CHARACTERIZATION OF THERMAL PROPERTIES OF SIC_F/SIC COMPOSITES FOR ENHANCED ACCIDENT TOLERANT FUEL

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ABSTRACT

The joint program of Framatome, CEA and EDF for the development of Accident Tolerant Fuel (ATF) led to consider SiC_f/SiC refractory composite materials as potential candidates to enhance accident tolerance for current Light Water Reactor (LWR). One of the key challenges in the development of LWR cladding design consists in ensuring that the heat exchange between pellet and coolant is optimal. In this purpose, assessing the through-thickness thermal properties of SiC_f/SiC is essential for the cladding design. Once adapted to cylindrical geometries and validated on monolithic SiC, an experimental flash method has been set up to measure the thermal diffusivity of SiC_f/SiC tubular samples, under two types of conditions (mechanical damage and LWR environment). The results of these characterizations are valuable for a better understanding of the thermomechanical behavior of the SiC_f/SiC cladding, which is one of the mandatory information to be provided for the ATF irradiation program planned for 2022.

1. Introduction

Tubular nuclear-grade SiC_f/SiC composites produced by CEA were investigated in the present work as cladding material to enhance the accident tolerance of Light Water Reactors (LWRs). The main advantage of a silicon carbide-based solution is that it keeps its dimensional stability and mechanical integrity when reaching accidental temperatures while exhibiting low interaction with steam. One of the key challenges in the development of LWR cladding design is to have an optimal heat transfer between the pellet and the coolant. In particular, the thermal conductivity of the cladding must be high enough so as to minimize thermomechanical damage under high heat fluxes.

The flash method, which is the most used experimental technique to measure the thermal diffusivity of solids, is usually run on flat samples. Hence, the method has been adapted to cylindrical, highly diffusive and heterogeneous multilayered samples. A homogeneous monolithic SiC cylinder has been tested first to validate the experimental method that has then been applied to SiC_f/SiC tubular samples. The thermal properties were evaluated on samples exposed to two types of conditions. First, the method has been used in order to assess the evolution of thermal properties before and after exposure to a representative PWR environment (without irradiation). Second, tubes have been adapted on an axial tensile test device so as to measure the evolution of thermal diffusivity with intentional mechanical damage.

2. Materials and experimental details

2.1. Samples

Three types of samples have been used in this study. The first one, which is used for the validation of the experimental process, is a monolithic tubular sample of silicon carbide, with an inner radius of 5.4 mm, a thickness of 2.1 mm, and a thermal diffusivity of ~ 70.10^{-6} m².s^{-1} (manufacturer data). The two other samples are SiC$_f$/SiC tubes, manufactured with braided SiC fibers chemically infiltrated by a SiC matrix, with an inner radius of ~3 mm and an external radius of ~4 mm (Figure 1).

Figure 1: Representative SiC$_f$/SiC tubular specimen processed at CEA

2.2. Estimation model

The flash method, which has been first introduced by Parker & al. [1], consists in following the thermal behavior of a sample after a pulse heating on its front face by observing the evolution of the temperature at its front and rear faces (Figure 2).

