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APPLICATION OF PULSED ELECTRON BEAM VAPORIZATION TO STUDIES OF UO_2

D. A. Benson

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APPLICATION OF PULSED ELECTRON BEAM
VAPORIZATION TO STUDIES OF UO_2 *

by

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ABSTRACT

A method for determining the pressure versus internal energy coordinates of the liquid-vapor saturation curve is applied to the study of UO_2 . The experimental details and results of an initial series of tests are described. A comparison of the measurement results to models of the UO_2 equation of state illustrates the role of the heat capacity in describing the P-E characteristics of the state surface. A discussion of the available heat capacity information suggests that additional modeling and measurements of the heat capacity may be needed to give a complete temperature and energy dependent state surface description. Because of these modeling uncertainties, a method of thermodynamically describing the P(V, E) state surface entirely through the use of dynamic vapor measurements is given. Such a model satisfies transient thermomechanical analysis requirements. Next the effect of the state surface on one type of core disruptive reactor analysis is examined. And finally, the property determinations and models for UO_2 are reviewed with requirements for future work being outlined.

*This work supported by the U. S. Energy Research and Development Administration

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I. INTRODUCTION

Extensive hypothetical core disassembly accident (HCDA) analysis^{1,2,3} has been performed to assess consequences of a fuel vapor driven hydrodynamic disassembly of a partially voided liquid metal fast breeder reactor (LMFBR) core. Peak fuel temperatures in these calculations range from 4000 to 7000 K, a temperature range where limited knowledge of fuel vapor properties exists to support such analysis. To fill this gap in information, initial experiments have now been carried out using a new electron beam heating technique to determine UO_2 properties under the pressure and internal energy conditions involved in such calculations. The high temperature vapor data is also needed to interpret the mechanism of pressure generation and related fuel motion in pulsed test reactor experiments carried out on prototypic reactor fuel pins. Measurement and description of the high temperature vapor properties are the subjects of this report.

Previous UO_2 vapor information has been based upon relatively low temperature static measurements.^{4,5,6} Included in these methods are vapor effusion, transpiration and spectroscopic techniques. Extrapolations of such data by three to four orders of magnitude or more in vapor pressure have been used to apply such results to accident analyses. More recently, pulsed laser heating experiments have given vapor data^{7,8} as a function of temperature for higher pressures. Potential problems related to high vaporization rates in these free evaporation experiments have recently led some to question the technique validity at the highest temperatures and pressures.⁹ Also worth consideration is the fact that transient thermomechanical calculations, of which the HCDA analysis is an example, require vapor information in a pressure versus energy format. Using the pressure versus

temperature data in this application requires the extreme temperature heat capacity to be measured or estimated. This heat capacity information, as we shall see later in the report, is not well defined and strongly affects the pressure energy relation. A new means of directly measuring the saturation curve pressure versus energy coordinates over the high pressure and energy range of interest for fuel materials will be described.

The new method uses a pulsed electron accelerator to heat vapor samples confined by a graphite container material. Measurements of the sample pressure and energy state are made in each pulsed experiment. The means for determining the UO_2 vapor pressure and specific energy states will be described in Section II. More discussion of the method can be found in a recent paper.¹⁰

In addition to describing the method of measurement, this report will present initial results for the vapor pressure of UO_2 .¹⁸ A comparison of the data to currently used estimates of the high temperature equation of state (EOS), will be made. Also, a new representation of the vapor EOS which allows a direct description in the $P(E, V)$ format required for calculations of transient thermomechanical phenomena will be described.

II. EXPERIMENTAL DISCUSSION

The vaporization samples in this work were heated by the electron beam from the Sandia Relativistic Electron Beam Accelerator (REBA).¹¹ This accelerator was modified¹² to couple the Marx high voltage generator directly to a field emission diode which produces the electron beam. A pulsed magnetic solenoid directs the beam from the diode to the target region. The accelerator's beam for these measurements had a 1.2 MeV average electron energy spectrum and a 50 kA peak current. Its energy pulse is approximately Gaussian in time with a .6 μ s (full width at half maximum) duration. Electron energy in excess of 20 kJ is delivered to the target plane. By varying the beam cross-sectional area, a specific energy (energy/mass) absorbed by the target may be selected over a range from low levels to greater than 4000 J/g.

The vapor pressure measurements are carried out with apparatus shown schematically in Fig. 1. Each target is composed of a thin vaporization sample sandwiched between two thin graphite pistons pressed into a graphite cylinder thus forming a crucible which confines the vapor. The target is positioned in the electron beam path where it is heated by an electron pulse. After the sample is heated, the pistons which inertially confine the mixed liquid-vapor material are accelerated in opposite directions by the vapor pressure corresponding to the specific energy absorbed by the vapor sample. Two types of observations are made during each experiment. First, the normal displacement of each target piston is measured by streak camera photography. The piston expansion trajectories obtained from these records together with simple equations of motion are used to determine the pressure of the vapor sample. Second, the specific energy absorbed at the center

Fig. 1. Schematic view of the pulsed vaporization experiment. A thin UO_2 vapor sample is confined between pistons in the graphite vapor container. Pulsed-electron-beam vaporization of the sample induces container expansion which is monitored by a streak camera. An optical pyrometer measurement and other beam diagnostics determine the vapor sample energy state.

of a calorimeter plate positioned behind the target is measured with a time resolving optical pyrometer.¹³ This specific energy is closely related to the specific energy of the vapor sample since the target and dosimeter are thin compared to the range of the electrons. These two measurements give the total vapor pressure as a function of energy along the liquid saturation curve. The energy state, which is volume dependent, closely corresponds to a value at the fully liquid end of the saturation curve.

