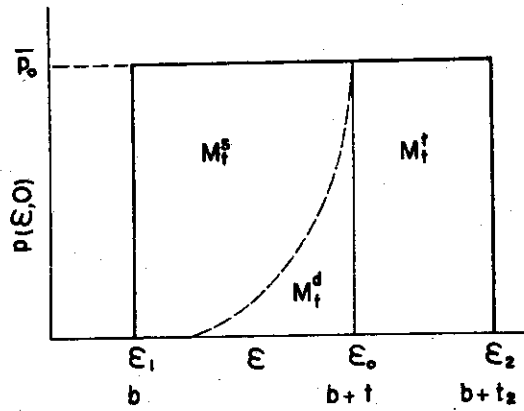


Fig. A1 The Initial Activation Energy Spectrum Having Uniform Distribution ( $M_i^t$  indicates the amount of  $^{135}\text{Xe}$  trapped,  $M_i^d$  that diffusing,  $M_i^s$  that escaped).

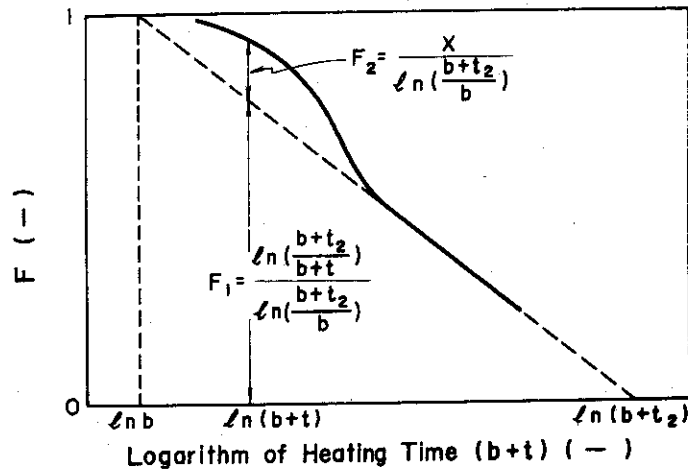


Fig. A2 Relation between F and  $\ln(b+t)$

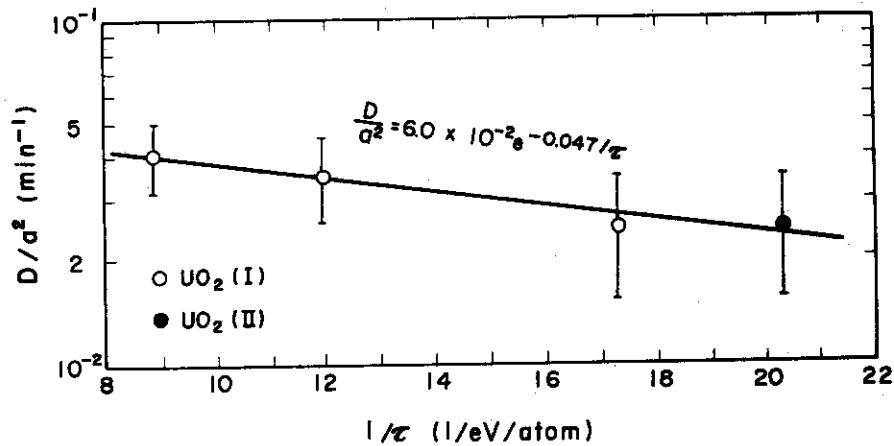


Fig. A3 Dependence of  $D/a^2$  upon Temperature