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A facility for the out-of-pile heating of nuclear fuel samples to above 2000 °C

M T Zharaspaev, D S Kim, R E Zhumagulova

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Abstract. International experience in nuclear energy safety issues shows that the in-pile tests of samples of various kinds of fuels (mixed, low-enriched, regenerated, etc.) involve considerable expenditure and present a higher radiation risk than out-of-pile experiments. The paper describes a facility that allows the out-of-pile investigation of nuclear fuels.

1. Introduction

Results of analyses of nuclear accidents in nuclear power reactors show that the main cause of depressurizing of reactor containment and leakage of ejecta polluted with products from the fission of nuclear fuel into the environment is the buildup of gas pressure in the reactor core. In the temperature range below the melting point ($\approx 1800\,^\circ$C), this increase in gas pressure in the irradiated nuclear fuel (uranium dioxide UO_2 of varying degree of enrichment, mixed uranium–plutonium fuel, etc.) is mostly produced by gaseous and volatile fission products, and in the temperature range above 2000 °C, it is produced by the fuel itself and less volatile fission products [1].

To analyze the distribution of damage in an individual fuel element of the reactor core, we need to know the thermodynamic relations between the variables. Thermodynamic relations in a closed system depend on the spatial positions of fission products. For a hypothetical reactor accident in which most of the reactor core melts, it is appropriate to use thermodynamic relations for an isothermal homogeneous mass.

Presently, several methods are available for the experimental determination of the thermal and thermic equations of state of nuclear fuel materials and fission products at temperatures up to 2000 °C.

2. Advantages of laser heating of nuclear fuel to temperatures above 2000 °C

When the fuel is heated above 2000 °C for the purpose of measuring the equilibrium temperature of the liquid–vapor system, the methods used are those of open evaporation. Heating by electron beams cannot be used because the interaction of the electron beam with the gas cloud above the target affects both the evaporation rate and the reaction pressure in the gas cloud, thus distorting the results of temperature measurements. Furthermore, the large depth of penetration of the electron beam results in unpredictable explosion-type evaporation.

Heating by a laser beam is a suitable method of vaporization of samples of nuclear fuel at temperatures above 2000 °C. It allows measuring thermal equilibrium by finding the rate of evaporation of the target material and the gas pressure.

Measurements of the gas pressure over nuclear fuel using laser heating result in significant changes in the composition of an unevenly evaporating surface. A surface depleted of evaporating components during open evaporation cannot be restored through diffusion from the bulk of the material.

When specimens of nuclear fuel are subjected to laser beam heating, the process of evaporation from the surface becomes stationary after a relatively short transient period. The chemical composition of the surface at this stage is very different from the original composition of the fuel and depends on the actual evaporation temperature. Once this

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M T Zharaspaev Kazakh National Technical University
named after K I Satpayev,
Satpaev Str. 22, 050013 Almaty, Republic of Kazakhstan
Tel. (727) 257 70 87. E-mail: zharaspaev.41@mail.ru
D S Kim Institute of Nuclear Physics, National Nuclear Center,
Republic of Kazakhstan,
Ibragimov Str. 1, 050032 Almaty, Republic of Kazakhstan
Tel. (727) 386 68 00. Fax (727) 386 52 60. E-mail: kim2@inp.kz
R E Zhumagulova International Education Corporation,
Ryskulbekov Str. 28, 050043 Almaty, Republic of Kazakhstan
Tel. (727) 309 63 22. E-mail: roza_j@mail.ru

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stationary, uniform, driven evaporation is achieved, the overall composition of the gas becomes well defined and identical to the isotopic composition of the fuel, which is significantly different from the actual composition of the laser-heated surface.

3. Facility with laser vaporization

Figure 1 shows a schematic diagram of the laser-heating facility that is capable of heating specimens of nuclear fuel to temperatures above 2000 °C.

3.1 Principle of operation

Carbon dioxide (O_2), weakly ionized by glow electric discharge and heated to the plasma state, emits laser beam *1* (see Fig. 1) of about 200 MW in the infrared range at a wavelength of about 5.10 μm. To reduce the high-amplitude pulse fed to the electrodes of plasma injectors of surface discharge and at the same time maintain the density and volume of the plasma produced by them, a plasma circuit breaker is used (*2*). It includes the source of a current pulse connected to two extended electrodes. The electrodes are separated by an insulator, and together they form a vacuum interelectrode gap and a load in the form of a vacuum or plasma diode connected to the electrodes. At least one plasma injector is located on one of the electrodes.

