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VAPORIZATION BEHAVIOR OF NON-STOICHIOMETRIC REFRACTORY CARBIDE MATERIALS AND DIRECT OBSERVATIONS OF THE VAPOR PHASE USING LASER DIAGNOSTICS

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ABSTRACT. Transition metal and actinide carbides, such as ZrC or NbC and UC or ThC, exhibit a wide range of stoichiometry. As a consequence, these materials vaporize incongruently. At relatively long times steady state vaporization can be achieved where the relative concentrations of atomic species on the solid surface equals that in the gas phase. The surface composition under these steady state conditions is termed the congruently vaporizing composition, or CVC. Modeling the vaporization or corrosion behavior of this dynamic process is complex and requires an understanding of how the surface composition changes with time and a knowledge of the CVC, which is both temperature and atmosphere dependent. This paper describes the vaporization and corrosion behavior of non-stoichiometric refractory carbide materials and, as an example, will describe a thermokinetic model that characterizes the vaporization behavior of the complex carbide $U_xZr_{1-x}C_y$ in hydrogen at 2500 to 3200 K. This model demonstrates that the steady state corrosion of $U_xZr_{1-x}C_y$ is rate limited by the gaseous transport of Zr where the partial pressure of Zr is determined by the CVC.

This paper also briefly describes our efforts to image and characterize the vapor phase above the surface of ZrC in static and flowing gas environments using planar laser induced fluorescence or PLIF. We have developed the method for the purpose of monitoring and controlling the corrosion behavior of nuclear fuels in nuclear thermal rockets. However, the techniques that are described can be used, for example, to image boundary layers, and could be used for the purpose of verifying corrosion models.

1. Introduction

The corrosion behavior of nuclear fuel materials in hot hydrogen is a critical issue for a variety of nuclear thermal propulsion (NTP) concepts for missions to Mars. In order to maximize the rocket performance certain reactor designs presume fuel operating temperatures in excess of 2800 K in a high velocity flow of high pressure, gaseous hydrogen. For this reason refractory carbide materials such as the $U_xZr_{1-x}C_y$ solid solution are candidates for such applications¹ due to their high melting points, good nuclear properties, and relatively good resistance to corrosion by hydrogen. Because the rate of corrosion can limit the reactor life, it is important to have a thorough understanding of the corrosion kinetics of nuclear fuel materials in hydrogen under a variety of possible operating conditions. The development of models which can account for changes in the many possible operating variables is therefore critical. Due to economic and environmental constraints, in any piloted Mars mission, modeling will inevitably become a major effort in predicting rocket performance and safety.

We need the ability to calculate vaporization/corrosion rates *a priori* so that extrapolations can be reasonably made from laboratory or test conditions to actual operating conditions. In this paper a model is presented that can account for variables such as temperature, pressure, gas flow rate, percent dissociated hydrogen, and geometry. In theory, the model presented here should also be able to account for impurities in the gas phase, such as hydrocarbons. The model assumes that the vaporization/corrosion behavior is controlled by gas transport. Thus, when corrosion is rate limited by surface reactions, such as cracking of hydrogen, the model is not applicable. As a general rule, gas/solid reactions remain or tend to become rate limited by gaseous transport as the temperature and total pressure are increased. Thus, the general model should be applicable to a real reactor environment where the pressures are significantly higher than 1 atm.

Additionally, it is important to be able to monitor the fuel corrosion products in the laboratory or in a reactor not only for understanding the corrosion kinetics, but also to assess the performance of NTP systems during operation. A space NTP system will require instrumentation that provides continuous monitoring of various operational parameters, such as gas temperature and density. Because the environment in the core of the rocket will be extremely harsh, many common diagnostic instruments (e.g., thermocouples) can not easily be used. Laser diagnostic methods provide some unique advantages over other diagnostic techniques. In particular, measurements can be done *in situ* with all the hardware located remotely. Laser diagnostics provide imaging ability, excellent temporal ($< 1 \mu\text{s}$) and spatial resolution (20-1000 μm), and in general are not affected by high temperatures. High temperatures can limit one's ability to resolve certain emission lines and, therefore, can potentially affect concentration measurements. It is thought that a laser diagnostic specific to a certain corrosion product can, when probing a rocket's exhaust, supply information on local gas stream concentrations and temperatures. This information could be used, for example, to adjust the reactor operating temperature or propellant flow rate, both of which influence the rate of corrosion.

