Determination of density and open porosity of irradiated fuel by the immersion method

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Abstract

The methods used at the Studsvik Hot Cell Laboratory to measure densities, mainly of fuel but also of other materials are described. Specifically a comparison is made of measurements of fuel density of separate fragments to measurements on fuel disks contained in its surrounding cladding. The two methods are shown to give very similar values.

Background

Density measurements are mainly performed to assess the densification and swelling of irradiated fuel, but can also be used to determine the density and thus swelling of other materials, such as zirconium alloys (which can swell due to hydriding) or stainless steel (void swelling).

Method

Studsvik currently employs an immersion density measurement method usually using bromoform $CHBr_3$ with a density of 2,74 g/cm^3 as the immersion liquid. The method is based on Archimedes' principle, but also accounts for open porosity. It consists of weighing the sample in air (=W_1), then weighing it after evacuating and immersing in the immersion liquid (=W_2), and then finally weighing again in air with the open porosity filled by the immersion liquid (=W_3). The bulk density and open porosity are then determined as follows (see Figure 1):

$$V_{op} = \frac{W_3 - W_1}{\rho_{liq} - \rho_{air}} \qquad (1a)$$

$$V_{cp} + V_m = \frac{W_1 - W_2}{\rho_{liq} - \rho_{air}} \qquad (1b)$$

$$\rho_{bulk} = \frac{m}{V_{op} + V_{cp} + V_m} \qquad (1c)$$

where V_{op}, V_{cp} and V_m are the volumes of the open porosity, the closed porosity and the matrix material respectively, ρ_{air} and ρ_{liq} are the densities of air and the immersion liquid, ρ_{bulk} is the sample bulk density, and m is the sample mass (approximately equal to its dry weight in air W_1).

The accuracy and precision of this technique, when used for irradiated fuel samples, is around ±0,03 g/cm^3. In addition, the measurements can be repeated, this improves the statistics.

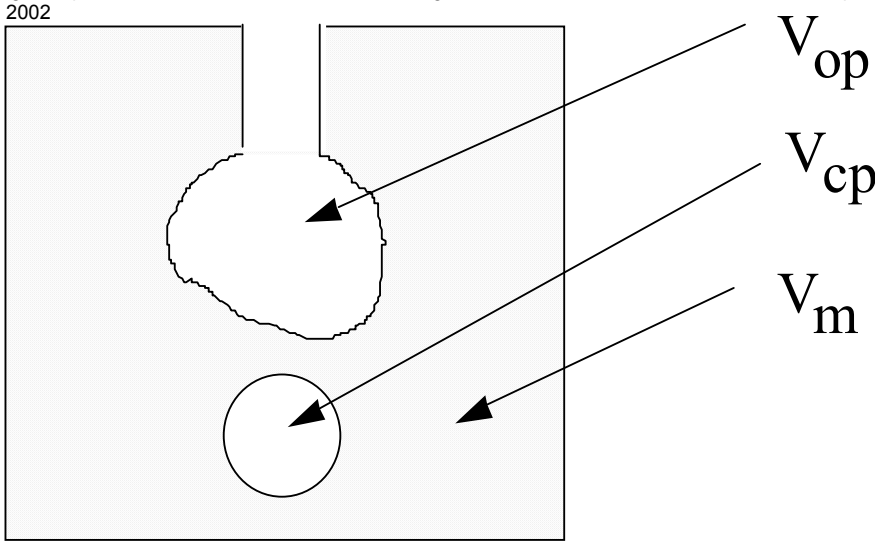

Figure 1 Definition of volumes.

A balance (Mettler AE 200) with a weighing accuracy of 0,0001 g is used in the measurements. The balance is calibrated to weigh samples with a density of 8 g/cm^3. When the density deviates from this, an equation is used to compensate for the difference. A wire basket of platinum is used to suspend the samples while submerged in the immersion liquid. To check the density of the immersion liquid which may vary slightly a standard density sample of Zircaloy is always used in connection to each measurement to measure the density of the bromoform.

Samples

Irradiated fuel pellets are frequently cracked, so the largest individual fragments are used as seen below. It is known that the density varies along the pellet radius especially in high burnup fuel. The chosen fragments can not represent the entire radius of a cross section since the position of the fragments in the original pellet is unknown and hence, it is not possible to determine exactly the average density based on the entire cross section.

