2.2 A TECHNIQUE OF MELTING TEMPERATURE MEASUREMENT AND ITS APPLICATION FOR IRRADIATED HIGH-BURNUP MOX FUELS

Takashi NAMEKAWA and Takashi HIROSAWA
Alpha-Gamma Section, Fuels and Materials Division
O-arai Engineering Center
Japan Nuclear Cycle Development Institute
O-arai-machi, Higashiibaraki-gun, Ibaraki-ken, Japan

ABSTRACT

A melting temperature measurement technique for irradiated oxide fuels is described. In this technique, the melting temperature was determined from a thermal arrest on a heating curve of the specimen which was enclosed in a tungsten capsule to maintain constant chemical composition of the specimen during measurement. The measurement apparatus was installed in an alpha-tight steel box within a gamma-shielding cell and operated by remote handling. The temperature of the specimen was measured with a two-color pyrometer sighted on a black-body well at the bottom of the tungsten capsule. The diameter of the black-body well was optimized so that the uncertainties of measurement were reduced. To calibrate the measured temperature, two reference melting temperature materials, tantalum and molybdenum, were encapsulated and run before and after every oxide fuel test.

The melting temperature data on fast reactor mixed oxide fuels irradiated up to 124 GWd/t were obtained. In addition, simulated high-burnup mixed oxide fuel up to 250 GWd/t by adding non-radioactive soluble fission products was examined. These data shows that the melting temperature decrease with increasing burnup and saturated at high burnup region.

INTRODUCTION

The Alpha-Gamma Facility (the AGF) is consisted of equipments for PIE, i.e., twenty one gamma-shielding cells and sixteen glove boxes, and has been operated successfully since 1971. Physical and chemical examination were conducted for the fuel pins irradiated in "JOYO" and overseas reactors such as the Phenix and the FFTF.

In a design of fuel pin, temperature of fuel pellet centerline is limited not to exceed the melting temperature of a fuel pellet. During irradiation, the melting temperature of fuel is considered to decrease slightly with increasing burnup due to buildup of soluble fission products in fuel matrix.

For irradiated and unirradiated UO_2 and $(U, Pu)O_2$ fuels, many workers have determined melting temperatures by so-called V-shaped filament method [1]. However, there have been

large variation in the results which might be the change of the oxygen to metal ratio and/or the vaporization of the specimen at high temperature. To avoid these effects, Lyon et al. applied the thermal arrest technique to determine solidus and liquidus temperatures for unirradiated UO_2-PuO_2 phase diagram [2]. In this technique, an oxide fuel specimen is heated within an sealed tungsten capsule, so that effects of the composition change and vaporization of specimen can be avoided during a measurement . Aitken and Evans also evaluated the melting temperature of unirradiated (U, Pu)O_{2-x} as a function of plutonium content and oxygen stoichiometry, 2-x, using this technique [3].

At the AGF, some apparatus for this technique had been introduced in cells and melting temperature has been measured with good accuracy on UO_2 and (U, Pu)O_2 fuels irradiated in fast breeder reactor (FBRs), an advanced thermal reactor (ATR), and boiling water reactor (BWRs) for more than fifteen years.

This paper describes the melting temperature technique and recent results.

SPECIMEN PREPARATION

Specimens to be examined are both irradiated and unirradiated UO_2 and (U, Pu)O_2 fuel pellets. At first, fuel pellets are granulated by either drilling or crushing in order to pack the fuel specimen into a tungsten capsule. For irradiated fuel pellets, an irradiated fuel pin is cut into small pieces about 40 mm in length and claddings are removed, before granulation. Then about nine grams of granular fuel is carefully introduced into capsule. The capsule containing the granular fuel is then sealed in a high vacuum of 10^{-2} to 10^{-3} Pa by an electron beam welding machine. These preparations are carried out remotely in an alpha-tight steel box(an in-cell box) within a gamma-shielding cell.

On this process, the temperature of the whole capsule is liable to rise up by welding heat. It will cause that FP gas retained in the fuel is put away, the pressure in the capsule heightens, the lid of a capsule is pushed up, and then the normal welding becomes impossible. In the worst case, the fuel in the capsule scatters out. Therefore, it is necessary to keep the temperature of the capsule as much as possible low temperature. Then, cooling method of the capsule under welding was devised in this system. In an electron beam welding machine, the tungsten capsule is set by a chuck. The outline drawing of the chuck is shown in Fig. 1. The base design is a three point scroll chuck , and it was improved into the plane chucking system for fitting to the capsular shape. So that the contact area with the capsule drastically increased, and the heat radiation effect was heightened. The chuck is made of copper metal and tantalum liner so that it has also a function of heat sink. The tantalum liner of the high melting point prevents depleting the copper heat sink by heat in a high vacuum condition. In addition, the stainless plate improves the handling by the manipulator is installed in the upper surface of the chuck.