In this paper we briefly describe a study initiated to develop, test, and subsequently use non-intrusive, laser-based diagnostics to characterize the gaseous products that evolve during the exposure of representative fuel samples to hydrogen. In the portion of the work described here, laser ablation is used to produce high temperature, vapor plumes from uranium-free ZrC in static and flowing gas environments. The plume is probed and imaged using planar laser induced fluorescence (PLIF).

2. The Thermodynamics of Incongruent Vaporization/Corrosion of Carbide Fuels

Uranium carbide and thorium carbide form complete, nearly ideal solid solutions with the refractory carbide compounds ZrC, NbC, HfC, and TaC. By incorporating, for example, UC into a solid solution with ZrC, a very high melting point nuclear fuel may be attained. Refractory carbide nuclear fuels are nonstoichiometric, meaning the compounds exist over a wide range of stoichiometry. To illustrate this, figures 1 and 2 show the binary phase diagrams for the systems U-C and Zr-C, respectively; and figure 3 shows an isothermal ternary phase diagram for the system U-Zr-C at 2473 K.^{2,3}

As will be discussed in a subsequent section, in order to model the vaporization behavior of refractory carbide fuels, it is necessary to know the equilibrium partial pressures of important vapor species. When a $\text{U}_x\text{Zr}_{1-x}\text{C}_y$ fuel is undergoing hydrogen corrosion, the most important vapor products are U, Zr, and hydrocarbons.^{3,4} These gaseous products

Figure 1. The U-C binary phase diagram. Figure 2. The Zr-C binary phase diagram.

Figure 3. The U-Zr-C isothermal phase diagram at 2473 K.

leave the solid surface at different rates due to the differences in equilibrium partial pressures and transport kinetics. Because this solid solution exhibits a wide range of stoichiometry, the fuel composition can change significantly during vaporization or corrosion. In particular, the equilibrium concentration of carbon vacancies is a strong function of temperature and atmosphere. Any change in composition will affect the equilibrium vapor pressures. For simplicity, if we consider the corrosion of the binary compound ZrC in 1 atmosphere of hydrogen at temperature T as follows:

the equilibrium partial pressures of Zr and C_2H_2 may be calculated from the equation:

$$p_{C_2H_2}^{x/2} \cdot p_{Zr} = \exp\left(\frac{\Delta G_{rxn}}{-RT}\right) \quad (2)$$

where ΔG_{rxn} is the Gibbs free energy of the reaction and RT has its usual meaning. Knowing the stoichiometry of ZrC_x at the surface we can calculate its free energy of formation using the relationship:

$$\Delta G_{ZrC_x} = RT \ln(a_{Zr}) + xRT \ln(a_C) \quad (3)$$

where a_{Zr} and a_C are the activities of Zr and C, respectively. Figures 4 and 5 show how the activities and partial pressures, respectively, of relevant species change with carbon to metal ratio at 2000, 2500, and 3000 K. As shown in figure 5, the partial pressures of Zr, C_2H_2 , and CH_4 change by two to ten orders of magnitude across the compositional range $ZrC_{0.6}$ to $ZrC_{0.99}$. Thus, as ZrC_x (or other transition metal or actinide carbide material) undergoes hydrogen corrosion the equilibrium partial pressures must change as the surface composition changes. Calculating or modeling the rate of change of the surface composition is a relatively complex topic deserving significant space; Thus, it will not be discussed in this brief paper, but the reader can find further details of the authors' work elsewhere.⁵

Figure 4. Composition dependence of the activities of zirconium and carbon in ZrC_x at 2000 to 3000 K. Data are based on the measurements of Storms.^{3,6}

Figure 5. Calculated equilibrium partial pressures above the surface of ZrC_x in 1 atm of hydrogen.

As the corrosion proceeds, the surface composition approaches what is referred to as the congruently vaporizing surface composition or CVC. The temperature dependence of the CVC of ZrC , and other refractory carbide materials of interest, in 1 atm of hydrogen is shown in figure 6. Therefore, at sufficiently long corrosion times the corrosion rate reaches a steady state condition where the relative concentration of atoms on the surface equals that in the gas phase just above the surface. It is this steady state condition that is of concern in this paper and will be discussed further below.

