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Tungsten coatings repair: An approach to increase the lifetime of plasma facing components

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The lifetime of tungsten coatings used for Plasma Facing Components (PFCs) in fusion devices is mainly limited by two degradation mechanisms occurring during the operation of the tokamak: erosion resulting from exposure to long plasma discharges (long term) and local damages caused by transient events (runaway electrons,...). An alternative solution to the current replacement of damaged PFCs would be to repair their surface by stripping the damaged tungsten coating and depositing a new one. In this paper, the process of removing tungsten was investigated on CuCrZr samples coated with a 15 μm layer of W using four techniques: plasma etching, sandblasting, chemical attack and laser ablation. For each technique, etching properties were assessed by means of microscopic observations, topographical surface analyses and chemical analysis. The results presented here also highlight the issues related with controlling the etching depth and size of the treated surface.

1. Introduction

Tungsten (W) coatings have received a great deal of attention as a technical solution for Plasma Facing Components (PFCs) in present day tokamaks, owing to their advantages over bulk tungsten such as a lower cost and weight [1]. However, their brittle behavior often leads to a weak adhesion bond with the substrate material. Exposure to long plasma discharges, thermal cycling and runaway electrons have been identified as the main sources of surface degradation [2], like erosion and local damages (delamination, cracks, etc.), potentially decreasing the performance of the plasma by releasing particles into it.

In European tokamaks where W coatings are used (JET, ASDEX) damaged PFCs are usually entirely replaced to avoid any contamination of the plasma and to ensure the structural integrity of the machine. However, in fusion devices like WEST, a vast majority of PFCs are actively cooled (baffle, divertor, VDE protection, heating system protection). In the present WEST configuration, such actively cooled components represent about 60% of the total PFCs surface exposed to the plasma, making their replacement more difficult and more time consuming than for inertially cooled components due to the assembly (weldings) to the cooling system and the associated control tests (leak tests). Moreover, the amount of spare components may be restricted because of the high technology associated with actively cooled components (swirl, bi materials junction, etc.) [3]. Repairing damaged

coatings seems therefore of a big interest and may represent an alternative solution to the replacement of the whole component.

In WEST, all PFCs are coated with tungsten [4,5]. The repair procedure presented in this paper was developed for actively cooled components only, which are made of CuCrZr heat sink with an about 15 μm coating layer of tungsten deposited by physical vapor deposition (PVD). As a first approach the work has focused on removing the damaged layer of tungsten so that a new layer could later be applied. For such systems, hard coatings on soft substrates, removing the top layer without damaging the substrate material can prove challenging. In addition, tungsten coatings have a relatively small thickness, a high density and a high adhesion to the substrate. The melting temperature of CuCrZr (1000 °C) is also much lower than the one of tungsten (3400 °C), adding another degree of complexity in the stripping process.

In this paper, several stripping techniques have been investigated to remove the tungsten coating from the CuCrZr substrate: plasma etching, sandblasting, chemical etching and laser ablation. These techniques were selected either for their ease of operation or for our experience and expertise developed in this field. In order to explore the possibility of repairing PFCs in the tokamak in the future, it was also important to select processes that could be implemented in situ. For each technique the ability to treat large surfaces and complex shapes was evaluated as well as the homogeneity of the process and its selectivity. This paper concludes with a discussion of the repair strategy, by presenting the

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results of ex situ and in situ etching processes as well as the future work that must be done as for depositing a new coating layer.

2. Materials

Extruded CuCrZr copper alloy (0.8 at.% Cr, 0.15 at.% Zr, balance Cu) supplied by Le Bronze Industriel was machined and used in the form of cylindrical samples 30 mm in diameter and 5 mm in height. A tungsten layer of about 15 μm was then directly applied on the top surface by physical vapor deposition (PVD), using the same experimental conditions than for WEST components. The characterization of the coating properties (performed on 3 samples) showed a density of about 90%, a thickness of 15 μm and a chemical composition of 95% W and 5% O.

