

FISSION-GAS-BUBBLE MOBILITY IN OXIDE FUEL -
A CRITICAL ANALYSIS

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The issues of fission-gas release from UO_2 fuel rod is an integral part of light-water (LWR) reactor safety analysis. Recent investigations¹ have indicated that the release of fission gases from UO_2 fuels have a close connection to the release processes of the other volatile fission products (VFP) (e.g., Cs, I, Te, etc.). In fact, according to the scaling model of volatile fission product release² the fractional release rates of the VFPs (μ_{VFP}) scale as that of the fission gases, μ_{FG} . In other words one has

$$\mu_{VFP} = \alpha_{VFP} \mu_{FG} \quad (1)$$

Here α_{VFP} is the scaling factor for that particular VFP as determined by the fuel chemistry. In this way a simple recipe has been provided within a potentially complex situation to give reliable estimates of the release rates of the VFP and their associated compounds. Broad agreement with the available VFP release data has confirmed the general viability of this approach.¹

The structure of the basic equation of the scaling model (Eq. 1) implies that its usefulness is related closely to one's ability to obtain reliable estimates of the scaling factor α_{VFP} and the fission gas fractional release rate μ_{FG} . To estimate α_{VFP} depends on a clear understanding of the fuel chemistry. Its analysis has been reported recently.^{1,2} The other important factor is of course μ_{FG} . Despite well over a decade of intensive theoretical and experimental research of fission gas behavior in nuclear fuel several

issues still remain open and controversial. Foremost among these is the problem of fission gas bubble mobility. Most workers in the field agree that gas bubbles do move (except for some uncertainty for very small bubbles of diameter below about 20 \AA).³ The major controversy is whether the gas bubbles can move sufficiently fast to be a significant mechanism responsible for fission gas release from the fuel.

For temperature regime below about $1800\text{-}1900^\circ\text{C}$ there exist substantial amounts of steady state^{3,4} (i.e., with the bubble in presumably mechanical equilibrium with the surrounding matrix) mobility (μ_B) data. For bubble sizes below about $1 \text{ }\mu\text{m}$ μ_B is generally below that predicted by a conventional surface diffusion mechanism.^{5,6} This discrepancy in general increases as the bubble sizes decreases.⁶

A recent model that seeks to rationalize this discrepancy invokes the concept of non-localized surface diffusion.⁷ Here the migrating surface UO_2 molecule is supposed to execute a long range gas-like translational motion over a narrow region above the bubble-matrix interface. The interference by the fission gases within the bubble on this gas-like migration serves to suppress severely the efficiency of this mass transfer process. As a result within this and another similar model⁸ the gas bubble mobility is significantly lower than what it would otherwise have been without consideration of the fission gas effect. This is consistent with the available data on μ_B .^{6,7,8}

A central parameter of this particular mechanism that has not been addressed is the mean jump distance per jump (R_M) of this hopping process. We have analyzed this parameter from the point of view of a variable range hopping mechanism. The jump distance per jump is now no longer a fixed quantity but a random variable itself. Its average value R_M is determined by the coupling of the surface to the excited internal degree of freedom of the hopping entity

(e.g., an UO_2 admolecule). Among the rotational, vibrational and electronic internal degree of freedom of an admolecule the first one is probably the most important in determining R_M . Using the technique of dissipative trajectory similar to that used in calculation of surface sticking coefficient⁹ the mean jump distance per jump has been estimated within a model in which successive interaction due to the surface/internal degree of freedom coupling is taken into account in a simple manner. This scheme also allows one to incorporate easily the influence of external perturbations (e.g., collision with fission gas atoms and surface imperfections such as steps as well as the effect of the bubble finite sizes). The relative importance of their impact depend crucially on the ratio of the mean jump distance to the relevant length scales (such as the mean free path of the fission gas atoms, mean step separation, or the bubble radius). In general these perturbations act to reduce the mean jump distance and hence the bubble diffusivity.

The picture of a migrating entity executing variable range hopping thus leads to behavior of the bubble mobility that is consistent with the experimental trends for $T \lesssim 1800\text{-}1900^\circ\text{C}$. In fact such long range hopping processes has also been observed in recent molecular dynamics simulation on surface diffusion phenomena.¹⁰

However application of these low temperatures (i.e., $T \lesssim 1800^\circ\text{C}$) bubble mobility results to fuel element modeling under transient heating condition results in very marginal amounts of fission gas release.¹¹ This is in contrast to the tens of percent of gas release prior to melting observed in a wide range of laboratory data.^{12,13} As the peak temperature reached in these transient heating experiments are in general significantly above 1800°C it is not obvious that the low temperature bubble mobility (i.e., $T \lesssim 1800^\circ\text{C}$) results are at all applicable.

A common feature of the low temperature models is that the fractional concentration of surface defects responsible for mass transport leading to bubble migration is low (i.e., $\ll 1$). In fact this is one reason why bubble mobility is rationalized to be small as indicated by the available data (for $T \lesssim 1800^\circ\text{C}$). Direct observations via TEM³ have indicated that the gas bubbles are often faceted and have an octahedron shape with the [111] faces truncated at the corners by 100 planes. The physical picture of a typical bubble/matrix interface is thus of a relatively smooth surface punctuated by a low concentration of surface defects and defect clusters including ad-molecules, surface vacancies, etc. However when the temperature increases so does thermal fluctuation leading to increases in surface defect concentration. When the temperature increases to a threshold temperature T_R thermal disorder is so great that complete surface roughening takes place.

Very recently a criterion has been proposed basing on renormalization group argument that relates the surface roughening temperature T_R to the interfacial tension $\Gamma(T)$ and the surface structure.¹⁴ Using the criterion together with a temperature-dependent $\Gamma(T)$ for UO_2 that is broadly consistent with experimental data from $T \approx 300^\circ\text{C}$ to about 1900°C ¹⁵ (including the value of 1000 erg/cm^2 at $T \approx 900^\circ\text{C}$ commonly adopted in fission gas release modeling¹⁵) one could obtain an estimate of T_R about $2500\text{-}2600^\circ\text{K}$.

Above that temperature the fractional defect concentration on the surface is of order unity. This translates into a significant increase in μ_B , the bubble mobility. An alternate viewpoint is that the surface disorder is so great that the interface region essentially is in a quasi-liquid state. Subsequently bubble mobility is governed by liquid-like surface diffusion in the interface region. Either way one would observe significant increases in bubble mobility.

In summary we have analyzed the recently suggested model of bubble mobility via nonlocalized surface diffusion⁷ from the point of view of a variable range hopping process. The physical mechanism underlying such a process was suggested to be due to surface/internal degree of freedom coupling. It was shown that within this approach the effect of external perturbation on the mean jump distance per jump R_M can be incorporated easily leading to attenuation of R_M . This is consistent with experimental trend^{3,4} ($\approx 1800^\circ\text{C}$) as well as previous analysis.^{7,8}

However we emphasize that this type of bubble migration mechanism need not be dominant over all temperature range. In fact at a significantly higher temperature regime than 1800°C other processes (e.g., surface roughening) could assume greater significance. As a result the bubble mobility at high temperature (e.g., $\approx 2300^\circ\text{C}$) could operate via a qualitatively different mechanism from that at the lower temperature regime (e.g., $\approx 1800^\circ\text{C}$). Thus for transient processes that reach a peak temperature $\approx 2300^\circ\text{C}$ fission gas release could result from concurrent processes¹⁶ including that of gas bubble migration.

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