Figure 6. Congruently vaporizing surface compositions (CVCs) of selected refractory carbide materials in 1 atm of hydrogen.

3. Kinetics and Modeling of the Vaporization/Corrosion Behavior of Refractory Carbide Materials. Case Study: $U_xZr_{1-x}C_{1-y}$ in 1 atm of H_2 at 2500-3200 K

Under the assumption that gaseous diffusion is rate limiting, the corrosion rate of $U_xZr_{1-x}C_{1-y}$ may be described using a flux equation, analogous to Fick's first law of diffusion, as follows:

$$J_{UZrC} = J_i = -\frac{h_i \Delta p_i}{RT} \quad (1)$$

where J_{UZrC} is the molar flux of $U_xZr_{1-x}C_{1-y}$ away from the surface, J_i is the molar flux of the rate controlling gaseous product species, i , away from the surface, Δp_i is the change in the partial pressure of species i between the bulk gas stream and the CVC surface (see figure 7), and RT has its usual meaning. The average or effective mass transfer coefficient, h_i , may be determined from boundary layer theory and, for the particular bullet-shaped geometry⁷ used in these studies, was estimated to be:⁸

$$h_i = \frac{D_{ij}}{L} (0.70 Re^{1/2} Sc^{1/3}) \quad (2)$$

where L is the characteristic dimension of the specimen (bullet diameter), D_{ij} is the gaseous interdiffusion coefficient for species i through, in this case, hydrogen, Re is the Reynolds number, and Sc is the Schmidt coefficient. The gaseous interdiffusion coefficient may be expressed as follows:⁹

$$D_{ij} = \frac{3kT}{8P} \sqrt{\frac{kT}{2P} \left(\frac{m_i + m_j}{m_i m_j} \right)} \cdot \frac{1}{\sigma_{ij}^2 \Omega_D \left(\frac{kT}{\epsilon_{ij}} \right)} \quad (3)$$

where k is the Boltzmann's constant, P is total pressure, and m represents molecular mass. The terms σ and ϵ are Lennard-Jones potential parameters (collision diameter and

Figure 7. Temperature dependence of the CVC above U.05Zr.95C1.07 in 1 atm of hydrogen.

interaction energy parameter, respectively) and Ω_D is referred to as the Lennard-Jones potential. Values of or methods for calculating these parameters are well documented.^{10,11} The dimensionless terms R_e and S_c in (2) may be calculated from the following relationships:

$$R_e = \frac{Lv}{v} = \frac{\rho v L}{\eta} \quad (4)$$

and

$$S_c = \frac{v}{D_{i-mix}} = \frac{\eta}{\rho D_{i-mix}} \quad (5)$$

where v is the bulk gas stream velocity, ν is the kinematic gas viscosity, η is the dynamic viscosity, and ρ is the gas density. The monomolecular dynamic viscosities were calculated using the equation:¹¹

$$\eta_i = \frac{5}{16} \cdot \frac{\sqrt{\pi m k T}}{(\pi \sigma^2) \cdot \Omega_D(kT/\epsilon)} \quad (6)$$

In determining the gas viscosity at these very high temperatures it is important to consider both molecular and dissociated hydrogen since these species have different individual gas viscosities. In the case of the binary mixture H-H₂, the dynamic gas viscosity, η_{mix} , was calculated using the following mixture rule:⁹

$$\eta_{mix} = \frac{\sum_{i=1}^N \sqrt{M_i} y_i \eta_i}{\sum_{i=1}^N \sqrt{M_i} y_i} \quad (7)$$

where M is molecular weight and y is the volume fraction of gas species i . In these studies, the concentrations of H and H_2 were assumed to be near equilibrium since the time for both dissociation and recombination at these temperatures and pressures is on the order of 10 to 100 milliseconds.¹² Likewise, the complex gaseous interdiffusion coefficient may be calculated using the following mixture rule:^{9,13}

$$D_{i-mix} = (1 - y_i) \left[\sum_{j=1}^N \frac{y_j}{D_{ij}} \right]^{-1} \quad (8)$$