The welding process was also devised that the capsule is welded in the low power condition temporary, and complete welding is carried out afterwards.

DESCRIPTION OF THE MELTING TEMPERATURE APPARATUS

The main part of the apparatus is located in a gamma-shielding cell. Schematic drawing of the apparatus is shown in Fig. 2 and Fig. 3. The furnace unit is installed in an in-cell box. The vacuum chamber is evacuated to 10^{-2} Pa. The specimen, enclosed in a tungsten capsule, is located in a tungsten crucible which is set in the center of the induction furnace to heat up the tungsten capsule uniformly. The water cooled concentrator is equipped to provide the magnetic field with the crucible efficiently and to shield the glass chamber from high temperature radiation.

The temperature of the specimen is continuously monitored by two sets of two-color pyrometers. Heating power is controlled by a signal from the upper pyrometer. The temperature of the specimen is monitored with the lower pyrometer sighted on a small diameter well at the bottom of the capsule which simulates black-body.

The light path traverses through the revolving protection glass disk, the vacuum enclosure window, and the shielding cell window. The revolving protection glass disk which is nearest to the furnace, prevents the vacuum enclosure window from being coated by vapor deposition. When the protection glass disk is dimmed, the disk is rotated to provide a new clean path.

MAINTENANCE OF THE APPARATUS

On the measurement operation for above 15 years, the capsule occasionally leaked, because of rise of the capsule internal pressure by the volatile FPs, coarsening of crystal grain by the recrystallization of the tungsten and embrittlement being generated in high temperature heating. The volatile FPs which was discharged out under heating deposited to the low temperature portion of the apparatus that is mainly the concentrator in the furnace unit and the exhaust duct of the vacuum pumping system, and the radiation dose rate of the equipment rose very much.

The contaminated concentrator can be exchanged periodically, because the remoteness disassembly is possible. However, the exchange is not possible for the exhaust duct, since it is being fixed in the in-cell box. In the point maximum, the dose rate reached 320 mSv/h. Such a high radiation condition interfered with ordinary maintenance of the apparatus.

Then, the equipment for the remote decontamination was devised, and the exhaust duct inside and heating furnace inside were tried to decontaminate. However, it was not possible to lower only to about 190 mSv/h, since the inside is very narrow and sufficient decontamination was impossible.

Some radiation shielding devices was designed and used for ordinary maintenance such as exchange of the oil of the vacuum pump and exchange of the globe under the environment of such high dose rate. The shielding screen consisted of the lead on 1800 mm height, 1300 mm width, 30 mm thickness. By using these shielding devices, the environment of maintenance area was secured and maintenance of the apparatus was able to be carried out. However, this work is hard to be carried out frequently. Therefore, the whole equipment will be renewed in FY 1999.

EXPERIMENTAL DATA AND DISCUSSION

1. Accuracy

It is most important to obtain the accurate temperature of a specimen in the melting point measurement. It is necessary that absolute temperature is calibrated by the calibration formula in the two-color pyrometer.

The temperature reproducibility is high for this pyrometer itself, and the measurement with the high accuracy becomes possible, if the calibration formula is obtained. There is the ratio of observed emissivity to rue one of measured object (i.e., tungsten) as a main error factor of the pyrometer. The emissivity of tungsten is well known, but there are some differences by the surface state (e.g., roughness). Then, the temperature measuring well was made at the capsule bottom to simulate black-body, in which the effective emissivity was about 1. At first, the well was designed the dimension with 1 mm in diameter and a depth of 10 mm. However, adjusting condition for focusing of the pyrometer is very severe, since a distance from the tungsten capsule is about 2 m and a sight range of the pyrometer is not less than 1 mm in diameter. Then, diameter of the well was enlarged up to 2 mm with the same length that is the limit to get a black-body condition. It contributes to good accuracy of measured temperature data. The uncertainties of measured temperature on each capsule were reduced by this method.

Furthermore, transmission of the glass and reflection by the mirror in the measurement optical path affect the emissivity, and it becomes a factor of the indication error. Then it must be corrected as an emissivity ratio. The correction is possible to be carried out by these condition being fixed. However, it must be evaluated at each measurement, since the transmittance of the window glass gradually changes by each measurement operation. At the AGF, melting point of reference sample (i.e., Mo metal of mp 2630℃ and Ta metal of mp 2990℃) is measured before and after the measurement of the fuel as an error evaluation method. These reference sample were selected since they are metal sample which melting point is around and near to the fuel's one.

