Preparation of Cesium (Cs) Release Test for Verifying Diffusion Coefficient in the IG-110 Graphite Matrix

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1. Introduction

The high temperature gas-cooled reactor (HTGR), which is kind of 4th generation type reactor, consists of the graphite matrix with tristructural-isotropic (TRISO) coated particle fuel. TRISO particles are composed of several coating layers, and the coating layers prevent fission products from their release to the environment. But if fission products are released out from TRISO due to cracked regions or diffusion through intact coating layers, the fission products would be retained in the graphite matrix. Most of fission products are expected to be retained in the graphite matrix, but some of the fission products such as cesium (Cs), silver (Ag), and strontium (Sr) have a possibility of releasing out to coolant channels [1-3]. For estimating how the fission products are diffused through the graphite matrix and released out, the diffusion behavior of fission products in graphite is important.

In the previous studies [4-6], there are the preliminary studies for performing diffusion test: check the results of diffusion test in the literature, prepare the samples for using diffusion test, analyze the prepared samples, and so on. In order to prepare samples, the IG-110 graphite pellets are impregnated by CsNO3 solution. After impregnation steps, the IG-110 pellets are annealed at 1100°C for converting CsNO3 to Cs metal and distributing Cs in the sample. The analyses after preparing samples have performed to compare before and after release test. Prepared samples are analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) to quantify Cs concentration in graphite. The concentration gradients of prepared samples are measured by Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). The results are about the saturated Cs concentration [4-5] when the impregnation time, CsNO3 solution concentration, and heat treatment time are above threshold conditions, and the uniform concentration is discovered along both radial direction and axial direction [6].

In this study, the prepared samples have been treated second annealing to release Cs. Cs concentration in graphite after annealing, Cs concentration gradient in graphite after annealing, and released Cs quantities are verified after second annealing. Furthermore, theoretical approach is used to analyze the data. The approach includes concentration gradient after annealing, release rate, and cumulative fractional release.

2. Prepared sample analysis (Previous study overview)

The prepared samples are made for simulating graphite which includes Cs inside it. The samples have analyzed by using ICP-MS and LA-ICP-MS to verify Cs concentration and concentration gradient in graphite respectively. The same work will be performed after second annealing to compare Cs concentration and their gradient in samples before and after annealing.

2.1 ICP-MS measurement with prepared samples

ICP-MS is a good solution to analyze trace elements up to ppb level. That is the reason why the Cs quantities are analyzed by using ICP-MS in this experiment. The tests have been performed in KAIST Analysis Center for Research Advancement (KARA).

The samples to use ICP-MS have to be prepared as liquid form. Therefore, the graphite samples should be pre-treated to change liquid form. In this process, Cs is just dissolved out because graphite cannot be dissolved in the acid solution, so the graphite samples have been ground to fine powder by using sharpener and mortar. The powders is treated 70% HNO₃ 7ml + 35% HCl 3ml solution at 200°C during 30 minutes.

The measured concentration is in the range of 146-652 ppm. The data will be discussed in Chapter 3.

2.2 LA-ICP-MS measurement with prepared samples

LA-ICP-MS equipment consists of LA equipment and ICP-MS equipment, and LA equipment is installed on ICP-MS. The merits of LA-ICP-MS is what solid sample can be directly used without pre-treatment. Furthermore, the concentration gradient can be checked by LA-ICP-MS because it is available local region analysis.

LA-ICP-MS measurement has been performed in both Korea Institute of Science and Technology (KIST) and Korea Atomic Energy Research Institute (KAERI). The samples have cut as a way which can show both radial and axial direction concentration gradient simultaneously.





Figure 1. The LA-ICP-MS measurement results (line scanning). Axial (top) and radial (bottom) direction Cs distribution is almost uniform. The sample has prepared in the same condition with release test.

LA-ICP-MS measurement has performed with changing sample preparation condition, and the conclusion is what most of the samples have shown uniform Cs distribution. The representative data, which is the same sample as what the condition have been using to prepare the release test samples, is shown in Fig. 1. Axial and radial direction data show relative standard deviation 9.7% and 9.5% respectively. In conclusion, the sample contains uniform Cs distribution.