The function of the graphite crucible in this experiment is important. The large heat capacity per gram of carbon, relative to UO_2 , forces the initial graphite temperature to remain below 2000 C, while the liquid-vapor sample reaches a temperature many times that value. The short duration of the experiments (from 2 to 20 μs) and the low thermal conductivity of the vapor phase, limit thermal loss from the vapor sample to the graphite container during the vapor driven expansion. Thus, the expansion path traced by an experiment is approximately isentropic. For mixed phase states away from the critical region, an isentrope closely corresponds to isobaric expansion thereby giving displacement profiles which are parabolic in time. The significance of the small deviations from constant pressure during adiabatic expansion is discussed in Ref. 10. Also, deviations from adiabatic conditions, and non-equilibrium thermal effects, could lead to an under-estimation of the vapor pressure; hence a lower bound of the dynamic vapor pressure is determined.

One streak camera record of a vapor driven crucible expansion is shown in Fig. 2. The motion is photographed by backlighting the graphite crucible with flashlamp illumination, as shown at the left of this figure. The shadow

Fig. 2. Schematic diagram of the streak recording system and an example of one experiment record. As shown on the left, the shadows from tabs on the target faces are cast through a slit onto a moving film image thereby producing the streak record. A 50- μs -duration photo of a target expansion is shown at the right. The expansion asymmetry is due to the different piston masses on opposite sides of the target.

image of the target cross-section is projected through the slit and into the camera. The camera then records the motion of each graphite piston face. The shadow image on the slit is cast by small tabs centered on each graphite piston face to insure that the shadow motion measures an actual expansion, rather than a rotation, of the crucible. Photographs such as the one in Fig. 2 are optically scanned to produce numerical values for the graphite piston position as a function of time. Results for the UO_2 tests in this form will be discussed in the next section of this report.

The energy density in the vapor sample is determined by calorimetry and calculations based upon electron beam diagnostic information. During each vapor experiment, the temperature rise at the center of a graphite calorimeter plate positioned behind the sample is measured. This temperature rise is determined by infrared optical pyrometry after the electron pulse, but before thermal diffusion alters the initial temperature distribution. The measurement must also be completed before the calorimeter plate is destroyed by the rapidly expanding vapor container. Enthalpy data¹³ for the graphite dosimeter material is used to relate the measured temperature rise to the absorbed energy density at the plate surface.

The relation between energy density measured at the calorimeter plate position and the sample energy density is based upon electron transport calculations. An energy absorption profile in the target-calorimeter assembly is calculated by the electron-photon Monte Carlo method^{14,15} using the experimentally determined electron beam spectrum as the input.¹⁶ A typical absorption profile calculated for the UO_2 vapor experiments is shown in Fig. 3. The ratio of energy in the vapor sample to energy measured in the calorimeter

Fig. 3. Calculations of the electron energy absorbed on the target axis as a function of mass depth X for one vapor sample configuration used in these tests. The position of the UO_2 layer between layers of graphite is indicated by the curve discontinuities. The point at approximately $.4 \text{ g/cm}^2$ depth is the point corresponding to the optical pyrometer measurement.

plate is taken from this figure to deduce the energy in the vaporization sample for each experiment.

The powdered sample material contains some free volume in the form of voids. Once vaporization temperatures are reached, a small mass fraction of the liquid is converted to vapor, filling these void regions between particles. The internal energy required to produce this vapor is negligible, however, compared to the total absorbed energy. Thus, the energy absorbed from the electron beam can be directly identified as the internal energy on the liquid phase boundary, relative to the internal energy of the sample prior to heating. This energy state and the vapor pressure corresponding to that state are the quantities determined by the measurement method.

III. URANIUM DIOXIDE RESULTS

Initial experiments were carried out with powdered uranium dioxide samples ($\sim 2 \mu$ diam. powder) having an oxygen to metal ratio of 2.08. Expansion profiles from five electron beam tests are shown in Fig. 4. In each case these profiles correspond to the displacement of a $.092 \text{ g/cm}^2$ graphite plate. The displacement in this figure has been expressed in a reduced form by multiplying the measured graphite plate displacement (cm) by the graphite plate mass per area. The peak energy levels (J/g) in the UO_2 sample layers are indicated for each of these tests.

Values for the vapor pressure are obtained through numerical differentiation of the displacement profiles. The results of this differentiation for the five tests are shown in Fig. 5. There is some scatter associated with the velocity values determined by this means due to the finite resolution of the displacement photos. To indicate the resolution available in the current experiment, numerical velocity data points corresponding to the highest and lowest velocity tests are shown. The solid lines are linear fits to these points. In each case the linear curves give an accurate representation of the velocity points to within the data scatter and indicate no large negative curvature of the velocity data with time. Such a negative curvature would indicate a drop in vapor pressure during the observed expansion.

The offset of these curves from the origin at $t = 0$ is similar to the behavior previously reported for gold vaporization.¹⁰ There, the offset was found due to transfer of momentum from the condensed phase sample material. Possible contributions may also be present from small volume fractions of material having higher volatility than the UO_2 (adsorbed gas, free oxygen, etc.).

Fig. 4. Values measured for displacement as a function of time for five expansion experiments. The indicated specific energy level for each experiment corresponds to the peak value in the mixed phase sample layer. The values of displacement have been scaled by multiplying each with the value of mass per area of that plate (.092 g/cm²).