Equation (1) was used to calculate the corrosion rate of $U_xZr_{1-x}C_{1-y}$ in hydrogen at 1 atm between 2500 and 3200K. The values for Δp_i were obtained by calculating the partial pressures above the CVC using thermodynamic data from several sources.^{3,6,14-16} In a fast flowing gas stream $\Delta p_i \approx p_i$ where p_i is the equilibrium partial pressure of the rate limiting gaseous species. Thus, the values of Δp_i used in calculating the corrosion fluxes corresponded to the equilibrium partial pressures of the most stable gaseous products, for example, $U(g)$, $Zr(g)$, $C_2H_2(g)$, and $CH_4(g)$. Because it is well known that solid solutions of UC and ZrC deviate only slightly from Vegard's law, $U_xZr_{1-x}C_{1-y}$ was assumed to be an ideal solution of UC and ZrC_x.

4. Comparison of Calculated and Experimental Corrosion Rates and Extrapolations

Calculated corrosion rates were compared to hydrogen corrosion data from studies performed at Los Alamos National Laboratory which yielded the following Arrhenius relationship for $U_{0.05}Zr_{0.95}C_{1.07}$ in 1 atm of hydrogen at 2670-3190K at a Reynolds number of approximately 500:^{7,17}

$$\ln \text{rate}(\text{g}/\text{cm}^2 \cdot \text{sec}) = 10.891 - \frac{68618}{T} \quad (9)$$

Figure 8 shows a comparison of calculated versus experimental corrosion rates. The calculated data shown in figure 8 are based on two different assumptions: (1) $Zr(g)$ transport from the CVC was rate limiting, and (2) $C_2H_2(g)$ transport from the CVC was rate limiting. These two assumptions gave the best agreement between the model and the data. The relatively good agreement between the experimental data and assumption (1) above suggests that the corrosion of $U_xZr_{1-x}C_{1-y}$ in hydrogen may be rate limited or partially rate limited by gaseous transport of Zr atoms away from the surface.

As is apparent from equations (1) and (2), the corrosion flux is a strong function of the bulk gas stream velocity. Because h_j is proportional to the square root of the Reynolds number, one predicts from equation (1) that the corrosion flux increases approximately proportionally with the square root of gas velocity. By plotting the log of the corrosion flux versus the log of the gas velocity a slope of approximately 0.5 should be observed. Figure 9 shows the effect of gas velocity on the calculated and experimental corrosion rates at 2600 K in 1 atm of hydrogen. Although the slope of the experimental data is less than 0.5, there is a positive correlation between flux and velocity, and there is

Figure 8. Arrhenius plot of calculated and experimental steady-state corrosion rates above the CVC composition of $U_{0.05}Zr_{0.95}C_{1.07}$ in 1 atm of hydrogen.

Figure 9. Calculated and experimental steady-state corrosion rates plotted as a function of bulk gas stream velocity at 2600 K in 1 atm of hydrogen.

good agreement between the calculated and experimental values. The fact that the experimental corrosion flux was found to depend on velocity suggests that the underlying assumption of this model (*i.e.*, that the corrosion rate is controlled by gaseous diffusion) is valid since surface reaction rate limited processes are independent of gas velocity. Note that at very high gas velocities the model predicts that the corrosion rate exceeds the thermodynamic maximum (see figure 8) which is not possible. In reality the rate of corrosion can only approach the thermodynamic maximum asymptotically at very high gas velocities. Also, the mass transfer coefficient must be appropriate for the particular Reynold's number of interest and for the type of flow, *i.e.*, laminar or turbulent.

5. Laser Diagnostics of Vaporization Processes

In addition to the model described above, a number of other investigators have used boundary layer theory and principals of gas kinetics with some success to model the vaporization or corrosion of materials undergoing high temperature gaseous corrosion.¹⁸⁻²¹ However, to date, there have been no direct observations of the gaseous boundary layers to provide quantitative verification that the models are valid. Laser diagnostics are amenable to such verifications.

Much of the laser diagnostics work that we have done has been aimed at identifying, imaging, and measuring the temperature of specific species in laser ablated plasma plumes. In these experiments, a rotating disk of, typically, ZrC or NbC was ablated by an excimer laser into a low pressure (≈ 1 kPa) backing gas of either argon or helium (see figure 10). A second laser was then passed through the resultant plasma plume in the form of a laser sheet at a wavelength selected to excite a specific optical transition of the molecule of interest. The fluorescence image of the excited species was then captured using a CCD camera as illustrated in figure 11. This technique is referred to as planar laser induced fluorescence or PLIF. Many of the details of our ablation studies have been reported elsewhere and will not be discussed further here.²²⁻²⁵

Figure 10. Schematic illustration of the laser ablation/laser diagnostics facility.