The following experimental arrangement was used to enable changing the angle of the laser beam focus on the nuclear fuel specimen according to the purposes and conditions of specific experiments: beam *1*, after passing through shutter *3*, does not enter vacuum chamber *12* directly but undergoes a double reflection from the system of mirrors *8* and *11*. The optical power of the beam of refracted rays entering vacuum chamber *12* through window *9* is increased by adding converging lens *10*, whose focal length and power can be adjusted depending on the temperature to which the nuclear fuel specimen, mounted on turntable *7*, has to be heated. The sample is locally heated by the beam across ballistic collector *6* and melts at a temperature above 2000 °C. During the pulse, as a result of rotation of mirror *11*, the laser focal point travels uniformly along a circular path on the surface of the sample, and therefore a relatively large amount of the liquid target material is evaporated.

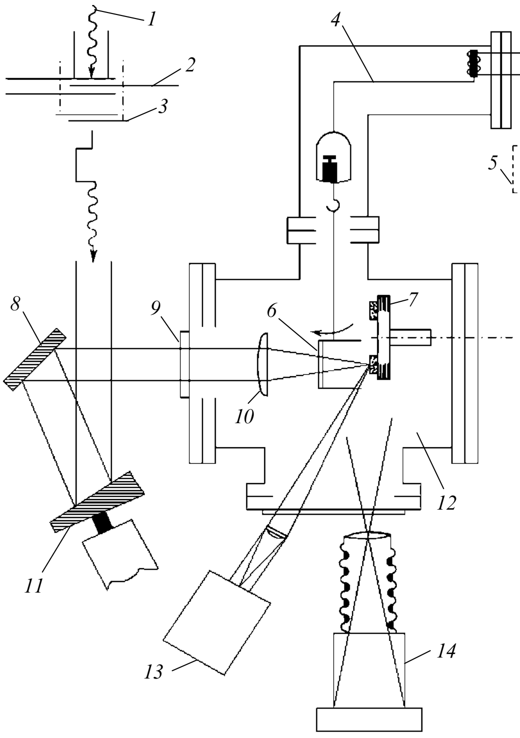

Figure 1. Schematic diagram of the facility for laser evaporation of nuclear fuel specimens at temperatures above 2000 °C: *1* — CO_2 laser beam, *2* — plasma circuit breaker, *3* — shutter, *4* — vacuum balance, *5* — compensation circuit, *6* — ballistic collector with the beam output window, *7* — turntable for targets, *8* — fixed mirror, *9* — beam window, *10* — lens, *11* — movable mirror, *12* — vacuum chamber, *13* — rapid-response micropyrometer for measuring the temperature in the focal spot of evaporation, *14* — camera to monitor the gas jet.

As a result of evaporation of the liquid nuclear material in vacuum chamber *12*, the specimen undergoes thermal decomposition and gases are absorbed. To measure the gas pressure during the evaporation of the fuel at temperatures above 2000 °C, to find the evaporation rate, and to determine the gas permeability of the medium over the heated fuel, the facility uses high-precision vacuum thermal balance *4* with a quartz helix; its sensitivity is about 1 mg mm^{-1} (the deformation of the helix is recorded using a cathetometer with an accuracy of 0.01 mm). Since vacuum balance *4* has a current supply unit (generator) connected in parallel to the laser generator, an equalizing current is generated (a reactive current with supersaturated excitation to the laser generator and capacitive current to the other generator). Compensation circuit *5* is used to reduce excitation in a reactively loaded generator and to increase excitation in a capacitively loaded generator so as to reduce the equalizing current to zero.

3.2 Purpose of heating nuclear fuel specimens to temperatures above 2000 °C

The purpose of experiments on heating specimens of nuclear fuel to temperatures above 2000 °C is the determination of the following indicators:

— time and area of evaporation;

— mass of gas formed;

— instant of emergence of a gas jet, measured from the amplitude of deflection of the pendulum collector mounted in the chamber to observe gas jet *14*;

— spectral thermal radiation from the evaporation surface, which is measured by rapid-response micropyrometer *13*, which allows finding the evaporation temperature of the fuel specimen by measuring the spectral emissivity of the liquid fuel.

Micropyrometer *13* is used for remote measurement of the gas temperature in vacuum chamber *12* because no physical interaction with the medium is possible at such a high temperature. In the cases where, instead of measuring the thermal radiation power, the radioisotope composition of the gas formed as a result of evaporation of a sample of nuclear fuel must be found, a spectrometer can be used instead of micropyrometer *13*.