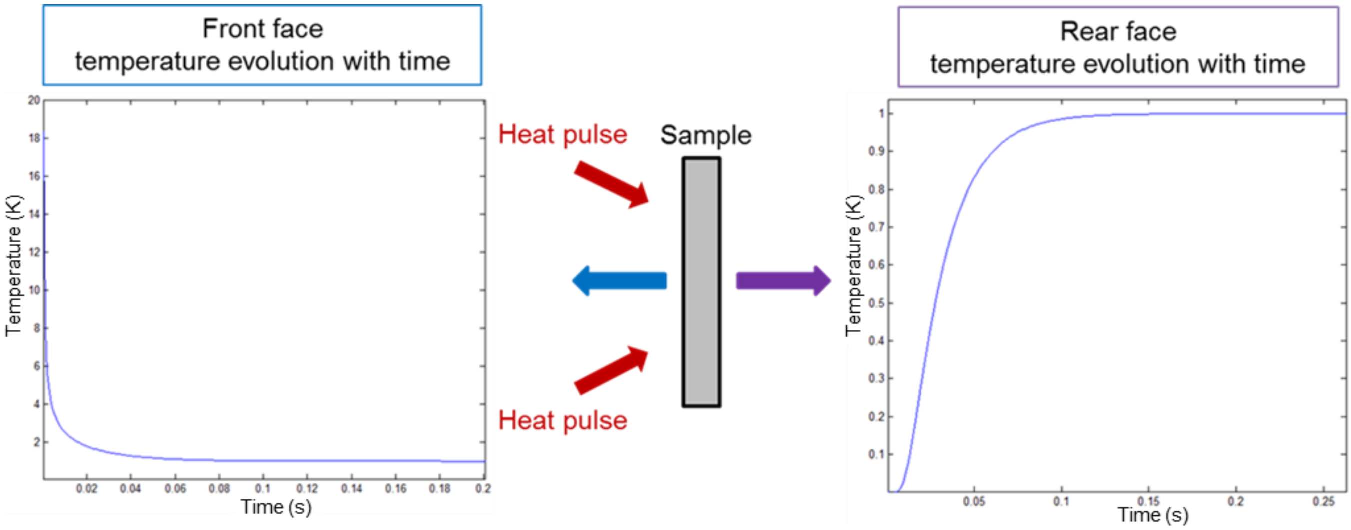

Figure 2: Flash method experiment and associated temperature responses

It allows the identification of the thermal diffusivity a (m².s^{-1}) over a large characteristic frequency range, which translates the ability of a material to transmit a temperature signal from one point to another, according to the heat equation (Eq. 1):

$$\partial_t T = a \Delta T \, , \qquad \text{(Eq. 1)}$$

with t the time (s) and T the temperature (K).

Originally, models have been developed for the identification of the diffusivity of isotropic homogeneous opaque solids, using direct methods. Progressively, models have become more representative and adapted to different types of materials. The flash method can be described accurately using the quadrupoles formalism [2], since it allows taking into account all the parameters involved in a flash experiment on tubular geometries [3] (cylindrical coordinates, heat losses, flash form and duration). After all due verifications, it has been shown that the evolution of temperature on both faces of our samples could be described in direct space and time [4] by the following analytical solutions (Eq. 2):

$$T(z,t) = \frac{Q}{\rho c e}\left[1 + 2\sum_{n=1}^{\infty} \cos\left(\frac{n\pi}{e} z\right) \exp\left(-\frac{n^2\pi^2}{e^2} at\right)\right] \qquad \text{(Eq. 2)}$$

With z the thickness variation (m), Q the flash power density (W.m^{-2}), ρ the density (kg.m^{-3}), c the specific heat (J.kg^{-1}.K^{-1}) and e the thickness of the samples (m). The identification of the diffusivity is performed using a least squared method to minimize the the error between the model and input signals for both front (z=0, (Eq. 3)) and rear faces of the sample (z=e, (Eq. 4)):

$$T(0,t) = \frac{Q}{\rho c e}\left[1 + 2\sum_{n=1}^{\infty} \exp\left(-\frac{n^2\pi^2}{e^2} at\right)\right] \qquad \text{(Eq. 3)}$$

$$T(e,t) = \frac{Q}{\rho c e}\left[1 + 2\sum_{n=1}^{\infty} (-1)^n \exp\left(-\frac{n^2\pi^2}{e^2} at\right)\right] \qquad \text{(Eq. 4)}$$

2.3. Experimental apparatus and validation

Flash devices are usually set up to test flat samples. Therefore, the classical flash test bench has been adapted to cylindrical geometries in order to characterize the tubular samples. In this purpose, samples have been settled at the focal center of four flash lamps (Digital 600RX – 300 J each), evenly spaced around the center to ensure a quasi-uniform heating. The recording of the samples surfaces temperature evolution after the flash heating is ensured by an infrared camera (FLIR SC7000 – 2.5 to 5.5 µm). The front face corresponds to the outer surface of the tubes, and is directly available with the IR camera, whereas the rear face (inner surface) is made accessible by the insertion of a 45° gold mirror, as seen in Figure 3:

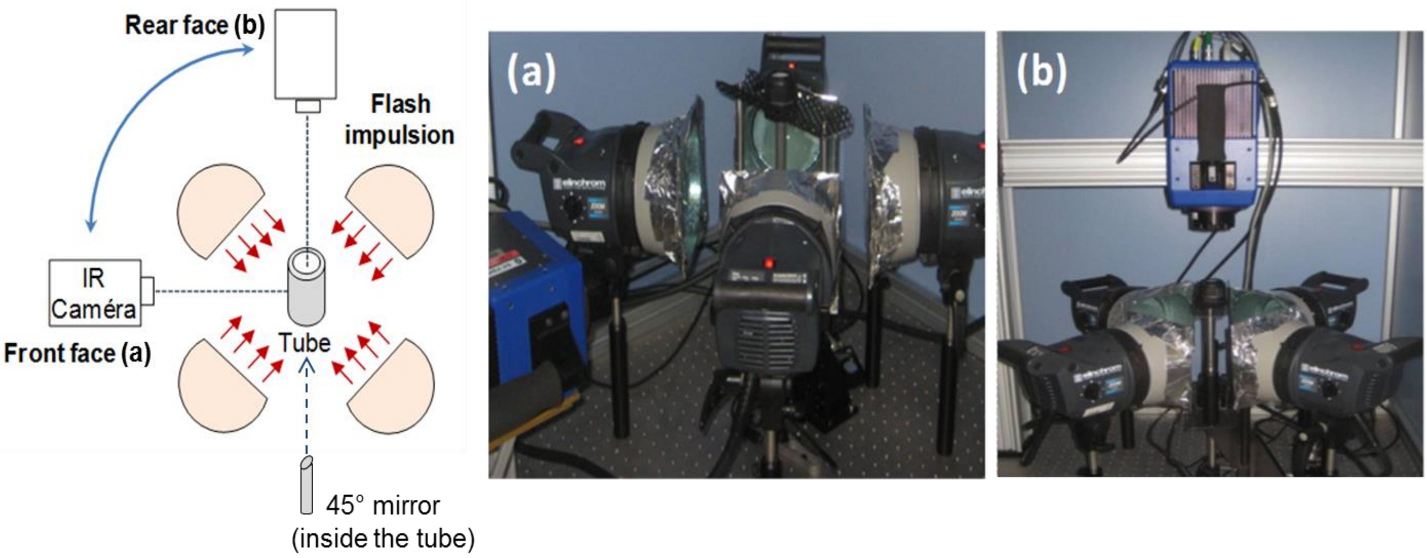

Figure 3: Experimental set-up for front face (a) and rear face (b) monitoring.

The validation of the experimental method is performed on a tubular monolithic SiC sample. It allows the verification of the homogeneity of the flash all around the tube, and the accuracy of the estimation model. A minimization of errors with respect to recorded signals by the front and rear faces analytical solution (Eq.3 & Eq.4) of the heat equation (Eq. 1) after one pulse heating is performed, yielding an identification of the thermal diffusivity (Figure 4):

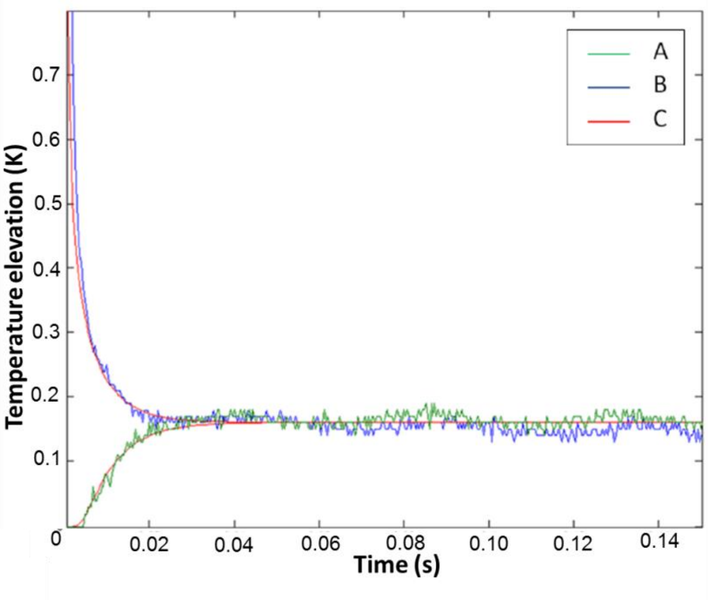

Figure 4: Plots of recorded rear face (A) and front face (B) temperature vs. time, and of the fitted associated mathematical models (C)

The flash method is performed every 10°, by rotating the inner gold mirror inside the tube to obtain the rear face signal, and by rotating the tube to obtain the front face signal. Results of the estimations are summarized in Figure 5. The average error between estimations and supplier's data is about 1.3% and the experimental bench is considered as accurate enough to be used on SiC$_f$/SiC with confidence in the results.