3. Plasma etching

High Power Impulse Magnetron Sputtering (HIPIMS) device from DEPHIS Company (France) was used to produce ion bombardment on the top surface of the sample for 1 h at a constant frequency of 500 Hz and with a 45 μs pulse duration in argon environment. A bias voltage of -900 V was applied to the sample holder. Although several studies [6,7] used fluorine based gas to etch tungsten with relatively high etching rates, it was decided here to use Cr^+ as etching ions to avoid any contamination of the component, and consequently the tokamak, with fluorine.

Fig. 1 presents SEM optical micrographs of the tungsten surface before etching (as coated) and after plasma etching. The non treated material is seen to have a uniform microstructure with elongated particles of about 2 μm . After 1 h of plasma etching, it exhibited a rather smooth surface with non oriented particles. Their size was found to be non homogeneous, ranging from 0.5 μm to 5 μm .

The boundary region between plasma etched and non etched tungsten was analyzed by confocal microscopy, with a $5 \times 6.5\text{ mm}^2$ surface cartography shown in Fig. 1. This system was developed at CEA [8] and was used to generate a 3D map of the height of the sample surface. The effect of the plasma on the material was then evaluated by measuring the relative height levels of the etched and non etched areas

indicated on the color scale of the figure. It can be seen that the height level of the W surface decreased from $\sim 281\text{ }\mu\text{m}$ to $\sim 280\text{ }\mu\text{m}$ after 1 h treatment. This result is highlighted in the line profile plotted on the surface.

This relatively low etching rate of 1 $\mu\text{m}/\text{h}$ is believed to be associated with the high density of the coating material rather than the experimental conditions for which HIPIMS was specifically chosen due to the high pulsed pulverization energy it provides. No traces of argon and chromium were detected in the tungsten material after etching.

4. Sandblasting

W coated CuCrZr specimens were sandblasted on the W side using a microsandblast equipment Sandmaster 55S, Unitoool Ltd. Aarburg (1982) with Al_2O_3 Corindon particles (available in our institute) of about 120 μm diameter under 7.4 bar compressed air. The sandblast gun was manually maintained at a distance of few centimeters from the W surface of the specimen. The samples were then rinsed with ethanol and air dried.

Fig. 2(a) shows the W coated CuCrZr sample after sandblasting for a few minutes. The original tungsten coating can be seen on the left hand side of the sample that was not exposed to abrasive Al_2O_3 particles. On the right hand side the CuCrZr substrate is clearly apparent, indicating that the entire layer thickness of tungsten was removed by this technique. It can also be noted that the boundary line between the sand blasted and non sandblasted region is very clean and precise, making this technique suitable for the treatment of patterned local areas.

The layer thickness removed by sandblasting was evaluated by confocal microscopy as mentioned before. The cartography obtained for the entire sample is shown in Fig. 2(b). As it can be seen, the non treated region exhibited almost the same elevation across its surface area with height levels ranging from 256 μm and 259 μm . However, a clear non homogeneity was found in the sandblasted area of the sample. The central part was more etched than the outer part, with a clear variation of the elevation in the radial direction. A surface height of 199 μm was measured in the central part while height levels slowly increased to 223 μm and then to 233 μm as the rim area of the sample was approached. This corresponds to an etched thickness between