Fig. 5. Numerical differentiation of the information in Fig. 4 results in this representation of velocity as a function of time. The specific energy levels corresponding to each experiment are shown for each curve. To illustrate the current resolution of the method, the numerically determined velocity values are shown for the upper and lower trace. Linear fits to these points are indicated by the solid lines. The slope of each of these curves is then proportional to the sample vapor pressure.

The vapor pressure which can be identified with each of the linear fits in Fig. 5 is proportional to the slope of these curves and the mass per area of the graphite plate inertially confining the vapor. In this way, values of vapor pressure corresponding to the absorbed energy are obtained. Pressure versus energy (measured relative to energy at room temperature) data from these experiments are shown in Fig. 6. Values of pressure and energy corresponding to the figure are also given in Table I. The curve drawn through the experimental points is from a representation that will be discussed in section VI of this report.

Fig. 6. A representation of the vapor pressure as a function of internal energy from the five experiments displayed in Figs. 4 and 5. The value E_0 is the initial energy state of the sample at room temperature conditions. Bars on each of these points illustrate the uncertainty in the measurement of energy. The solid line is a model which assumes the slope of the logarithm of pressure versus $1/(E - E_0)$ is constant. This model is discussed in detail in Section VI.

TABLE I. Experimentally determined vapor pressures for UO_2 as a function of absorbed specific energy. In each case the initial energy state was that at room temperature.

TEST #	PRESSURE (bar)	ENERGY (J/g)	(cal/g)
7142	66	1990	477
7141	96	2030	485
7140	43	1860	445
7134	1500	3030	725
7133	322	2490	595

IV. COMPARISON OF DATA WITH MODELS

Numerous theoretical high temperature equations of state have been formulated and used for safety analysis. By comparing the electron beam data to those models, one can begin to establish the limitations of a given model. A few of the models which have been proposed for the relationship of vapor pressure and temperature are shown in Fig. 7. The simplest comparison of the electron-beam data to these models can be made by choosing an average heat capacity to convert the internal energy scale to temperature. This was done using the liquid C_p value in the ANL-Menzies equation of state²¹ of .44 J/gK to give the points from the electron beam data shown on Fig. 7. Some justification for the choice of this average value will be presented later in the report. Because of the approximate nature of this heat capacity, the points on Fig. 7 are approximate and derived from the data which is in a pressure-energy format. This temperature diagram shows an approximate relation to the vapor models.

Also shown in Fig. 7 are the range of temperature and pressure values from the laser heating experiments of Bober.⁸ Additional laser experiments have been reported recently by Ohse.⁷ Those data approximately coincide with the Booth EOS curve on Fig. 7 over the pressure range from 30 to 60 atm.

Among the models in Fig. 7, two will be chosen for additional calculations in this paper. The first is due to Menzies and has frequently been referred to as the ANL equation of state (curve 5). It is described by

$$P(\text{bar}) = 10^{-6} \exp[69.979 - 76800/T - 4.34 \ln T] \quad (1)$$

for T in degrees K

Fig. 7. A representation of vapor pressure versus reciprocal temperature. The electron beam heating results have been placed on this figure through the use of a constant heat capacity, $C_p = .44 \text{ J/gK}$ for the liquid UO_2 . Six of the vapor pressure models have been discussed in reference 1. The Booth model is from Ref. 23 while the experimental results of Bober are described in Ref. 8. Recent data of Ohse⁷ (not shown in this figure) compare well with the booth curve between 30 and 65 atm.

A second equation which matches a constant heat of vaporization extrapolation of Ackermann's^{17,1} high temperature effusion experiments (curve #1) is given by

$$P(\text{bar}) = 10^{-6} \exp(37.63 - 82880/T(\text{K})) \quad (2)$$

Placing the above temperature dependent equations in an energy format requires a model of the heat capacity. This heat capacity can then be used to relate the energy and temperature scales. The relationship between the constant pressure heat capacity C_p (for the liquid phase material) and the variation of internal energy along the coexistence curve may be found using the thermodynamic relation,

$$\left(\frac{\partial E}{\partial T}\right)_{\text{SAT}} = C_V \left(1 + \beta\gamma T\right) \left(1 - \frac{P}{T \left(\frac{\partial P}{\partial T}\right)_V}\right) \quad (3)$$

Here, β , the volumetric thermal expansion coefficient, γ , the Gruneisen parameter and other state variables refer to the liquid phase. In the liquid phase along the saturation curve, the term $\frac{P}{T \left(\frac{\partial P}{\partial T}\right)_V}$ becomes vanishingly small except for the immediate critical region. For this approximation the remaining terms on the right hand side of Eq. 3 are equal to C_p . Hence $\left(\frac{\partial E}{\partial T}\right)_{\text{SAT}}$ may be approximated as

$$\left(\frac{\partial E}{\partial T}\right)_{\text{SAT}} \doteq C_{P_{\text{liq}}} \quad (4)$$

where $C_{P_{\text{liq}}}$ is the specific heat of the liquid phase at the pressure and

temperature on the saturation curve. In this paper, the energy and volume states on the liquid saturation curve will be defined to be those at the intersection of the coexistence curve with the liquid state surface. Preliminary analysis relating the pressure-temperature data to the electron beam pressure-energy data makes the assumption that the heat capacity, C_p , does not vary with temperature and pressure along the region of interest on the coexistence curve. No temperature dependent measurements of C_p have been made at these high temperature levels. In fact, significant variations from this constant value may occur.