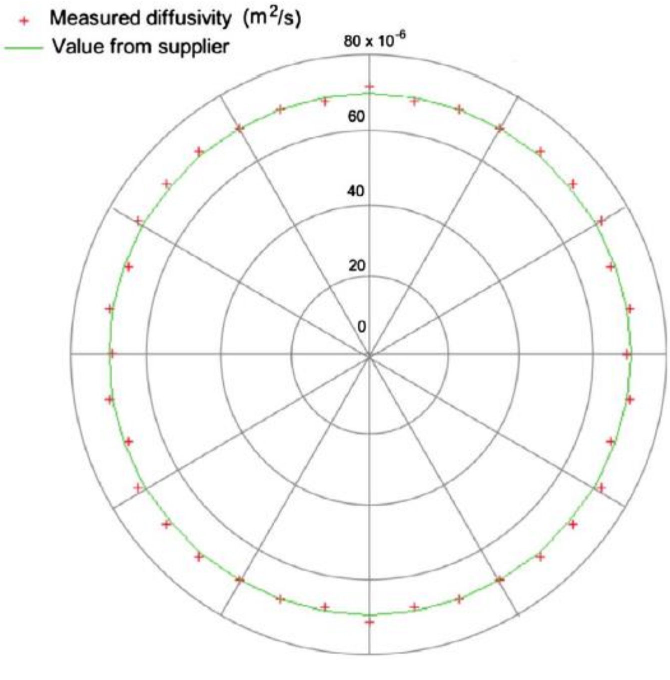

Figure 5: Estimation of the all-around through-thickness thermal diffusivity of a monolithic SiC sample versus supplier's value [4]

2.4. Adaptation on an axial tensile test device

In order analyze the evolution of thermal properties with mechanical damage, the apparatus depicted in Figure 3 has been adapted to a uniaxial tensile test machine. This set-up is an adaptation of the usual experimental device for flat samples, where front and rear face analyses are directly available [5]. The mechanical tensile device adaptation for tubular samples is described in [6]. Since the inner surface is no longer optically reachable (due to congestion), only front face analysis can be performed. Metal jaws are thermally isolated thanks to an adhesive bonding that was used to bind them to the tube. Given the space left, two lamps instead of four were used, involving an additional measurement error of 3% [7], as seen in Figure 6:

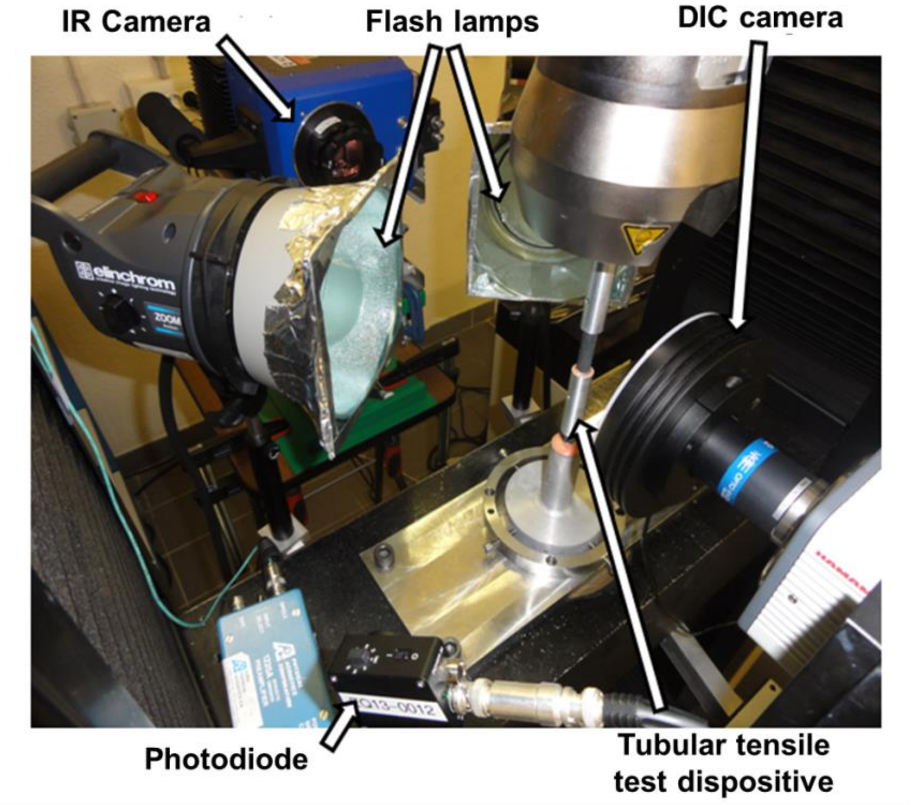

Figure 6: Experimental set-up used to assess the thermal properties evolution with intentional mechanical damage

3. Results

The experimental apparatus for the thermal diffusivity estimation has been used to perform flash experiments on SiC_f/SiC tubular samples under various conditions. First, it has been used to evaluate the evolution of the thermal behavior of samples after exposure to representative PWR (unirradiated) conditions in an autoclave. Second, it has been used to study the correlation between mechanical damage and thermal properties evolution.

3.1. Evolution of the thermal properties with PWR environment

CEA's SiC_f/SiC tubular samples have been corroded in a Framatome autoclave (Le Creusot Technical center, France) under representative PWR conditions (temperature, pressure and chemistry; no irradiation). Flash analyses have been performed at room temperature (25°C) after different exposure times (30, 80 & 110 days). Thermal conductivity is deduced from the thermal diffusivity through the specific heat (680 $J.kg^{-1}.K^{-1}$, [8]) and the density (2823 $kg.m^{-3}$, measured). Results of the estimations are summarized in Table 1.

Table 1: Thermal properties estimations after PWR operating conditions exposure (unirradiated)

Time of exposure	Initial	30 days	80 days	110 days
Thermal diffusivity ($mm^2.s^{-1}$)	15.09 ± 1.03	14.84 ± 0.74	14.92 ± 1.02	14.77 ± 0.26
Thermal conductivity ($W.m^{-1}.K^{-1}$)	28.97 ± 1.98	28.50 ± 1.42	28.64 ± 2.07	28.35 ± 0.50

After a PWR representative exposure of 110 days (out-of-pile), no significant fluctuation or degradation of the thermal properties of SiC_f/SiC tubular samples are measured. The thermal conductivity remains stable around a value of 28.5 $W.m^{-1}.K^{-1}$.

3.2. Evolution of the thermal properties with mechanical damage

CEA's SiC_f/SiC sample has been inserted in the LCTS tensile tests device. The device remains mechanically stable during every load and discharge step of the cycled tensile tests, so that a flash analysis can be performed. This arrangement allows a correlation between thermal diffusivity evolution and mechanical loading, which is described in Figure 7. The directions of the arrows indicate the progress in the hysteresis cycles of the test until failure (at a strain value of 0.7%).