4. Determination of gas pressure of the evaporated target material

Gas pressure over a heated target is determined from the values of mechanically measured quantities. The pressure of gases constitutes one of the factors affecting the movement of the fuel and coolant during the early stages of an accident in the core of a nuclear reactor: the presence of gases allows larger quantities of fuel and coolant to form a mixture. Unequal accelerations of the fuel and the coolant caused by the presence of fission products in the coolant introduce products of fragmentation or gaseous products of fission. Differences between accelerations in the transfer of the fuel and the coolant affect the process of spontaneous formation of fragmentation products in the fission environment. Nuclear fragmentation affects the propagation of a nuclear accident only slightly, but constitutes an important factor in the process of mixing the fuel and the coolant. The equilibrium vapor pressure of the gaseous component MO_B(g) evaporating from a solid or liquid metal oxide MO_A(cond) can be calculated for the evaporation reaction [2]

$$\mathrm{MO}_A\,(\mathrm{cond}) + \frac{B+A}{2}\,\mathrm{O}_2\,(\mathrm{g}) \rightarrow \mathrm{MO}_B\,(\mathrm{g})\,, \tag{1}$$

where MO is metal oxide, A and B are the stoichiometric coefficients of oxides of various metals, (g) indicates a gaseous state, and (cond) is a the condensed (liquid) state.

Reaction (1) also covers evaporation of pure metals ($A = B = 0$). In the case of thermodynamic equilibrium, the law of mass action yields [2]

$$\Delta G_T(\mathrm{MO}_B) - \Delta G_T(\mathrm{MO}_A) + RT\,\ln\left[-\frac{p_{\mathrm{MO}_B}}{a_{\mathrm{MO}_A}\,p_{\mathrm{O}_2}^{(B-A)/2}}\right] = 0$$

or, after the substitution $RT\,\ln p_{\mathrm{O}_2} = \Delta\bar{G}_{\mathrm{O}_2}$,

$$\ln p_{\mathrm{MO}_B} = \ln a_{\mathrm{MO}_A} + \frac{1}{RT\,\ln 10}\left[\Delta G_T(\mathrm{MO}_A) - \Delta G_T(\mathrm{MO}_B) + \frac{B-A}{2}\,\Delta\bar{G}_{\mathrm{O}_2}\right], \tag{2}$$

where p_{O_2} is the oxygen pressure, p_{MO_B} is the vapor pressure of the gaseous metal oxide MO_B, a_{MO_A} is the activity of the (evaporating) condensed oxide MO_A, $\Delta G_T(\mathrm{MO}_A)$ is the free enthalpy of formation of condensed oxide MO_A at a temperature T, $\Delta G_T(\mathrm{MO}_B)$ is the free enthalpy of formation of condensed oxide MO_B at the temperature T, $\Delta\bar{G}_{\mathrm{O}_2}$ is the oxygen potential of the oxide system, and $R = 8.314\ \mathrm{J\ mol^{-1}\ K^{-1}}$ is the universal gas constant.

The pressure of the gaseous component of the fuel can be calculated if we know the thermodynamic quantities in the right-hand side of Eqn (2).

5. Determination of the target evaporation temperature

In the first approximation, the evaporation temperature is found by detecting the instant of emergence of a gas jet from the heated surface and by applying the laws of gas dynamics to the expansion of the gaseous fuel in the vacuum. Figure 2 [3, 4] plots $\ln p$ as a function of T obtained in experiments with uranium dioxide UO_2, where p is the gas pressure in the medium above the heated fuel. Despite the scatter of the data, mostly due to defects in the material, we note some degree of curving of the line in the region of higher pressures at increased temperatures.

A high evaporation rate at extremely high temperatures alters the composition of the surface layer, which is not given enough time for recovering via diffusion. This change results in a driven uniform evaporation, at which vapor composition does not differ from the composition of the fuel. The curve of driven uniform evaporation *1* (see Fig. 2) was fit to experimental points. Then curve *1* was normalized to the ratio O/U = 2.00, which gave us curve *2*.