In these first comparisons, two heat capacity models will be used to place Eq. 1 and Eq. 2 in pressure-energy coordinates. The first model is based upon the Menzies-ANL equation of state with an average constant pressure heat capacity of .437 J/gK.^{21,1} The average in this case includes the heat of fusion energy. A second model uses measured values with an average value of C_p of .33 J/gK below melt and .50 J/gK above melt with a 276 J/g heat of fusion.

These two heat capacity models have been used to place each of Eqs. 1 and 2 in Fig. 8. The four curves obtained in this way can be compared with the electron beam data. For an energy of 2000 J/g the models in Fig. 8 span the range from 20 to 750 bars. The expressions in Eqs. 1 and 2 appear to bound the vaporization experiment when used with the above heat capacities. The pressure-temperature data from the laser heating experiments of Bober⁸ and of Ohse⁷ are also shown on this figure, each using the two heat capacity models.

Since the vapor pressure results of Ohse are measured as a function of temperature, while the present studies are measured in terms of energy,

Fig. 8. A comparison of the electron beam data to models of the equation of state based upon equations 1 and 2. Each of these equations are combined with the two heat capacity forms discussed in the test to give four pressure predictions as a function of the sample internal energy. Also shown are comparisons with the experimental results of Bober⁸ and Ohse.⁷ Those experimental pressure versus temperature data have been placed in energy coordinates using the same two heat capacity models.

the comparison could be used to estimate an average heat capacity. However, uncertainties which exist in the measurement of energy, vapor pressure and temperature limit the accuracy of this UO_2 heat capacity estimate. The average heat capacity of either model discussed above does give adequate comparison between Ohse's experiment and the present work.

V. HEAT CAPACITY DISCUSSION

It is important to recognize the significance of the heat capacity in describing the pressure-energy states on the equation of state surface from models of the P-T behavior. The heat capacity should be based upon experimental values determined under conditions which match those of the problem being analyzed (similar temperatures and heating rates, etc.). In this section of the report we will examine the data available to support such models, and then illustrate the effect of heat capacity variations on the pressure-energy representation of mixed phase states. Such a representation is needed to compare other vapor data with the present measurements or to carry out high temperature transient calculations.

Essentially all of the UO_2 heat capacity measurements have been made using standard calorimetry methods involving long duration heating and cooling cycles. We will refer to these as static measurements of the heat capacity. An excellent summary and evaluation of these heat capacities has been published previously.²⁴ The heat capacities are derived from enthalpy measurements which fall in a band about 6 percent wide. Recommended values from that study will be used in this section. In addition, a single measurement of the liquid phase heat capacity up to 3500 K has been reported and will be used in this analysis.³³

Based upon this limited data, models of the heat capacity up to ≈ 8000 K have been proposed. Fig. 9 shows three heat capacity models. Model 3 is based upon statically measured values up to 3500 K. Above that point the heat capacity is assumed to be constant at a value of .5 J/gK. A heat of fusion energy of 276 J/g is assumed. The pronounced peak in this

Fig. 9. Comparison of three heat capacity models proposed for UO_2 . Model 3 is from the Kerrisk and Clifton work together with the high temperature measurement of Liebowitz. Model 2 is the original Menzies-ANL average value where the heat of fusion is included in the average. Finally, Model 1 is based upon the measured values of heat capacity but excludes the strong peak occurring below melt. Above melt this model coincides with Model 3.

heat capacity curve below the melting temperature has been attributed to an activation energy process leading to the formation of oxygen interstitial defects in the UO_2 lattice.²⁵

The interstitial defect argument is complicated by the fact that an abnormally large concentration of defects is required to explain the magnitude of this peak. Therefore, diffusion and possibly segregation of oxygen resulting from oxide dissociation may also occur. Simple calculations suggest that a few to hundreds of milliseconds may be required for this diffusion. Hence the heat capacity peak would not be present using such a model for the electron beam experiments or for the disassembly phase of an HCDA accident, both of which occur on a faster time scale. Because of this possible rate effect, Model 1 which excludes this peak has been selected to represent one choice of the UO_2 heat capacity. Model 1 is based upon an extrapolation of the heat capacity measurements taken below 1500°C . The heat capacity in Model 1 has an average value of .33 J/gk below melt, a value of .50 J/gk above melt and a 276 J/g heat of fusion.

Finally, Model 2 is the heat capacity form of the Menzies-ANL equation of state. It assumes a single average liquid phase heat capacity over the entire range. The energy associated with the heat of fusion has been included in the heat capacity average for this model only.

Based upon these three heat capacity models and the Menzies-ANL P-T equation (Eq. 1), three pressure energy representations of the coexistence curve have been generated and are shown in Fig. 10. The measured values from the present experiments are shown from comparison. Reasonably close agreement with the present experiments is achieved with Models 1 and 2. The remaining deviation in those cases could result from vapor pressures which

Fig. 10. A comparison of the electron beam data to models of the dynamic equation of state. The formulations shown are based upon Menzies vapor pressure curve and the three heat capacity models in Fig. 9. The different heat capacity assumptions strongly affect the position of the liquid-vapor saturation curve in pressure versus energy coordinates.

differ from those predicted by the Menzies model for a given temperature. Producing a match between the present data and the static heat capacity values of Model 3 would require adjusting the P-T equation upward by $1\frac{1}{2}$ orders of magnitude. Such a large change would appear to be inconsistent with the experiments of Ohse or with the extrapolated equation of state.