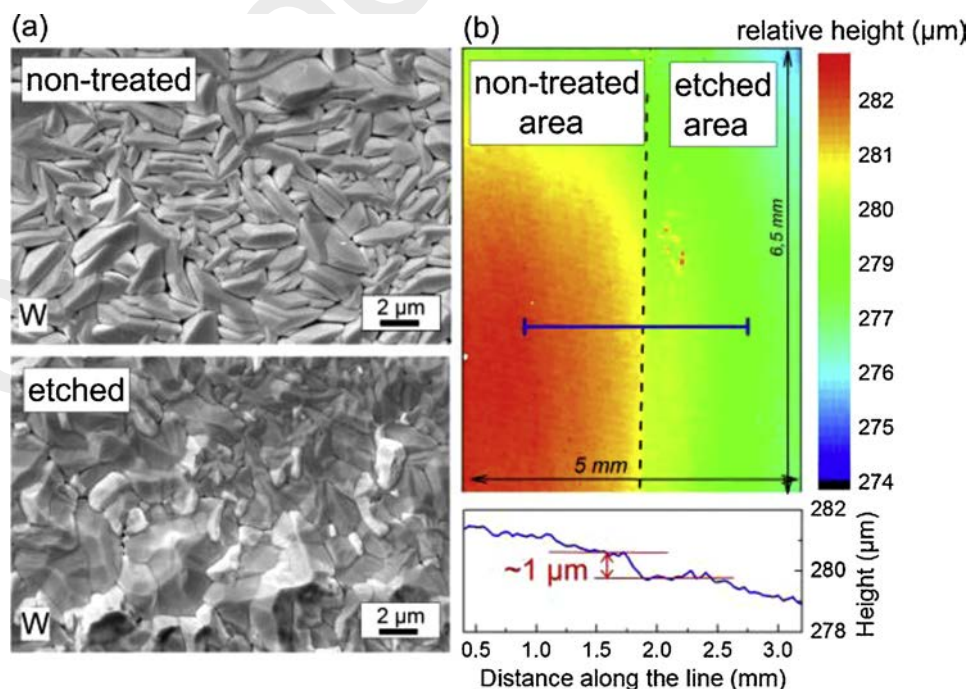


Fig. 1. (a) SEM images of tungsten structure before (non-treated) and after plasma etching for 1 h. (b) Height surface cartography of the boundary region between etched and non-etched tungsten.

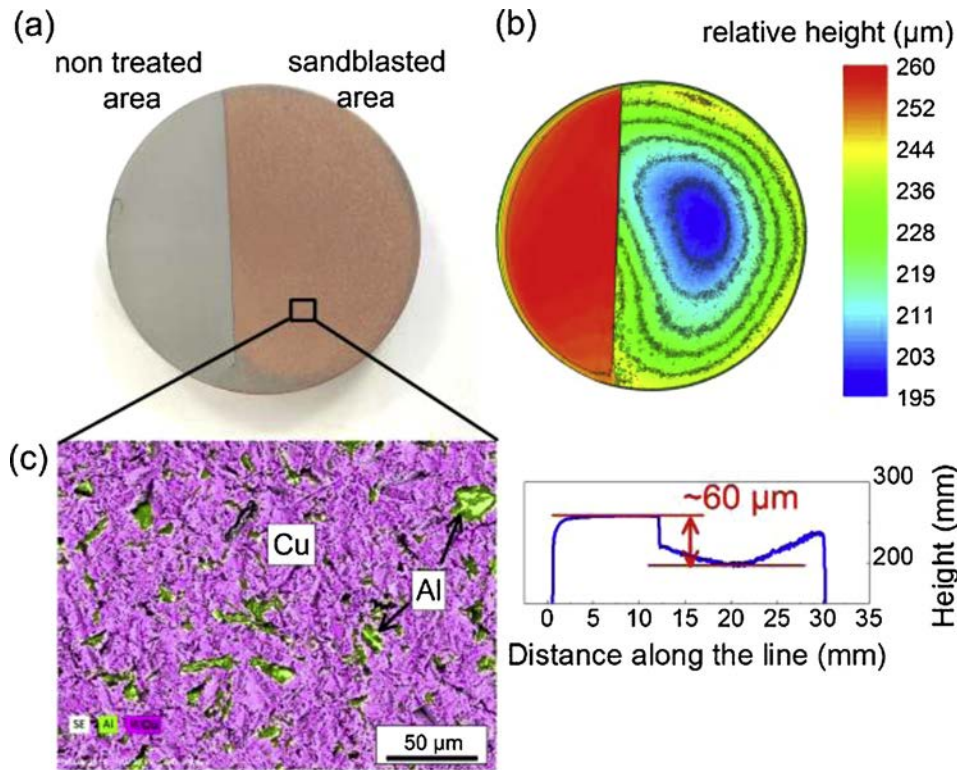


Fig. 2. (a) Image of the W coated CuCrZr sample locally etched by sandblasting and (b) the associated height levels map. (c) EDS chemical analysis of the CuCrZr after sandblasting.