As the above description, the slippage of melting point around the measurement of reference sample was adopted as an error of the pyrometer indicating temperature. The whole measurement error is added reading error of thermal arrest in the heating curve. The melting point of unirradiated UO_2 was verified being 2845 ± 12℃ at the AGF in which 2847 ± 25℃ is the recommendation value of the IAEA [4].

As the other report for unirradiated UO_2 using with the encapsulation method, Aitken & Evansns [3], Lyon & Baily [2] and Latta & Fryxell's [5] is representative, and the measurement accuracy is ± 25℃, ± 20℃, ± 15℃ each. These measurement error do not carry out the evaluation of the every sample like this report. For irradiated MOX fuel,Krankota &Craig [6] reported the error $\pm 52 \sim \pm 130$℃. This large error was caused by the V-shaped filament method, since the specimen is not enclosing and the composition change is easy to be caused by unequal evaporation and the oxygen transfer. Adamson et al.[7] mentioned about the reliability of the V-shaped filament method that this method gave melting temperatures approximately 50℃ lower than true one.

2.Melting Temperature Data

At the AGF, melting temperature has been measured with good accuracy on UO_2 and (U, Pu)O_2 fuels irradiated in FBRs, ATR, and BWRs. Typical data are shown in Fig.4.

The melting temperature of 29 wt% PuO_2-UO_2 obtained at the AGF are plotted,, comparing with the data of 25 wt% one reported by Krankota and Craig[6]. Their data locate at higher temperature range than the AGF's, and it is well-known that melting temperature decreases with an increase in PuO_2 concentration[6,7].

In this data, the maximum burnup of specimen is 124 GWd/t which was irradiated in "JOYO". This figure also includes the data obtained from simulated burnt MOX fuels. They were prepared by adding the non-radioactive oxide powder of eight soluble FP elements to unirradiated or irradiated fuel powder, that amount corresponds to the maximum burnup of 250 GWd/t.

These data shows that the melting temperature of 29 wt% PuO_2-UO_2 decrease almost linearly with burnup at a rate of 5 ℃ within the lower burnup region up to 170 GWd/t, and almost saturate above it. It may be necessary to measure the actual specimen of higher burnup in order to confirm this behavior.

SUMMARY

A technique and operation for melting temperature measurement of irradiated oxide fuels at the AGF is described. In this technique, the fuel is enclosed in a sealed tungsten capsule to prevent chemical change, and heated in the induction furnace. The melting temperature is determined from arrest of the heating curve.

Observed melting temperature of MOX fuel decrease with increasing burnup and saturated at high burnup region.

REFERENCES

[1] For instance; Christensen, J. A., et. al., WCAP-6065 (1965).
[2] Lyon, W. L., Baily, W. E., J. Nucl. Mater., 22 (1967) 332.
[3] Aitken, E. A., Evans, S. K., GEAP-12229 (1971).
[4] M.H.Rand et al., Rev. Int. Hautes Temp. Refract. 15 (1978) 355.
[5] R.E.Latta & R.E.Fryxell, J. Nucl. Mater., 35 (1970) 195.
[6] Krankota, J. L., Craig, C. N., GEAP-1315 (1969).
[7] M.G.Adamson, et al., J. of Nucl. Mater., 130 (1985) 349.