3. The relationship between ICP-MS measurement and powder size

The ICP-MS measurement results are reported in previous publications [4-6], and the concentration range is 146-652 ppm. But the maximum value is over four times higher than the minimum value, and it cannot be easily believed resulting from data error. Graphite don't be dissolved in acid pre-treatment of ICP-MS, as mentioned in Chapter 2.1, Cs is only dissolved out from powder which has been ground from bulk samples. But there is an expectation that Cs which is contained deeply in the powder cannot be solved out enough if the particle size is large because of long diffusion path. Therefore, the verification of relationship between particle size and ICP-MS measurement have been performed because the process can be affected by powder size.

3.1 Test procedure

A sharpener or mortar have been used to make powders. The size of particle which is ground by sharpener or mortar should be verified first, so the powder which does not contain Cs is analyzed by particle size analyzer. It is located on KAIST Energy & Environment Research Center (EERC). The sieves have been prepared to separate powders by their size based on particle size analysis results. The powders have been prepared by a specimen which contains Cs, and the powders have been separated with sieves by size. The prepared powders will be analyzed by ICP-MS.

3.2 Powder size distribution

The particle size analysis is a wet method, and ethanol is used to as solvent. The results of the analysis are shown in Fig. 2.

The powders prepared by using sharpener are distributed from 3 μ m to 260 μ m. The powders which are around 20 μ m show maximum volume fraction. On the other hand, the powders prepared by using mortar are distributed from 2 μ m to 700 μ m. The powders which are around 160 μ m show the maximum volume fraction. In conclusion, the powders prepared by using mortar are finer than the powders prepared by using the sharpener.

The ICP-MS results until now are identified that Cs concentration is 194-652 ppm when the powders are prepared by sharpening file, while Cs concentration is 146-262ppm when the powders are prepared by mortar. The ICP-MS results by sharpening file show relatively higher values than the results by mortar. In turn, the hypothesis can be obtained; if powder size is smaller, ICP-MS measurement values are higher. For verifying this hypothesis, ICP-MS result should be checked after classifying powders by size.



Figure 2. Particle size distribution of powders. The powders are prepared by using sharpening file (top) and mortar (bottom)

3.3 Data comparison with other studies

First of all, the Cs concentration in IG-110 after sample preparation have been reported in previous papers [4-6], 206-271 ppm from Carter's work [7]. But the results of this study have shown frequently higher concentration than Carter's work although the sample preparation condition is similar. When the particle size is larger, Cs inside the graphite may not be dissolved out perfectly because the large particle has a longer diffusion path. If so, the hypothesis mentioned in Chapter 2 has a critical contradiction with this Carter's result; the concentration in this study should be less than Carter's work when ICP-MS measurement depends on the powder size and Cs is saturated to a Cs concentration because of their solubility. Even if there is the error and scattering because Carter has measured Cs quantities by using Instrumental Neutron Activation Analysis (INAA), the result cannot be explained. Therefore, Cs concentration is calculated again in Carter's work. They have prepared 4 mm diameter sphere first, and ground to 3 mm diameter sphere. But the volume of sphere has been calculated as 4 mm diameter, instead of 3 mm diameter. The wrong result has to be corrected to 488-643 ppm, instead of 206-271

ppm. In conclusion, Cs in IG-110 matrix was 488-643 ppm in Carter's work. The result can be agreed with our ICP-MS results.

As a result, the verification of the hypotheses are needed: Cs is saturated with proper Cs quantities and ICP-MS results show a condition which uses a powder smaller than a threshold size. When the hypotheses are correct and the small powders are used, ICP-MS result should be in the range of 488-643 ppm.

4. Release test

4.1 Release test apparatus

The samples which have been prepared at verified condition are treated as second annealing for releasing Cs from graphite. The release test data will be used to verify diffusion coefficient of Cs in IG-110 graphite. The system, as shown in figure 3, for performing diffusion test consists of tube furnace and other subcomponents. Firstly, fly ash filters are used to prevent contamination of the environment, and Cs release quantities are estimated by using them. The small tube is installed inside of the original tube for reducing temperature variation, reducing contaminated part, and increasing Cs capture ability of fly ash. The atmosphere is Ar gas flow.



Figure 3. The diffusion test apparatus system. A picture of real apparatus (top) and the schematic (bottom)

4.2 Test details

The test includes temperature stabilization stage, rapid heating stage, and rapid cooling stage to reduce the errors. The rapid heating is achieved by pushing the specimen into the small tube after the temperature is stabilized. On the other hand, rapid cooling is obtained by removing the small tube, which contains the specimen, entirely from the furnace.