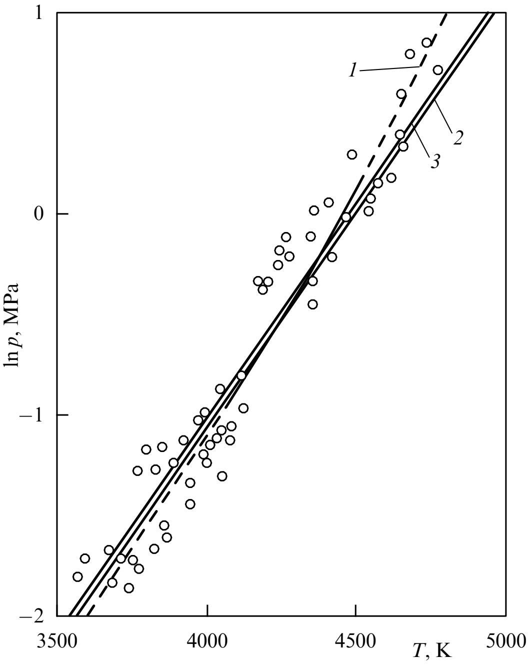

Figure 2. Analysis of experimental data for the total gas pressure over liquid UO_2 at different temperatures: *1* — computational fitting of the curve of driven uniform evaporation to the experimental data (○) by varying the input thermodynamic data within the acceptable error interval, *2* — the curve of driven uniform evaporation normalized to the ratio O/U = 2.00, *3* — the curve of driven uniform evaporation plotted using the equation of state of irradiated fuel UO_2 (the equation of vapor–liquid equilibrium).

In fitting the data, thermodynamic equilibrium was assumed, the Rand–Breitung method was used [3, 4], and we took into account the maximum possible error in the determination of free energies of formation of different fragmentation products suggested in [5]. After normalizing, curve *2* agrees almost completely with the initial data obtained in the temperature range 4400–4700 K [6, 7].

The need to measure deviations from linearity in the dependence $p(T)$ of pressure on the heating temperature stems from the need to obtain relations between the results of measurements of pressure at temperatures up to 4700 K and the range of possible values of the critical point for the heating of uranium–plutonium oxide.

Good agreement is expected between the thermodynamical data, the characteristics of the induced uniform evaporation, the results of measurements of pressure below the melting point, and the melting heat for nuclear fuels heated up to 5000 K. However, at 5000 K or higher temperatures, the deviation of the actual curve $p(T)$ from a straight line cannot be determined with high precision, as is needed for extrapolating the data to the critical point (Fig. 3). It is clear from Fig. 3 that as the nuclear fuel is heated to temperatures above the critical point, the gas pressure of the medium above the fuel changes parabolically, not linearly, as the temperature increases. The following final equation can be derived for UO_2 [1, 8]:

$$\begin{aligned} &\ln p\ [\mathrm{MPa}] = 0.770 \pm 0.013 - \frac{2790 \pm 52}{T}\,,\\ &\Delta H = 533.4 \pm 9.0\ \mathrm{kJ\ mol^{-1}}\,. \end{aligned} \tag{3}$$

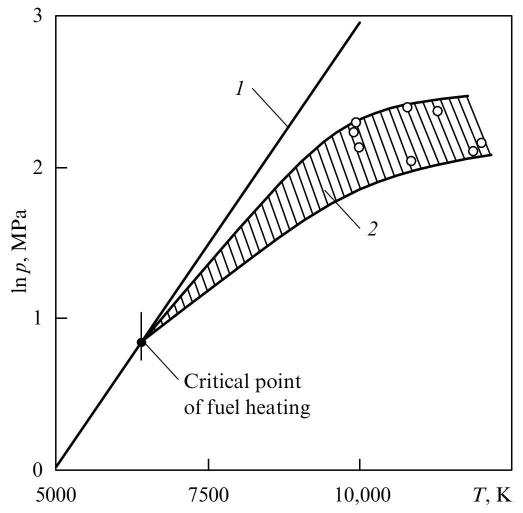

Figure 3. Deviation of the actual curve $p(T)$ from linearity: *1* — linear law, *2* — region of deviation of $p(T)$ from linearity, ○ — experimental data [1, 8].

In the second approximation, the evaporation temperature of a fuel specimen is found by measuring the spectral emissivity of the liquid fuel using a spherical integrating laser reflectometer.

6. Conclusion

Work on the preparation of out-of-pile experiments with nuclear fuel samples has been successfully conducted in a number of countries for a fairly long time. For instance, experiments to investigate the vaporizability of nuclear fuel in a wide range of pressures and temperatures were conducted at the Prokhorov General Physics Institute and at the Institute of Thermophysics of Extreme States, Russian Academy of Sciences, using dedicated laser facilities; the operating principles of these facilities are on the whole similar to those described in this article. The system for laser-beam heating of nuclear fuel samples in a vacuum chamber [9] is technologically the closest to the one presented in this paper.

The facility described above in this paper is specific in that the laser beam passes through a system of two mirrors, one of which is movable, and a collecting lens, and then enters the vacuum chamber. This results in a more uniform heating of the nuclear fuel samples; greater uniformity, in turn, should reduce the error of measuring data at thermal equilibrium, which is itself based on determining the evaporation rate of the target material.

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