If one chooses the P-T equation implicit in the Booth formulation (these values are consistent with the P-T measurement result of Ohse) and the same three models of heat capacity, the comparison shown in Fig. 11 results. Models 1 or 2 are consistent with the present data while Model 3 again produces unreasonably low values for vapor pressure.

If the differences in energy scale between model 3 (which is based on static heat capacity measurements) and the present experiments were due to an error in energy measurement in the present work, it would require a 30 percent error of energy measurement. This is inconsistent with our previous experience with the dosimetry measurements.¹³

At this time, two physical explanations have been proposed for the apparent disagreement in energy scale between the present work and enthalpy values from static measurements. First, that the present measurements carried out with hyperstoichiometric ($\text{UO}_{2.08}$) material have more than an order of magnitude larger vapor pressure compared to stoichiometric material. However this is inconsistent with the thermodynamic calculations of Breitung³² which show little dependence of the equilibrium vapor pressure on oxygen stoichiometry for pressures of the order of 100 atm. Second, a rate effect such as the one discussed in connection with the Model 1 heat capacity may result in different values for the static and dynamic experiments. The choice between these two explanations will require additional dynamic experiments on stoichiometric UO_2 .

Fig. 11. A comparison of the electron beam data with models of the dynamic equation of state. The figure is like that of Fig. 10 except that the pressure versus temperature coordinates are based upon the Booth formulation.

VI. AN ENERGY ORIENTED LIQUID-VAPOR EQUATION OF STATE

In the previous section of this paper, a strong dependence on heat capacity and heat of vaporization was shown for the description of a pressure-energy representation of the equation of state. A representation in energy coordinates is appropriate to transient problem analysis where the energy is the working state variable rather than an explicit representation of the temperature. In this section of the report, the concept of an energy oriented equation of state will be developed that permits a direct characterization of the mixed phase pressure states in terms of energy and density. While this form of the state surface does not contain complete thermodynamic information, it does contain all the information required for transient thermomechanical analysis.

Any complete theoretical equation of state formulation, since it contains full thermodynamic information can be placed in a $P(E,V)$ format. However, a complete theoretical model is difficult to support or even estimate in all its detail with experiments. By working with a $P(E,V)$ equation, only the data essential to adiabatic thermomechanical analysis is used. By including estimates of the heat capacity, perturbations from adiabatic conditions can be described as well. The essential characteristics of mixed phase thermodynamics required for transient thermomechanical problems will be discussed in this section of the report and expressed in terms of a few measurable quantities.

For the reactor safety analysis of an LMFBR core disruptive accident, the mixed phase volume state accessible to the fuel material generally lies within a factor of 10 of the fuel boiling point density. Within this area of the state surface one can accurately express the vapor pressure in terms

of the liquid saturation curve properties. The method we will present is an expansion of the state surface analogous to the condensed phase Mie-Gruneisen equation. In the Mie-Gruneisen case, the equation of state is expanded from the $T = 0$ compression curve using a parametric representation of the energy dependent term. By comparison, the present work will expand the state surface into the mixed phase region using the liquid saturation curve properties as the point of reference. The result is a representation of a large surface area on the state surface in terms of measurable saturation curve pressure and energy coordinates on the liquid saturation curve. Values for the heat capacities and saturation curve densities must also be estimated. For many problems such as the voided core reactor analysis, the sensitivity of the analysis to these values is not strong.

In a transient thermomechanical problem, the quantities E and V can be specified from the initial geometry and equilibrium temperature conditions. These are then updated to the current values as energy is added to the system from sources such as radiation, conduction, or internal heating. Frequently the energy is added rapidly to the system resulting in constant volume heating giving the specific energy and volume coordinates $(E_0 + E_{IN}, V_0)$. Where E_0 is based upon the initial equilibrium state, and E_{IN} is the energy added to the system. For analysis situations where incremental volume changes occur during heating, the energy input may be regarded as a sequence of short intervals. Then the energy and volume in each short interval may be corrected using the current system pressure to give the state $(E_0 + E_{IN} - PdV, V_0 + dV)$. The system is then followed through the sequence of short intervals to give the energy and volume state at any point in time. In such a way, the specification of energy and volume states is straightforward. A description of the mixed

phase pressure in terms of E and V needed for the sequence above will now be presented.

Expanding the two-phase states from the liquid saturation curve (in this paper we refer to the completely liquid phase end of the coexistence curve as the liquid saturation curve), we use:

$$P(E,V) = P(E,V_{SAT}) + \int_{V_{SAT}}^V \left(\frac{\partial P}{\partial V} \right)_E dV \quad (5)$$

where the term $\left(\frac{\partial P}{\partial V} \right)_E$ can be evaluated as follows. One can show for equilibrium mixed phase expansion, first at constant system entropy

$$\left(\frac{\partial P}{\partial V} \right)_S = - \frac{T}{C_V} \left(\frac{dP}{dT} \right)^2 \quad (6)$$

where (dP/dT) is the slope of the vapor pressure curve. Also at constant energy

$$\left(\frac{\partial P}{\partial V} \right)_E = - \frac{1}{C_V} \left(\frac{dP}{dT} \right) \left[\left(\frac{dP}{dT} \right) T - P \right] \quad (7)$$

The above equations are derived rigorously for the equilibrium two phase equation of state. However, some approximations are useful in evaluating the integral expression to permit rapid calculations. For modest expansion ratios (10:1) and for pressures well below the critical pressure, $P(E,V)$ is dominated by the value of $P(E,V_{SAT})$ so that small errors in the integral evaluation are not significant. Thus some approximations are useful in evaluating the integral expression to permit rapid calculations. The above expression for $(\partial P/\partial V)_E$ will now be simplified and expressed in terms of energy rather than temperature.