Upon the development of the laser diagnostic methods, we transferred the technology to a high temperature supersonic flow facility. A very basic, schematic illustration of this facility is shown in figure 12. In initial studies, we attempted to heat the specimen in the view port area, shown in the figure, using the high temperature plasma. However, the entropy transfer to the sample was inefficient and as a result the sample was heated to no more than 1400°C, which is too cool to produce significant amounts of vapor phase. Thus, in the near term we settled on ablating our specimens in the high velocity gas stream as illustrated. Figure 13 shows a PLIF image of the Zr atom distribution from a sample ablated in this flowing gas stream.

Figure 11. PLIF image of Zr atom distribution in an ablation plume; 840 μ s delay, chamber pressure (argon) = 850 Pa.

Figure 12. Schematic illustration of the radio frequency driven, supersonic flow system used to image the gas species produced by laser ablation into a flowing gas.

Figure 13. PLIF image of Zr atom distribution from a sample ablated in a supersonic flow of argon.

6. Concluding Remarks

In this paper we discussed the application of a boundary layer model to describe the vaporization or corrosion behavior of non-stoichiometric refractory carbide materials at elevated temperatures (>2000 K) in hydrogen. It was shown that, although these materials vaporize incongruently, the mass loss rates can be predicted if the surface composition is known. The basic assumption in the model is that the rate of material loss is controlled by gaseous transport through a static gaseous layer above the solid. Although the molecular theory of gases and gas/solid interactions is relatively well established, we have never directly observed molecular species in a gaseous boundary layer above a solid surface at very high temperatures and made correlations between model and boundary layer thickness. In this paper we describe a laser diagnostic method (referred to as planar laser induced fluorescence or PLIF) that allows for direct observation of atoms or molecules in high temperature gas phase. Although, we have only used our method to image gas phase above laser ablated materials, PLIF can be used

to image boundary layers and, therefore, may be useful for direct verification of the validity of boundary layer models.

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References

1. *America at the Threshold*, Report of the Synthesis Group on America's Space Exploration Initiative. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C., 1991.
2. D. P. Butt and T. C. Wallace, Sr., "The U-Zr-C Ternary Phase Diagram above 2473 K," *J. Am. Ceram. Soc.*, **76** [6] 1409-1419 (1993).
3. E. K. Storms, "The Behavior of ZrC_{1-x} and $U_yZr_{1-y}C_{1-x}$ in Flowing Hydrogen at Very High Temperatures," Los Alamos National Laboratory Report, LA-12043-MS, 1991.
4. D. P. Butt, "Corrosion of $U_xZr_{1-x}C_{1-y}$ Nuclear Fuel Materials in Hydrogen Gas at High Temperatures and Pressures," *Proceedings of the World Space Congress*, paper IAF-92-0570, Washington D.C., 1992.
5. D. P. Butt and T. C. Wallace, Sr., "Modeling the Hydrogen Corrosion of Refractory Carbide Materials: Coupling Solid State and Gaseous Diffusion," to be published in the *Proceedings of the Specialist Conference on Space Nuclear Power and Propulsion Technologies: Materials and Fuels*, published by the Russian Federation Ministry of Atomic Energy, Moscow, Russia, 1993.
6. E. K. Storms and J. Griffin, "Thermodynamic and Phase Relationships of the Zirconium-Uranium-Carbon System," *High Temp. Sci.*, **5**, 423-437 (1973).
7. D. P. MacMillan, "Mass Loss Rates of Uranium-Zirconium Carbide in Flowing Hydrogen and Hydrogen-Hydrocarbon Mixtures," *Proceedings of the 8th Symposium on Space Nuclear Power Systems*, Albuquerque, NM, January 6-10, 1991, published by the Institute for Space Nuclear Power Studies, UNM, Albuquerque, NM, Los Alamos National Laboratory Report LAUR-90-2503.
8. W. M. Rohsenow and H. Choi, *Heat, Mass, and Momentum Transfer*, Prentice-Hall, Inc., 1961.
9. D. E. Rosner, *Transport Processes in Chemically Reacting Flow Systems*, Butterworths, Boston, 1986.
10. J. O. Hirschfelder, C.F. Curtiss, and R.B. Bird, *Molecular Theory of Gases and Liquids*, John Wiley & Sons, NY, 1954.
11. R. A. Svehla, "Estimated Viscosities and Thermal Conductivities of Gases at High Temperatures," NASA TR R-132, 1962.
12. G. Dixon-Lewis, "Computer Modeling of Combustion Reactions in Flowing Systems with Transport," pp 21-125, in *Combustion Chemistry*, Ed.: W.C. Gardiner, Jr., Springer-Verlag, NY, 1984.
13. T. R. Marrero and E.A. Mason, "Gaseous Diffusion Coefficients," *J. Phys. Chem. Ref. Data*, **1** (1) 3-118 (1972).