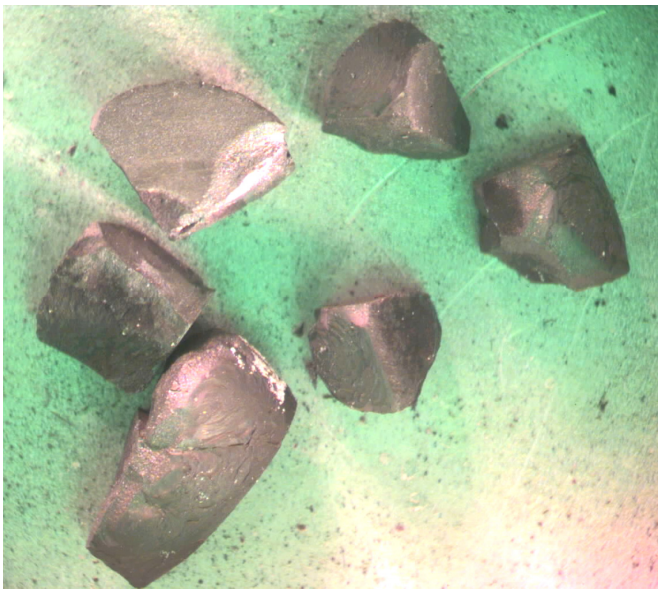

Figure 2 Separate fuel fragments measured individually.

Procedure for measuring separate fragments.

1. The samples are baked in vacuum at 110 °C to become completely dry.
2. The samples are weighed in air.
3. The samples are placed in a beaker in air in a vacuum jar. The jar is evacuated to remove gas from the open pores of the samples. The samples are then submerged in bromoform. Liquid enters the open porosity when atmospheric pressure is restored. The standard weight and the wire basket are placed in the beaker.
4. The samples and the standard weight are weighed, suspended in the wire basket and submerged in bromoform.
5. The specimens are dried on a wet cloth and weighed quickly before the liquid evaporates from the pores.
6. The samples are once again baked in vacuum, and weighed in air to check that nothing has happened to the samples during the measurement
7. The steps 3 - 6 are repeated.
8. The bulk density and open porosity are calculated from the measured data.

In some cases, for instance at high burnups, it is impossible to remove the entire fuel pellet from the cladding. In these cases it is necessary to first determine the volume and mass of the fuel plus cladding, then remove the fuel (mechanically and chemically) and determine the volume and mass of the remaining cladding and cladding oxide as seen below.

Figure 3 Fuel bonded to the cladding and the cladding after chemical fuel removal.

When measuring the density of fuel contained in a cladding ring as for high burnup fuel bonded to the cladding it is not possible to measure the open porosity as the cracks in the fuel will behave similar to open porosity. Thus, only the density of the fuel matrix including the closed porosity will be measured. Luckily, the open porosity is mostly very small in high burnup fuel making the density measured in this way very close to the real bulk density.

Procedure for measuring fuel enclosed in the surrounding cladding.

1. The samples are baked in vacuum at 110 °C to become completely dry.
2. The samples are weighed in air.
3. The samples are placed in a wire basket in a beaker in a vacuum jar. The jar is evacuated to remove gas from the open pores of the samples. The samples are then submerged in bromoform under vacuum. Liquid enters the open porosity when atmospheric pressure is restored.
4. The samples are suspended in a wire basket, submerged in bromoform, and weighed.

5. The fuel is chemically removed from the cladding rings and the cladding rings are measured once more in the same way as the complete samples.
6. The fuel weight and volume is obtained from the two measurements and the density is calculated.

Examples of measurements

To compare the two methods of density measurements, (on separate fragments and a complete cross section surrounded by cladding), samples were taken only 6 mm apart and measured with the two methods. Two rods were sampled in this way giving six fragments each with weights between 0,2 and 0,5g. The weight of the fuel disks was about 2g. The resulting data are shown below.

	Bulk density of fragments (g/cm^3)	**Open porosity (%)**	**Bulk density excluding open porosity[1] (g/cm^3)**	**Disk minus Fragment density (g/cm^3)**
Rod 1 (fragments)	9,984	0,053	9,989	0,008
Rod 1 (fuel disk)			9,997	
Rod 2 (fragments)	10,023	0,086	10,032	-0,005
Rod 2 (fuel disk)			10,027	

1) Density calculated from the fuel mass divided by the (volume of fuel matrix + closed porosity)

Conclusion

Apparently, the density of fuel can be measured with a high degree of accuracy even when the fuel is contained in the surrounding cladding. This method also ensures that the density value obtained is a correct representation of the average density of a cross section of the fuel pellet.