Temperature calibration between furnace read temperature and actual temperature inside tube have performed because diffusion is very sensitive to changing temperature. A thermocouple is inserted through gas outlet side, and read temperature is compared with tube furnace temperature. Temperature variation with time is also recorded to check temperature stabilization of the system. The test methods are organized based on this results.

For comparing the results with other research, the temperature is set on 1100K, 1200K, and 1300K. The initial Cs quantities and fractional release with time variation are reported in Carter's work [7]. The expected release quantities have been calculated based on Carter's work, and the test matrix is organized as Table 1. The test matrix is considered how much of Cs will be released out because each test set should have proper change of Cs quantities.

Table 1. Test matrix based on Carter's data

Temperature	Time set				
1027°C	30min.	1hour	3hours		
927°C	30min.	1hour	3hours	6hours	
827°C	30min.	1hour	3hours	6hours	24hours

In this experiment, the results will be analyzed by using three sources: remained Cs quantities, released Cs quantities, and Cs distribution in graphite after annealing. The theoretical approach is needed to analyze the data, and that will be introduced in next chapter.

4.3 Theoretical study

The sample which is used in this test is a pellet shape having radius R and height H. The cylindrical coordinate is used as shown in Fig. 4. The general diffusion equation is equation (1) [9] in the cylindrical coordinate.

$$\frac{\partial C}{\partial t} = D\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 C}{\partial \theta^2} + \frac{\partial^2 C}{\partial z^2}\right\} (1)$$

The concentration at the surface of the sample is always zero, and the concentration inside of the sample is finite. And the initial concentration is uniform when the time is zero. With these conditions, the boundary condition and initial condition are defined as following.

Boundary Condition (B.C.)

1. C=0 at z=0, H (with $t \ge 0$) 2. C=finite at r=0 (with $t \ge 0$) 3. C=0 at r=R (with $t \ge 0$)

Initial Condition (I.C.) C=C₀ at 0 < r < R, 0 < z < H at t=0



Figure 4. Cylindrical coordinate [8]

Cs concentration distribution is assumed as same along the radial direction, and that means there is no angular parameter.

$$\frac{\partial C}{\partial \theta} = \mathbf{0}$$
(2)

When the equation (1) and (2) is combined,

$$\frac{\partial C}{\partial t} = D\{\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C}{\partial r}\right) + \frac{\partial^2 C}{\partial z^2}\} (3)$$

Equation (3) is solved by using separation of variables,

$$C(\mathbf{r},\mathbf{z},\mathbf{t}) = R(r)Z(z)T(t)_{(4)}$$

$$RZT' = D\left(\frac{1}{r}\frac{\partial}{\partial r}(rR'ZT) + RZ''T\right)$$
$$= D\left(\frac{1}{r}(R'ZT + rR''ZT) + RZ''T\right)$$
$$= D\left(\frac{R'ZT}{r} + R''ZT + RZ''T\right)$$
(5)

$$\frac{T'}{T} = D(\frac{R'}{rR} + \frac{R''}{R} + \frac{Z''}{Z})_{(6)}$$

$$\frac{Z''}{Z} = \frac{R'}{rR} + \frac{R''}{R} - \frac{T'}{DT} = -\alpha^{2}_{(7)}$$

$$\frac{R'}{rR} + \frac{R''}{R} = \frac{T'}{DT} - \alpha^{2} = -\beta^{2}_{(8)}$$

$$Z'' = -\alpha^{2} Z \quad (9)$$

$$R'' + \frac{R'}{r} + \beta^{2} R = 0 \quad (10)$$

$$\frac{T'}{DT} = \alpha^{2} - \beta^{2} \quad (11)$$

 $Z = C_1 \cos(\alpha z) + C_2 \sin(\alpha z) \quad (12)$ $R = C_3 J_0(\beta r) + C_4 Y_0(\beta r) \quad (13)$ $T = C_5 \exp((\alpha^2 - \beta^2) Dt) \quad (14)$

Equation (12)-(14) are calculated by using boundary conditions and initial condition.