First, we assume that the mixed-phase heat capacity, C_V , is volume independent. If states sufficiently close to the liquid end of the saturation curve are considered, most material mass remains in the liquid phase thus causing the heat capacity to remain constant at the liquid density value. For example, a factor of 10 mixed phase adiabatic expansion from a subcritical point on the saturation curve will satisfy the above requirement.

The term $\frac{dP}{dT}$ in the mixed phase region may be simplified by relating energy in the mixed phase region to energy on the saturation curve. As discussed previously,¹⁰ it was found that,

$$\left(\frac{\partial E}{\partial T}\right)_{SAT} = C_V + \left[T \frac{dP}{dT} - P\right] \left(\frac{\partial V_{SAT}}{\partial T}\right) \doteq C_V \quad (8)$$

If we use the approximation at the righthand side of this equation, we then find the energy explicit form for dP/dT ,

$$\frac{dP}{dT} = \left(\frac{\partial P}{\partial E}\right)_{SAT} \left(\frac{\partial E}{\partial T}\right)_{SAT} \doteq \left(\frac{\partial P}{\partial E}\right)_{SAT} C_V \quad (9)$$

Using the result from equation 4, we recall that this value of mixed phase heat capacity, C_V , can be predicted from measured values of the liquid phase heat capacity, $C_{P \text{ liq}}$, since $C_V \doteq C_{P \text{ liq}}$.

Substituting this into the constant energy derivative expression gives the result,

$$\left(\frac{\partial P}{\partial V}\right)_E = -\left(\frac{\partial P}{\partial E}\right)_{SAT} \left[\left(\frac{\partial P}{\partial E}\right)_{SAT} C_V T - P \right] \quad (10)$$

A representation of the variation of heat capacity with temperature is needed to precisely evaluate the $C_V T$ term in energy coordinates. For high temperatures, however, a constant heat capacity is generally assumed

for lack of better information. In the spirit of that assumption, we replace $C_V T$ by $(E - E_F)$ where E is the total energy excursion from $T = 0$ and E_F the heat of fusion. This gives a simple energy explicit representation

$$\left(\frac{\partial P}{\partial V}\right)_E = -\left(\frac{\partial P}{\partial E}\right)_{\text{SAT}} \left[\left(\frac{\partial P}{\partial E}\right)_{\text{SAT}} (E - E_F) - P \right] . \quad (11)$$

Notice that in using the constant heat capacity assumption, one need not specify the constant value. If one can measure (as in section 2), or predict from heat capacity and vapor pressure versus temperature measurements, the value of P versus E on the saturation curve, then the above derivative and integral expression may be evaluated.

The mixed phase state surface for UO_2 will now be constructed with the use of equation (11) and the electron beam data discussed previously. The data will be used to define values for $(\partial P / \partial E)_{\text{SAT}}$.

Figure 12 shows the data from Section 2 plotted in a $\ln P$ versus $1/E$ representation. A linear fit of those points is shown which adequately describes the measurements. The linear fit is $\ln P = A - B/E$ where $A = 13.290$ and $B = 18110$ for P in bars and E in J/g , or $A = 27.106$ and $B = 1.8110 \times 10^{11}$ in CGS units.

Then,

$$P_{\text{SAT}} = e^{A - B/E} \quad (12)$$

$$\left(\frac{\partial P}{\partial V}\right)_{\text{SAT}} = \frac{PB}{E^2} \quad (13)$$

Substituting these into the above equations while neglecting the heat of fusion and energy excursion from $T = 0$ to room temperature in the derivative term, gives

Fig. 12. Vapor pressure versus reciprocal energy diagram. The energy in this curve is measured relative to the energy of the material at room temperature, E_0 . The electron beam points and uncertainty of the energy coordinates measurement are shown by the bass. A linear fit to those points is shown by the straight line.

$$\left(\frac{\partial P}{\partial V}\right)_E = K(E)P^2 \quad \text{where } K(E) = \frac{B}{E^2} [1 - B/E] \quad . \quad (14)$$

Summarizing, one can construct any desired energy and density point on the mixed phase surface by selecting the point on the saturation curve which corresponds to the energy of interest using,

$$\ln P = A - B/E \quad (15)$$

and then integrating at constant energy from V_{SAT} to the volume of interest V using

$$P(E, V) = e^{A - B/E} + \int_{V_S}^V \left(\frac{\partial P}{\partial V}\right)_E dV \quad (16)$$

with $\left(\frac{\partial P}{\partial V}\right)_E$ defined by Equation 14. Because of the slow variation of P with density, calculations of the volume dependence can be conveniently iterated from V_S to V using,

$$P(E, V + \Delta V) = P(E, V) + \left(\frac{\partial P}{\partial V}\right)_E \Delta V \quad (17)$$

where $\left(\frac{\partial P}{\partial V}\right)_E$ is evaluated using the pressure from the previous iteration in Equation 14. Generally in mixed phase problems, variations of the liquid phase density are negligible when compared to changes of the mixed phase density. To include this small effect however, information for the energy dependence of V_{SAT} can be estimated from thermal expansion and heat capacity data. Then, one could specify a linear dependence on energy using,

$$V_{SAT} = V_0 + \beta \cdot E \quad (18)$$

where V_0 and β are selected constants. To illustrate the state surface

which results from the calculation described above, a P - V mixed phase surface was generated with the linear fit of Fig. 12 and using $V_{SAT} = .2 \text{ cm}^3/\text{g}$. Constant energy traces of the PV surface are shown in Fig. 13.