14. *JANAF Thermochemical Tables*, 3rd Ed., M.W. Chase, Jr., C.A. Davies, J.R. Downey, Jr., D.J. Frurip, R.A. McDonald, and A.N. Syverud, Editors, Am. Chem. Soc. and the Am. Institute of Phys. for the NBS, Vol. 14, 1985.
15. Private communications with M. W. Chase, Jr.: corrections to erroneous data for $C_2H_2(g)$ cited in reference 16.
16. C. E. Holley, Jr., M. H. Rand, and E. K. Storms, *The Chemical Thermodynamics of Actinide Elements and Compounds, Part 6, The Actinide Carbides*, International Atomic Energy Agency, Vienna, 1984.
17. E. K. Storms, D. Hanson, W. Kirk, and P. Goldman, "Effect of Fuel Geometry on the Life Time-Temperature Performance of Advanced Nuclear Propulsion Reactors," *Proceedings of the AIAA/NASA/OAI Conference on Advanced SEI Technologies*, AIAA Paper 91-3454, 1991.
18. D. P. Butt, R. E. Tressler, and K. E. Spear, "Corrosion of SiC Materials in N_2 - H_2 -CO Gaseous Environments: I, Thermodynamics and Kinetics of Reactions," *J. Am. Ceram. Soc.*, **75** [12] 3257-3267 (1992).
19. D. W. Readey, "Gaseous Corrosion of Ceramics," in *Corrosion and Corrosive Degradation of Ceramics*, Eds. R. E. Tressler and M. McNallan, published by American Ceramic Society, Inc. Westerville, OH, 1990.
20. Y. S. Kim and H. W. Pickering, "Kinetics of Chlorination of Co and Co-10At. Pct Pt Alloy by Reaction with HCl Gas," *Met. Trans. B*, **13B**, 249-356 (1982).
21. J. W. Hinze and H. C. Graham, "The Active Oxidation of Si and SiC in the Viscous Gas-Flow Regime," *J. Electrochem. Soc.*, **123** [7] 1066-1073 (1976).
22. D. P. Butt, P. J. Wantuck, and A. D. Sappey, "Laser Diagnostics of Zirconium Carbide Vaporization," *Am. Ceram. Soc. Bulletin.*, in print.
23. P. J. Wantuck, A.D. Sappey, and D. P. Butt, "Measurement of Fuel Corrosion Products Using Planar Laser-Induced Fluorescence," in the *Proceedings of the 10th Symposium on Space Nuclear Power and Propulsion*, Eds. M. S. El-Genk and M. D. Hoover, Albuquerque, NM, American Institute of Physics, Vol. 1, pp. 237-243, 1993.
24. D. P. Butt and P. J. Wantuck, "Imaging and Characterization of Plasma Plumes Produced During Laser Ablation of Zirconium Carbide," in *Laser Ablation in Materials Processing: Fundamentals and Applications*, Eds. B. Braren, J. J. Dubowski, and D. P. Norton, Materials Research Society Symposium Proceedings, Vol. 285, Pittsburgh, PA, 1993.
25. P. J. Wantuck, S. J. Rehse, and D. P. Butt, "Laser Diagnostics of Vapor Phase Produced above Laser Ablated ZrC and NbC," to be submitted for publication in *J. Nucl. Mater.* (tentatively)

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