Towards Density Determination of Pyrolytic Carbon Layers Deposited on Nuclear Fuel Microspheres

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In High-Temperature Gas-Cooled Reactors (HTGR), the fuel is applied in the form of small spherical kernels, some 100 µm in diameter depending upon the fuel cycle concept. The fuel kernel is coated by a sequence of pyrolytic carbon layers. The inner layer adjacent to the kernel is always highly porous to provide the particle with a free volume for the fuel kernel swelling and the gaseous fission product reception. The outer high density layers form a pressure vessel and fission product barrier. It is obvious that because of the importance of the particular layers the apparent, bulk or geometric density has to be controlled carefully.

The APPARENT, BULK OR GEOMETRIC DENSITY is the mass per unit volume. It is assumed that the volume is homogeneously filled with mass. It includes the volumes of the substance, displacements, micro and macro pores - open and closed - and of the internal voids.

The geometric density is measured by determining the increase in volume and mass after the deposition of the particular pyrolytic carbon layer. The diameter can be determined by use of microscopic measurements or x-ray micrographs.

For statistical considerations and on-line quality control an automatic optical particle analyzer developed by ÖSGAE Seibersdorf/Austria is used (1). A quantity of coated particles is moved along a constant light source and a very small measuring gap with intervals of approximately 20 particles per second. The gap area shadowed by a particle is measured by means of a photodiode and is a measure for the diameter. The apparatus is controlled and the data are evaluated by an on-line process computer. This technique is suitable for diameters of 150 to 1500 µm. After weighing the investigated portion of particles, the apparent density can be calculated.

As far as the porous buffer layer is concerned, it is not sufficient to determine the density directly after its deposition. During the deposition of high dense pyrolytic carbon on the buffer layer it may occur that at least the outer zone is infiltrated by carbon and thus densified. Therefore it is necessary to measure the densities of both the inner and outer layers once more after fuel production by applying the SINK-FLOAT method.

The purpose of this method is to determine the density of layers by suspension in a density gradient column. The gradient column consists of isobutanol and bromoform chemicals mixed continuously in varying proportions so as to cover a range of densities from 0.8 $g \cdot cm^{-3}$ to 2.8 $g \cdot cm^{-3}$. Pyrolytic carbon fragments cracked from coated particles are suspended into the column. The density is defined at the point in the column where the test specimen of pyrolytic carbon has reached the same density as the liquid in which it is suspended (fig. 1) and thus floating in the column.

The geometric and sink-float densities of a sample are equal, provided the substrate is impermeable. The permeation depends on the pore size as well as surface tension, but not on the viscosity of the liquids used. The densities mentioned above are not equal in the case of low density carbon, because the liquid in the column penetrates the pores with an open entrance greater than 10nm. Therefore the volume measured is too small, so that the reading of the sink-float density is too high (fig. 2). PECHIN et al. have proposed an equation to correct these influences (2).

In KFA this problem has been solved experimentally. Studies have been carried out to prove the combination of two measuring procedures to yield true values for the apparent or geometric density of the buffer layer even after the deposition of the high dense layer:

- sink-float method
- quantitative image analysis.

The quantitative image analysis measures macropores with diameters greater than 200 nm. For this procedure, the pyrocarbon coatings are applied in the form of ceramographic sections of the coated fuel particles.

The measurement is carried out by an electronic, automatic image analyser. Working mode is the classification of the coating to be inspected on the basis of the gray value contrast between pores (dark) and carbon (bright).
From the sink-float density, d_s, one can calculate a porosity, P_s, which is too small because the closed pores only are considered

$$P_s = 1 - \frac{d_s}{d_x} \qquad (1)$$

with d_x as X-ray density. By the quantitative image analysis the total macro porosity, P_M, is measured. For buffer layers the total porosity, P, results in

$$P = P_s + P_M \qquad (2)$$

This equation is valid for pyrolytic carbon with closed pores smaller than 200 nm.
The apparent, bulk or geometric density, d, of the porous pyrolytic carbon can be calculated by applying the equation

$$d = d_x(1 - P) \qquad (3)$$

The results obtained have shown, that density values from mass and volume measurements and from the application of the combined methods described above agree well for porous buffer layers (3).

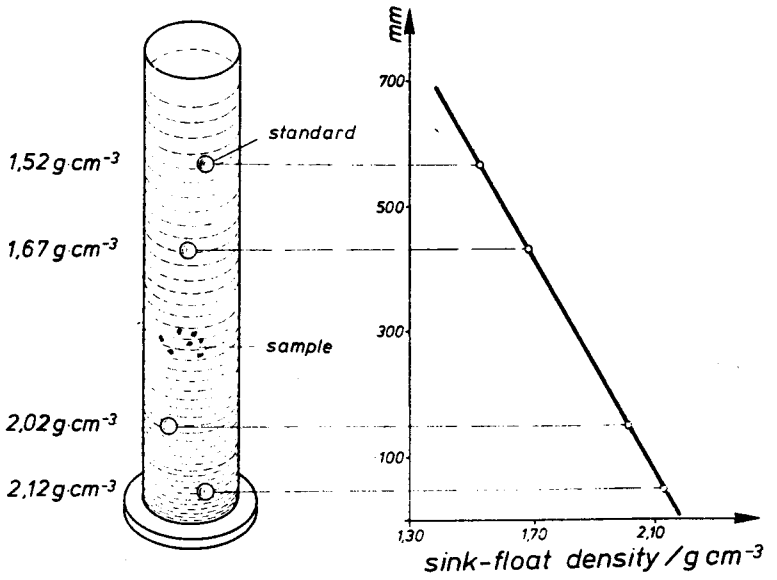

Fig. 1: Gradient column for the measurement of sink-float density

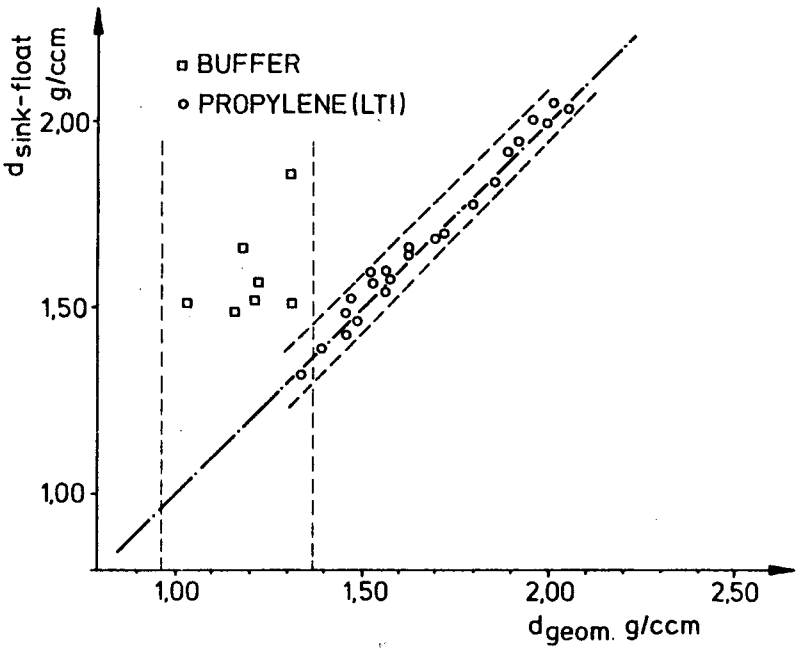

Fig. 2: Interrelation between sink-float and geometric density

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