Effect of testing conditions on the diffusivity measurements of nuclear fuels via the flash method

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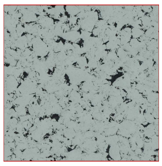
Context and objectives

$\lambda(T) = a(T).C_p(T).\rho(T)$
a : thermal diffusivity
$C_p(T)$: heat capacity
$\rho(T)$: density

Thermal conductivity of the nuclear fuel :
→ A key parameter to understand the performance of the fuel under irradiation
→ Highly dependent on microstructure, porosity and its distribution…
→ Influences almost all important processes such as fission gas release, swelling…

Knowledge of the nuclear fuel thermal conductivity (**derived from a thermal diffusivity measurement**) :
→ A fundamental data for a better prediction of its performance in reactor
→ **Must be measured accurately** (including a reliable uncertainty assessment)



Determination of thermal diffusivity by the flash method with a LFA 467 HyperFlash® NETZSCH apparatus

Principle : heating of the front surface of a **plane-parallel sample** by a **short energy light pulse** and measurement of the **resulting temperature excursion** of the rear face

Technical data

Temperature range from ambient to 500 °C
Xenon-pulse delivery source
Liquid nitrogen-cooled InSb **IR detector**
ZoomOptics system (optimized field of view)
Gas control unit

Range of measurement : 0,01 to 1000 $mm^2.s^{-1}$
Accuracy : ± 3 %
Repeatability ± 2 %

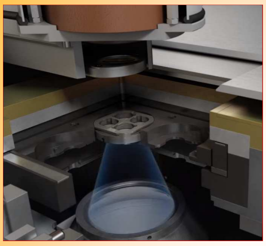
Thermal diffusivity identification

Fitting of a calculated model to experimental data (**Proteus® software**)
- Ideal case : **adiabatic model** (no heat loss)
- **Cowan model**: 1-D mode, purely axial heat transfer (no radial losses)
- Improved **Cape & Lehman model** : 2-D axial-symmetric mode, (radial and facial losses)

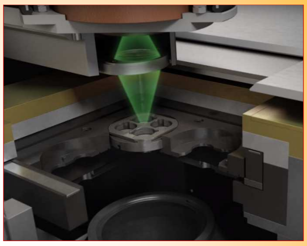
Description of the Flash method

a transient heat flux technique

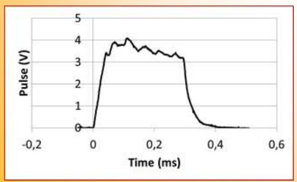
Heat pulse generation



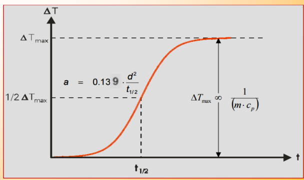
Optical detection



Light pulse shape



Signal measurement

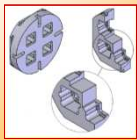


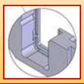
Test specimens

Thin ceramic samples (~ 2 µm thick)
- different shapes (circular, square, rectangular)
- different sizes (8 to 12.7 mm in diameter or side length)
- **Deposit of an opaque layer on both faces** to allow full absorption of the energy pulse
  - tungsten or gold coated and graphite sprayed dense **Pyroceram® 9606** (BCR -724, standard)
  - graphite sprayed porous **uranium dioxide** (obtained by a powder metallurgy route)

Sample holders

- Steel supports, designed for each sample geometry
- Loosely thermally coupled to the specimen

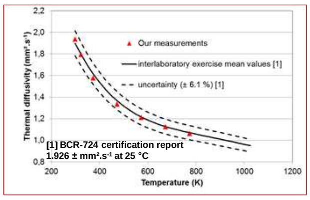




Experimental data

Verification

BCR-724 certified thermal diffusivity

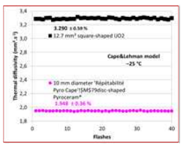


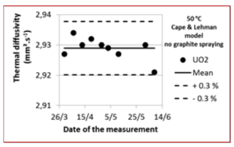
Interlaboratory comparisons

Pyroceram® sample	Thermal diffusivity at ~ 25°C ($mm^2.s^{-1}$)							
	Our measurements			LNE measurements		Netzsch measurements		
10 mm disc-shaped	Cowan model	1.969	± 0.003	De Giovanni model	1.94	/		
	Cape Lehman model	1.948	± 0.004					
12.7 mm disc-shaped	Cowan model	1.940	± 0.004	/		Cowan model	1.932	± 0.007
	Cape Lehman model	1.926	± 0.006					

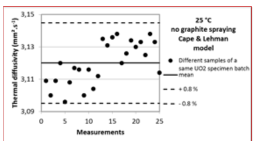
Good reproducibility

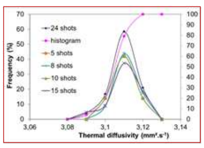
Repeatability





NB: A measurement result is the average of several shot results (usually ~ 20)

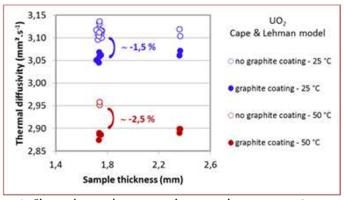




Within-batch repeatability better than 1%
Repeatability better than 1%

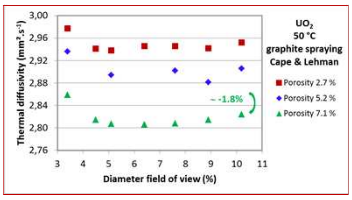
Effect of testing conditions

Effect of graphite spraying

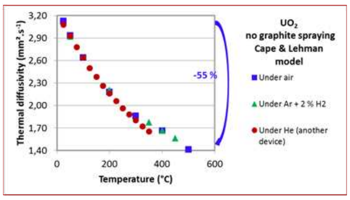


- Clear dependence on the sample preparation
- Choice of spraying on both faces of the sample

Effect of the field of view

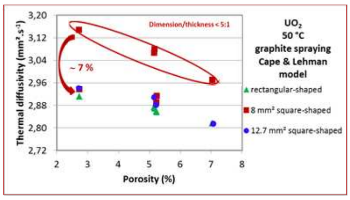


Effect of the temperature



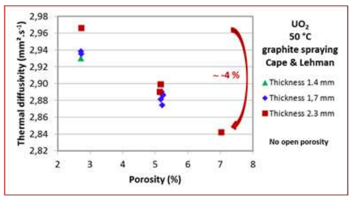
Effect of material characteristics

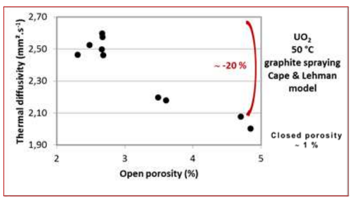
Effect of the sample geometry



- Slighty higher apparent diffusivity for 8 mm^2 square-shaped samples (differences between geometries not taken into account in the proposed models)
- Dimension to thickness ratio must be > 5:1

Effect of the specimen porosity





Strong dependence on the open porosity

Conclusions

- Flash method → **Appropriate to discriminate differences in the thermal diffusivity measurements of UO_2 samples** caused by a variation of the porosity
- **Recommendations for the operating conditions** : geometry (dimension to thickness ratio > 5:1) and preparation of the samples (graphite spraying on both faces)

Perspectives

- Carrying out of tests under different atmospheres (**prevention of the oxidation of UO_2 at temperatures > 300°C**)
- Evaluation of the **effect of the fabrication process/material structure**
- Use of image analysis in order to **quantify the pore size morphology and distribution** and to correlate these data with the thermal diffusivity measurements