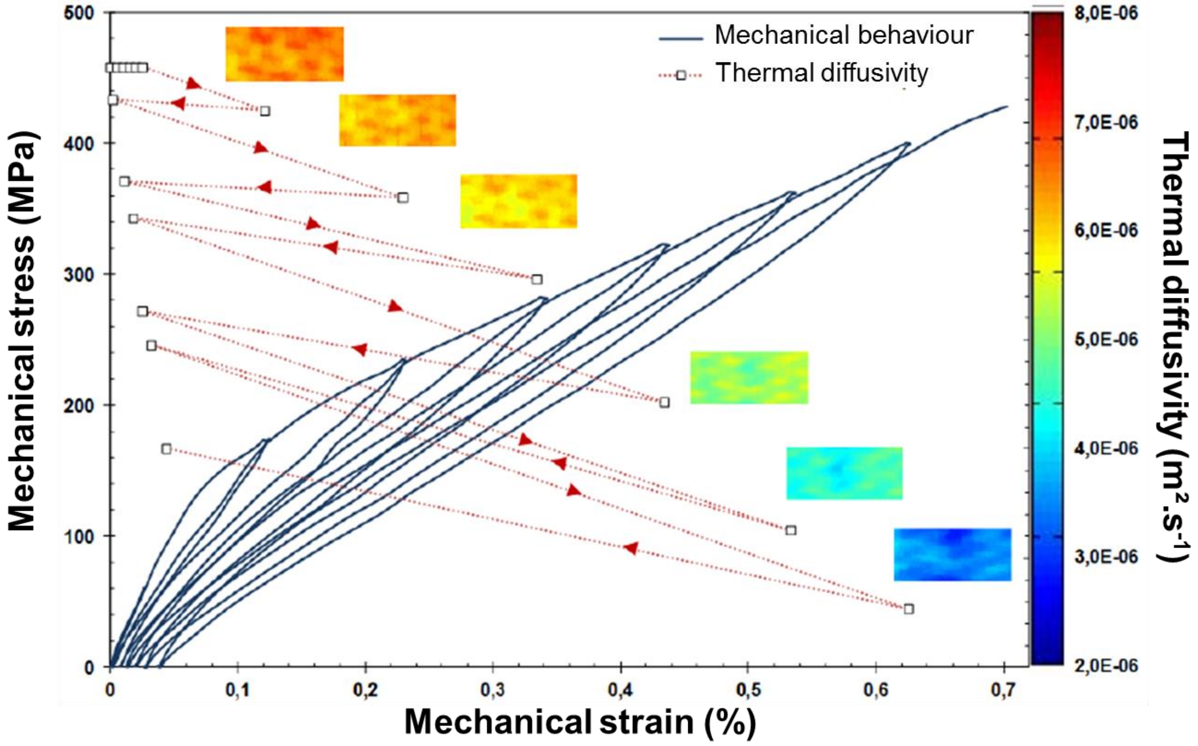

Figure 7: Through-thickness thermal diffusivity evolution of a SiC_f/SiC tubular sample under intentional mechanical damage

Through the elastic domain, there is no crack formation within the material. Therefore, the thermal diffusivity remains stable, which is characterized by a plateau of values at low strains. Then, increasing the mechanical stress leads to the formation of the first cracks that do not close up. These cracks form air gaps in the material and act as thermal barriers (Figure 8).

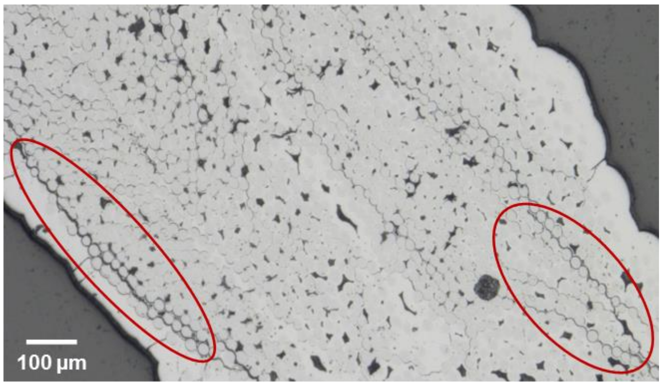

Figure 8: Thermal barriers formed by micro-cracks within the SiC_f/SiC tubular sample after mechanical damage

The thermal diffusivity decreases gradually with the increase in mechanical stress, since the cracks are continuously enlarged. The thermal diffusivity does not return to its initial value after each hysteresis cycle, because of the micro-cracks that do not entirely close up during the relaxation phase. The decrease reaches up to 70% of the initial value for the deformation levels close to breaking. The thermal diffusivity of SiC_f/SiC tubular samples is therefore highly correlated to the amount of micro-cracks, and consequently to the imposed mechanical damage.

4. Conclusions

An experimental apparatus for the thermal characterization suitable for tubular geometries has been developed and adapted to a tensile testing machine. Therefore, thermal properties have been estimated under two types of conditions. It has been proven that the PWR representative exposure had no influence on through-thickness heat diffusivity (out-of-pile). By contrast, this quantity is highly dependent to the mechanical damage, and decreased by as much as 70% close to failure.

The study of thermal properties evolution of the SiC_f/SiC cladding samples under various environments (including irradiation) is necessary since it allows a better understanding of their thermomechanical behavior. The given suitable experimental apparatus is highly valuable, as it gives a piece of compulsory information to be provided for the ATF irradiation program. This approach should be continued and improved in order to characterize irradiated samples.

5. References

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