In situations where limited excursions into the single phase liquid region may occur, a test to see if $V < V_{SAT}$ should be made. If this is satisfied, then the pressure should be computed using

$$P(E,V) = P(E,V_{SAT}) + B_M \cdot (V_{SAT} - V) \quad (19)$$

where B_M is an input value of the liquid phase bulk modulus. This crude approximation of the liquid phase surface is generally all that is required for problems dominated by the mixed phase EOS surface.

If one wishes to carry out a reactor safety calculation involving hydrodynamic disassembly, the only other temperature dependent quantity which appears in the analysis is the Doppler coefficient of reactivity. This coefficient has been described in the past most commonly using the parameter $\alpha(T)$ of the form $\alpha(T) = T \frac{dk}{dT}$ where k is the neutron multiplication factor. For a constant heat capacity (this has generally been assumed for the high temperature region of the state surface) $\alpha(T) = \alpha(E) = E \frac{dk}{dE}$. A better estimate of the $T(E)$ relation may be included, of course, in place of this constant heat capacity form if such information is available.

The result of the discussion in this section has been to place the equation of state for transient thermomechanical analysis in energy and density coordinates. The role of the temperature variable is not explicit during the transient calculation. The initial or equilibrium temperature is used, however, to define the initial energy state, E_0 , from which the

Fig. 13. Constant energy traces of the mixed liquid vapor state surface. The model discussed in the text and the electron beam data were used to generate these pressure-volume points for the indicated mixed phase energy levels.

thermal transient calculation beings. The resulting thermodynamic state surface may be described in terms of a few measurable quantities without detailed consideration of the high temperature physics and chemistry involved. As such, it represents a useful experimental EOS description for calculational purposes.

In contrast to the simple thermodynamic EOS model just discussed, other equations of state based upon simple theoretical descriptions of the vapor have been proposed. For example, models have been discussed using the corresponding states approach,^{21,23} significant liquid structures,²⁷ as well as theories based upon interatomic potential constructions of the equation of state surface.²⁸ While these give considerable insight to high temperature vapor states, such models generally omit or only approximately model physical processes such as ionization or chemical reactions which may be quite important. The experimentally oriented mixed phase EOS is designed to minimize such problems. One thermodynamically complete formulation used widely at Sandia is the CHARTD or ANEOS equation of state.²⁸ Obviously this complete representation describes the $P(E, V)$ characteristics in the mixed phase region. A recent fit of the ANEOS model²⁰ to UO_2 has been used to define the $P_{SAT}(E)$ curve shown in Fig. 14. The coordinates of the liquid saturation curve are compared with the previously described experiments. Table II contains the values of parameters used for this first try at matching the EOS properties. Because of the simple approximations for the heat capacity and compressive properties used, the freedom in varying the P-E curve is limited. Typically one must sacrifice the modeling of some thermodynamic properties to match others. The parameter values in Table II represent one possible

Fig. 14. A description of the pressure vs energy coordinates of the saturation curve based upon the ANEOS32 equation of state model. An initial fit to the UO_2 properties using the parameter values of Table II is shown. The electron beam points are also shown for comparison.

Table II. Values of the equation of state parameters used for representing UO_2 in the ANEOS Equation of State. Discussions of the units and meaning of these parameters are given in Ref. 32 where the designation ZB(I) is used to describe the equation of state input array.

I	ZB(T)	I	ZB(I)	I	ZB(I)
1	2.0	9	0	17	0
2	3.0	10	2.0	18	0
3	10.968	11	5.1×10^{10}	19	0
4	.02568	12	.268	20	0
5	0	13	8.8×10^{12}	21	0
6	2.0×10^{12}	14	.855	22	0
7	2.10	15	0	23	2.82×10^9
8	.025	16	0	24	.980

compromise in matching this thermal data. Improving agreement with the data may require adding chemical and ionization processes which are currently not in the model. Until this can be done in detail, a thermodynamic representation of the state surface based upon experiments should prove useful.

VII. VAPORIZATION IN TRANSIENT REACTOR ANALYSIS

The high temperature vapor equation of state for UO_2 is currently used as an input to calculations designed to assess the consequences and related safety of LMFBR reactor systems under abnormal transient conditions. In such a sequence, the vapor equation of state becomes an undeniably reliable driving force for neutronic disassembly, thus assuring termination of the prompt power generation in the reactor. The goal of a safety calculation for this sequence is to define the mechanical work component of the energy release and the related structural loads on the containment system. Such calculations are made for a particular reactor design to absolutely bound the consequence of an accident condition.

Fig. 15 shows the calculative scheme which is being used in the analysis. The upper loop shows the skeletal features of the disassembly computation carried out by computer codes such as VENUS,²⁹ PAD,³⁰ KADIS,³¹ etc. Initial conditions hypothesized for a prompt critical disassembly are evolved in time by a neutronic calculation of the power developed in the current fuel geometry. This gives an energy density at each point in the reactor core. Then, using an equation of state surface, vapor pressure levels corresponding to each of these points may be input to a mechanical analysis. The predicted material motion then modifies the neutronic analysis and so on in a closed loop calculation. After termination of the prompt power generation, the EOS properties continue to be important during expansion. The expansion properties then result in values being defined for the mechanical energy release or work as well as impulsive loads upon the reactor containment features.