20 μm and 60 μm , well above the 15–20 μm thickness of W, indicating that a part of the copper substrate was also removed during the process. This non homogeneity can be explained by the experimental setup of the process that did not allow controlling properly the distance and the angle impact of the blast stream.

Fig. 2(c) illustrates the results of a chemical analysis carried out on the apparent CuCrZr substrate after sandblasting by FESEM (Field Emission Scanning Electron Microscopes ZEISS Sigma 500 equipped with Energy Dispersive X ray Spectroscopy, Bruker). The element distribution map of the microstructure showed a rather rough surface with Al inclusion particles of up to 20 μm diameter trapped into the copper matrix. The associated composition analysis confirmed the presence of aluminum (18.8 at.%) but revealed also the presence of O (21.4 at.%) and W (1.4 at.%) that may be associated with the projection of previously ablated material. The clear presence of abrasive particles residues in the material after processing requires therefore a supplementary cleaning process which, added to the need to automate the process, makes the application of this technique difficult.

5. Chemical etching

Specimens were held in a vertical position and partially immersed in a tungsten etchant solution (Sigma Aldrich ref. 667498, tripotassium hexacyanoferrate and potassium hydroxide) for 8 h at room temperature so that the back of the sample of CuCrZr and the top surface of tungsten were etched in the same conditions. The solution was changed after 4 h to avoid any saturation of the reaction.

Chemical etching was the third etching technique investigated in this study. In addition to its ability to treat large surface areas, this process enabled to study the effect of the etchant solution on the coating material and the substrate material simultaneously. Fig. 3(a) presents optical images of the sample after treatment. In the front view picture, the part of the sample that was immersed into the liquid solution is located on the right hand side of the specimen where the

copper substrate can be seen. After 8 h treatment it was seen that the entire coating layer was dissolved, with no traces of tungsten or oxides observed on the copper surface. The left hand side of the sample was kept outside the solution and therefore did not receive any etching treatment. The middle area that appears in bright grey in the photo was not immersed at first but was immersed after renewing the solution (the solutions had different liquid levels). This region was therefore etched for 4 h only. The effect of the chemical etching process was less visible on the back side of the sample where only a slight change of color was observed, probably due to the deoxidation of copper.

Fig. 3(b) shows the surface cartographies of the two sides of the specimen. In the front side, it can be seen that the non treated area had an initial dome shape that resulted from the coating deposition process, with a height variation of about 5 μm between the center (350 μm) and the rim of the sample (345 μm). However, the same relative height of 333 μm was observed all over the surface area of the chemically etched area, indicating a maximum removed thickness of 17 μm , as shown also in the plotted height profile. The back side of the sample exhibited a rather uniform topography all over the sample surface, with only visible circumferential marks and waviness associated with the machining process. A height profile of the non etched area confirmed the strong resistance of CuCrZr to the chemical etching process with the used solution. It can be noted that the resolution of the confocal microscopy did not allow the detection the deoxydation of copper.

The results achieved here show therefore the great potential of using chemical etching for selectively removing tungsten material from CuCrZr substrate using tripotassium hexacyanoferrate and potassium hydroxide solution. Additional etching tests were then conducted with stainless steel to study the effect of the process on PFCs cooling tubes. No traces of etching were observed.