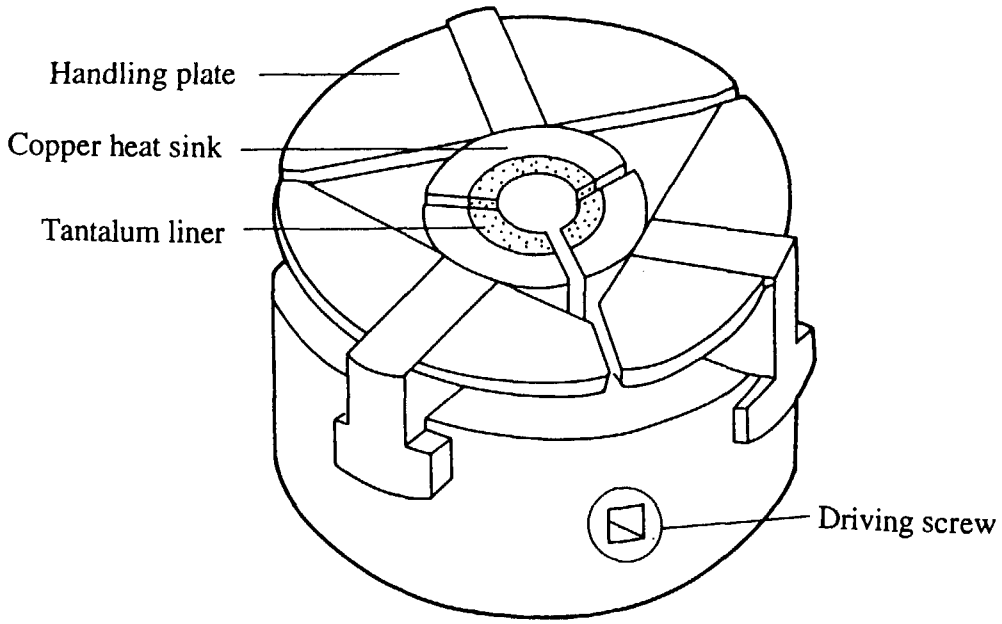
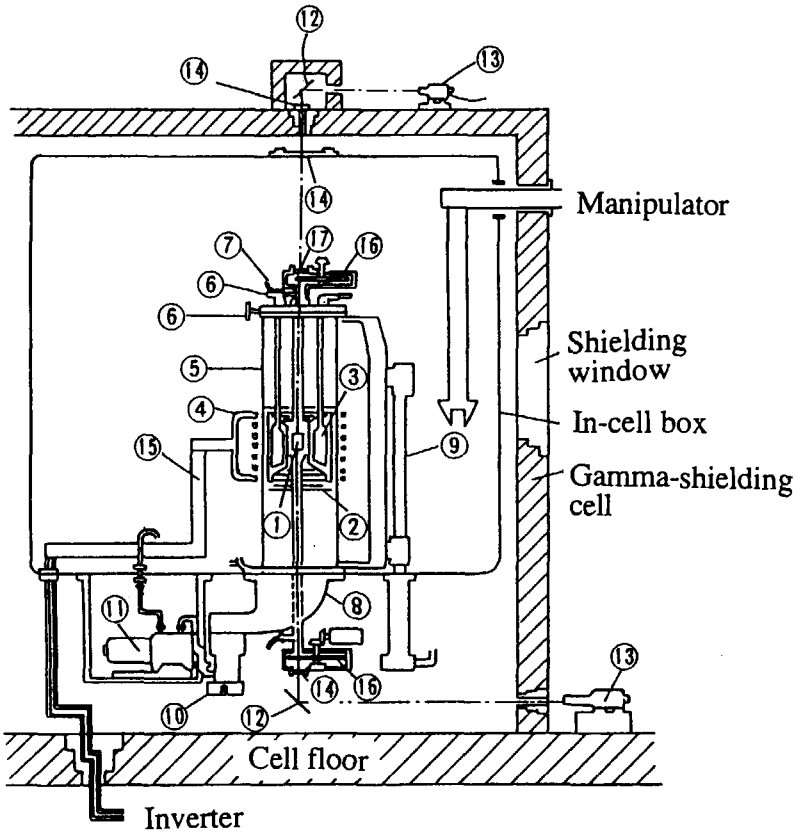


Fig. 1 Structure of the capsule chuck in an electron beam welder



① Specimen (Tungsten capsule)
② Heat shielding
③ Concentrator
④ Induction heating coil
⑤ Vacuum chamber (Quartz)
⑥ Cooling water
⑦ Argon gas supply
⑧ Exhaust duct
⑨ Hydraulic lift
⑩ Diffusion pump
⑪ Rotary pump
⑫ Mirror
⑬ Two-color pyrometer
⑭ Window
⑮ Induction cable
⑯ Protecting glass