i)
$$C(r, 0, t) = C(r, H, t) = 0$$

 $C_1 = 0_{(15)}$
 $C_2 \sin(\alpha H) = 0_{(16)}$
 $\alpha H = n\pi, \alpha = \frac{n\pi}{H} (17)$
 $Z(z) = C_2 \sin(\frac{n\pi}{H}z)_{(18)}$
ii) $C(0, z, t) = \text{finite}$
 $C_4 = 0_{(19)}$
iii) $C(R, z, t) = 0$
 $C_3 J_0(\beta R) = 0_{(20)}$
 $J_0(\beta R) = 0_{(21)}$
 $R(r) = C_3 J_0(\beta r) (22) [\text{at } J_0(\beta R) = 0]$
iv) $C(r, z, 0) = C_0$
 $C_{nm} = A_{nm} \sin(\alpha_n z) J_0(\beta_m r) \exp((\alpha_n^2 - \beta_m^2)Dt)$
... (23)
when α_n and β_m are nth and mth α and β , respectively.

$$C(r,z,t) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \sin(\alpha_n z) J_0(\beta_m r) \exp((\alpha_n^2 - \beta_m^2) Dt)$$

...(24)
$$C(r,z,0) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \sin(\alpha_n z) J_0(\beta_m r) = C_0$$

...(25)

Fourier-Bessel and Fourier series formula are used to calculate $A_{\text{nm}}\text{,}$

$$A_{nm} = \frac{2}{H} \frac{2}{R^2 J_1^2(\beta_m R)} \int_0^H \int_0^R C_0 \sin(\alpha_n z) \, r J_0(\beta_m r) dr dz$$
(26)

Therefore, the calculation result by using separation of variables with B.C. and I.C. is like equation (27).

$$C(r,z,t) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \sin(\alpha_n z) J_0(\beta_m r) \exp((\alpha_n^2 - \beta_m^2) Dt)$$
(27)

Where

$$A_{nm} = \frac{4}{HR^2 J_1^2(\beta_m R)} \int_0^H \int_0^R C_0 \sin(\alpha_n z) r J_0(\beta_m r) dr dz \quad (28)$$

$$\alpha_n = \frac{n\pi}{H} \quad (29)$$

$$J_0(\beta_m R) = 0 \quad (30)$$

Release rate at a specific time can be obtained by using concentration profile, the basic formula is like equation (31).

$$\dot{m}(t) = -D(2\pi R \int_0^H \frac{\partial}{\partial r} C(r, z, t)|_{r=R} dz + 2 \int_0^R 2\pi r \frac{\partial}{\partial z} C(r, z, t)|_{z=0} dr$$
...(31)

Integral form is calculated first,

i)
$$\int_{0}^{H} \frac{\partial}{\partial r} C(r, z, t)|_{r=R} dz$$
$$\int_{0}^{H} \frac{\partial}{\partial r} C(r, z, t)|_{r=R} dz$$
$$= \int_{0}^{H} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \sin(\alpha_{n} z) \left[\frac{\partial}{\partial r} J_{0}(\beta_{m} r) \right]_{r=R} \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt) dz$$
$$= \int_{0}^{H} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \sin(\alpha_{n} z) \{-\beta_{m} J_{1}(\beta_{m} R)\} \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt) dz$$
$$= \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \left\{ \int_{0}^{H} \sin(\alpha_{n} z) dz \right\} \{-\beta_{m} J_{1}(\beta_{m} R)\} \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt)$$
... (32)

$$\left\{\int_0^H \sin(\alpha_n z) \, dz\right\} = \left\{-\frac{1}{\alpha_n} \cos \alpha_n z\right\}_0^H = -\frac{1}{\alpha_n} \left\{\cos \alpha_n H - 1\right\}$$
...(33)

$$\int_{0}^{H} \frac{\partial}{\partial r} C(r, z, t)|_{r=R} dz$$

= $\sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} [\frac{\beta_m}{\alpha_n} \{\cos \alpha_n H - 1\}] \{J_1(\beta_m R)\} exp((\alpha_n^2 - \beta_m^2) Dt)$
...(34)

ii)
$$\int_{0}^{R} r \frac{\partial}{\partial z} C(r, z, t)|_{z=0} dr$$
$$\int_{0}^{R} r \frac{\partial}{\partial z} C(r, z, t)|_{z=0} dr$$
$$= \int_{0}^{R} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \{\frac{\partial}{\partial z} \sin(\alpha_{n} z)\}_{z=0} r J_{0}(\beta_{m} r) \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt) dr$$

$$= \int_{0}^{R} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \alpha_{n} r J_{0}(\beta_{m} r) \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt) dr$$
$$= \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \alpha_{n} \{ \int_{0}^{R} r J_{0}(\beta_{m} r) dr \} \exp((\alpha_{n}^{2} - \beta_{m}^{2}) Dt)$$
...(35)

$$\int_{0}^{R} r J_{0}(\beta_{m}r) dr = \{\frac{1}{\beta_{m}} r J_{1}(\beta_{m}r)\}_{0}^{R} = \frac{1}{\beta_{m}} R J_{1}(\beta_{m}R)$$
...(36)

$$\int_{0}^{R} r \frac{\partial}{\partial z} C(r, z, t)|_{z=0} dr$$

= $\sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \frac{\alpha_n}{\beta_m} R J_1(\beta_m R) \exp((\alpha_n^2 - \beta_m^2) D t)$
...(37)