One may estimate the influence of the fuel vapor EOS on the expansion calculations for the vapor using the vapor relations described in this paper.

Fig. 15. A flow diagram of the analysis used to calculate the neutronic disassembly is an HCDA sequence. The mechanical energy or work is calculated from the core expansion which occurs after the termination of the prompt neutron transient pulse. The equation of state contributes to both segments of the calculators and is particularly important in the calculation of the conversion of thermal to mechanical energy.

The major assumption is that contributions to the expansion work induced by interaction of the fuel with sodium coolant are negligible. Current theory and experiments suggest that fuel vapor is the important working fluid. In this section we will evaluate the expansion properties of a mixed liquid-vapor region characterized by average values of temperature, pressure, and specific volume T , P , V . The pressure dependence of an incremental volume change along a mixed phase adiabat can be described by,

$$P = P_0 + \left(\frac{\partial P}{\partial V} \right)_S \Delta V \quad (20)$$

where the derivative may be evaluated using the technique of the previous section. The mechanical energy generated by fuel adiabatic expansion may then be determined by integrating the expression,

$$W = \int_{V_1}^{V_2} P(V) dV \quad (21)$$

where $P(V)$ is determined by iteration of the above pressure volume curve for a given form of the equation of state. This adiabatic analysis neglects the heat transfer from the fuel which can substantially reduce the expansion energy. Thus W gives an upper bound on the mechanical energy release (commonly called work potential) for situations in which the coolant is not vaporized.

To illustrate the effect of the equation of state on the behavior of this work potential, the above integral was evaluated for initial temperatures (before expansion) between 5000 K and 9000 K using the equation of state forms described by Eqs. 1 and 2. The heat capacity assumed for the analysis was the constant value in the ANL-Menzies model. Expansion energy was

calculated for an average density ranging from an initial 5 g/cm^3 to a final value of 2.5 g/cm^3 . The precise density values of interest to a reactor calculation are those which range from the initial average fuel density to the fuel density at the time the sodium slug impacts on the reactor vessel containment. As stated previously, the thermal interaction of the fuel with its surroundings is assumed to be negligible.

The mechanical energy calculated as a function of initial temperature for two equations of state is shown in Fig. 16. For comparison, the temperature values are related to an energy scale as well. This energy scale has been defined as the energy required to reach the indicated temperature when starting from an initial temperature of 1000 K.

Two conclusions are evident from the calculations in Fig. 16. First, the work potential values from these selected equations of state differ by more than an order of magnitude for a given temperature prior to expansion. Second, if the EOS is fixed, variation of the initial energy state on disassembly by 20 percent will result in a work potential variation of a factor of 3. Thus in considering a disassembly analysis, the work release is demonstrated to be sensitive to the EOS parameters as well as to the value of total energy release on disassembly.

Fig. 16. Calculated values of the work potentially available to act on the reactor containment as a function of fuel temperature prior to expansion. Equations 1 and 2 in the test have been used to represent the pressure-temperature behavior of the fuel. For comparison, the temperature scale has been related to thermal energy on the lower scale using the Menzies-ANL heat capacity model (Model 2 of Fig. 9). The low efficiency of thermal to mechanical energy is shown for this twofold volumetric expansion example.

VIII. SUMMARY

The details of an initial series of UO_2 vaporization experiments have been discussed. A recently developed method was used for this work which gives the first vapor pressure versus internal energy data for UO_2 . Vapor pressure data which span an energy range from 1900 J/g to 3000 J/g above the ambient internal energy state have been measured. Corresponding vapor pressures range from tens of bars to 1.5 kbar.

These data have been compared with a variety of models for the UO_2 vapor. Particularly important in such a comparison is the correct choice of the high temperature heat capacity. While good agreement can be obtained with the present data using dynamic models of the heat capacity, comparisons with lower temperature statically measured heat capacity data are unfavorable. This result suggests that high temperature UO_2 behavior may be more complex than previously assumed.

Because of these potentially complex aspects in modeling UO_2 properties, a thermodynamic representation of the state surface without detailed assumptions has been presented. This representation allows one to carry out a thermomechanical analysis based entirely on vapor data from transient experiments and a few non-restrictive assumptions. An experimentally oriented equation of state should allow adequate analyses until a detailed understanding of the high temperature processes in UO_2 is attained.

Finally, the significance of these state surface assumptions in a transient analysis has been examined by exploring their effect of the mechanical energy release in a liquid-vapor expansion. This is similar to the process

used to determine work potential in a reactor transient analysis. A significant EOS dependence to the work potential is demonstrated. Strong sensitivity of the work term to the initial temperature or total energy state prior to expansion is also observed.

Further study of the uranium dioxide equation of state is needed to understand the vaporization and chemical processes involved. Additional electron beam, as well as pulsed reactor experiments have been planned to address this problem. Additional data for the adiabatic release profile as well as heat transfer and other rate effect should become available from this effort and contribute to development of specific physical models.

In addition to this UO_2 work, experiments are needed to obtain data for more prototypic fuel materials to meet reactor analysis requirements. The nuclear fuels of interest in such analysis have significant concentrations of volatile fission products. Under some circumstances the vapor pressure behavior of the stainless cladding adjacent to the fuel can be important. Each of these areas require experiments before a fuel vapor analysis can be considered complete. The present studies represent the first part of the experimental input needed for such reactor analysis.

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