Fig. 2 A schematic drawing of the melting temperature measurement apparatus

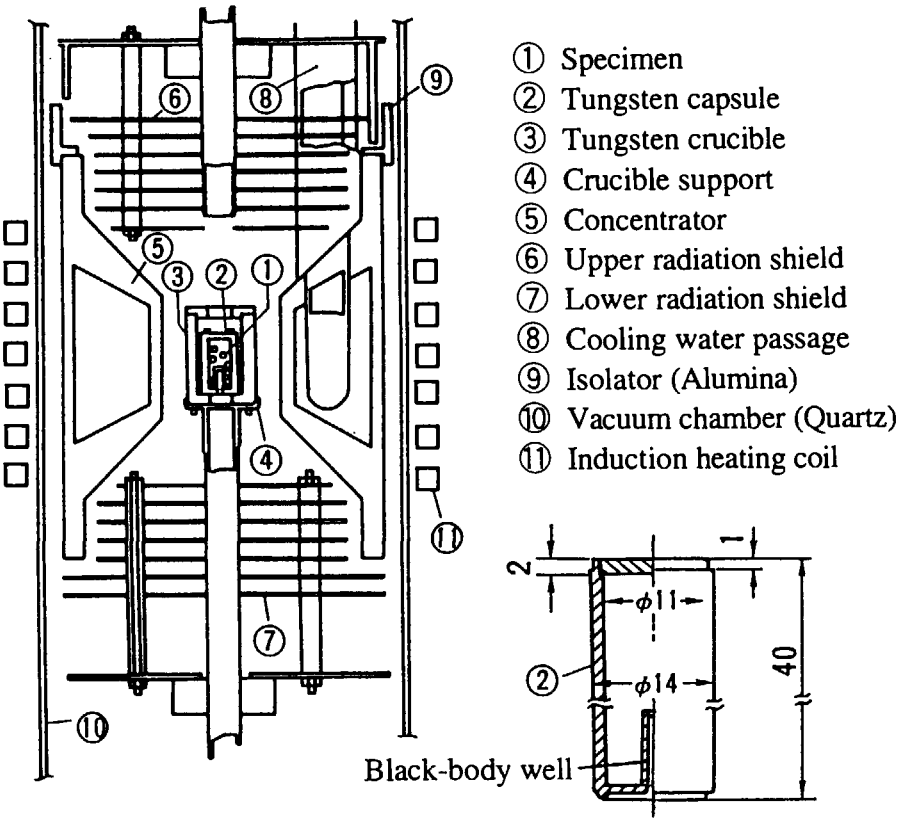


Fig. 3 A schematic drawing of the induction furnace unit and a tungsten capsule

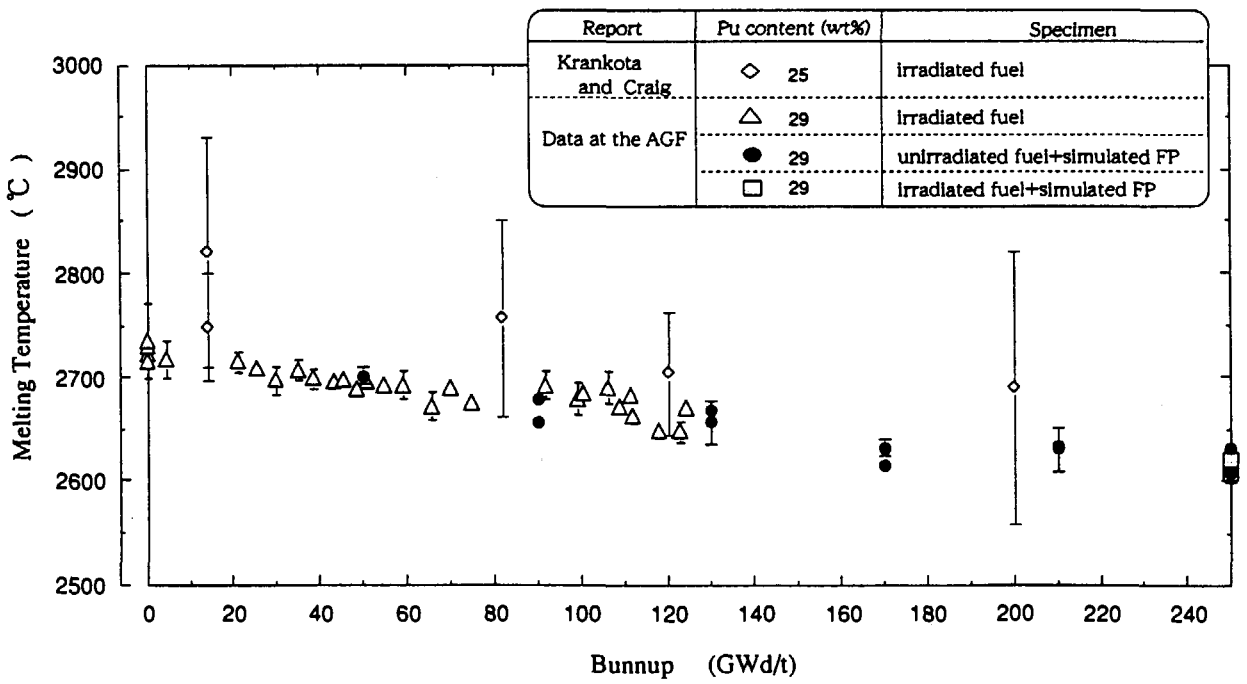


Fig. 4 Melting temperature of MOX fuels as a function of burnup