Finally, the mass release rate is following,

$$\dot{m}(t) = -2\pi RD\left(\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}A_{nm}J_1(\beta_m R)\left(\frac{\beta_m}{\alpha_n}(\cos(\alpha_n H) - 1) + \frac{2\alpha_n}{\beta_m}\right)exp((\alpha_n^2 - \beta_m^2)Dt)\right)$$
...(38)

Cumulative fractional release also can be obtained by equation (39)

$$F(t) = \frac{1}{m_0} \int_0^t \dot{m}(t') dt^n$$

= $-\frac{2\pi RD}{m_0} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} J_1(\beta_m R) (\frac{\beta_m}{\alpha_n} (\cos(\alpha_n H) - 1) + \frac{2\alpha_n}{\beta_m}) \int_0^t \exp((\alpha_n^2 - \beta_m^2) Dt') dt$
...(39)
$$\int_0^t \exp((\alpha_n^2 - \beta_m^2) Dt') dt'$$

= $[\frac{1}{(\alpha_n^2 - \beta_m^2) D} \exp(\alpha_n^2 - \beta_m^2) Dt']_0^t$
= $\frac{1}{(\alpha_n^2 - \beta_m^2) D} \{\exp(\alpha_n^2 - \beta_m^2) Dt - 1\}$ (40)

The initial concentration is assumed as uniform, so the initial mass is following,

$$\mathbf{m}_0 = C_0 \pi R^2 H \, (41)$$

The final form of cumulative fractional release is equation (42).

$$F(t) = -\frac{2}{C_0 R H} \left(\sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{A_{nm}}{\alpha_n^2 - \beta_m^2} J_1(\beta_m R) \left(\frac{\beta_m}{\alpha_n} (\cos(\alpha_n H) - 1) + \frac{2\alpha_n}{\beta_m} \right) (exp\left(\left(\alpha_n^2 - \beta_m^2 \right) Dt \right) - 1) \right) \cdots (42)$$

5. Conclusions

Obtaining the Cs diffusion coefficient is a purpose of this work. The preparation of samples, analysis of prepared samples have been performed in the previous studies. The relationship between the particle size and ICP-MS measurement have been verified. The trend is observed that the ICP-MS measurements with smaller powders shows higher concentrations. The ICP-MS measurements by using powders classified by size will verify this hypothesis. Based on the data in the previous studies [4-6], the prepared samples will be performed high temperature release test. The remained Cs quantities, the released Cs quantities, and remained Cs distribution will be used to analyze diffusion coefficient. For this experiment, the new system which includes fly ash filter has been prepared, and test plan has been organized based on Carter's data [7].

The diffusion equations have been solved to analyze the data theoretically. The Cs distribution and cumulative fractional release quantities are defined by the following equations.

i) Cs distribution

$$C(r,z,t) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \Lambda_{nm} \sin(\alpha_n z) J_0(\beta_m r) \exp((\alpha_n^2 - \beta_m^2) Dt)$$

Where

$$A_{nm} = \frac{4}{HR^2} \int_{1}^{2} (\beta_m R) \int_{0}^{H} \int_{0}^{R} C_0 \sin(\alpha_n z) r J_0(\beta_m r) dr dz$$
$$\alpha_n = \frac{n\pi}{H}$$
$$J_0(\beta_m R) = 0$$

ii) Cumulative fractional release

$$\mathbf{F}(t) = -\frac{2}{C_0 R H} (\sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{A_{nm}}{\alpha_n^2 - \beta_m^2} J_1(\beta_m R) (\frac{\beta_m}{\alpha_n} (\cos(\alpha_n H) - 1) + \frac{2\alpha_n}{\beta_m}) (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) - 1) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 - \beta_m^2) D t\right) + \frac{2\alpha_n}{\beta_m} (exp\left((\alpha_n^2 -$$

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