6. Laser ablation

The CORIA laboratory's Laser Induced Breakdown Spectroscopy

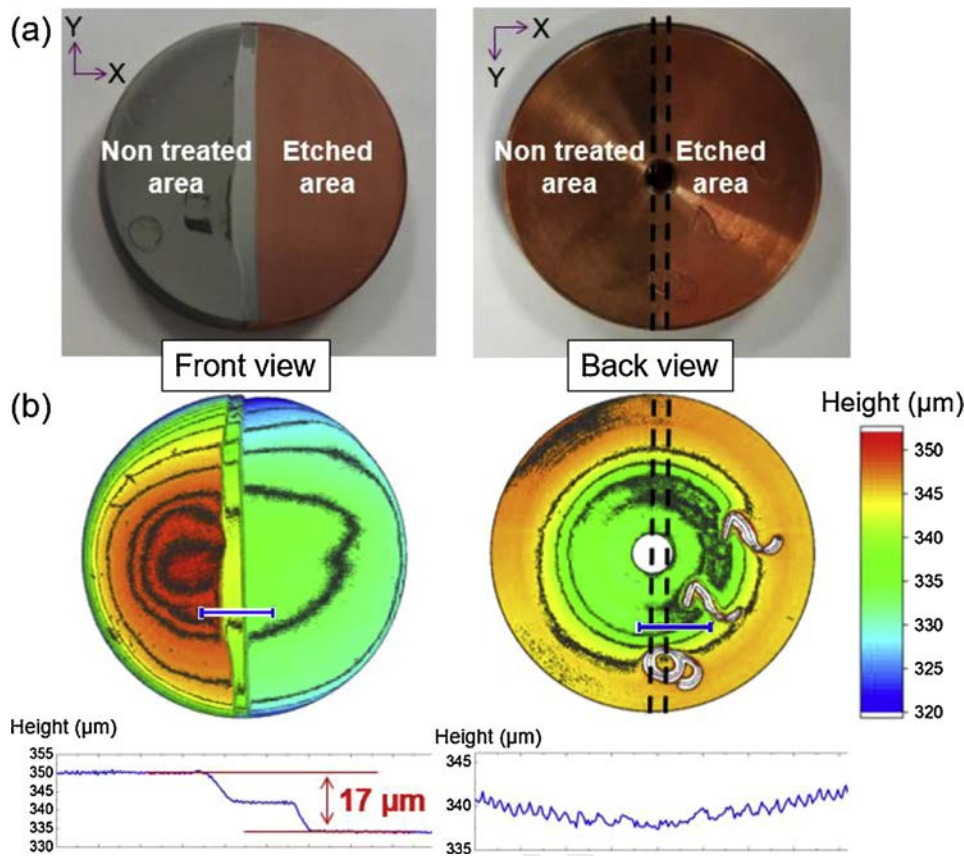


Fig. 3. (a) Front view and back view optical images of the W-coated CuCrZr sample locally etched by chemical attack for 8 h and (b) associated surface cartographies.

(LIBS) platform [9] was used to remove the tungsten coating via ablation craters. A picosecond laser pulse provided by a EKSPLA source (10 Hz, 30 ps, 532 nm, 14 mJ) was focused on the W surface using a converging lens (focal length of 40 cm) under atmospheric pressure. In order to modulate the ablation, the distance between the lens and the sample surface varied from 328 mm to 388 mm. The light emitted by the plasma resulting from the interaction between the sample and the laser pulse was analyzed by a Ebert Fastie spectrometer ($f = 2$ m, aperture of $f/15$). Finally, the obtained craters were observed with an optical microscope and their depth was measured to an accuracy of $2 \mu\text{m}$ using a focus variation method.

In order to study the ablation rate of W coated CuCrZr by laser, laser pulses were delivered to the top surface to ablate the material. Five ablation craters of 25, 50, 100, 150 and 200 pulses were produced and the ablated depth for each of them was measured, as illustrated in Fig. 4. The craters depth was found to increase almost linearly with the number of pulses, with an ablated depth of $6 \mu\text{m}$ after 25 pulses and about $16 \mu\text{m}$ after 200 pulses. This corresponds to an average ablation rate of 90 nm/pulse . It can be noted that similar experiments performed with ITER like bulk W (not shown here) lead to much higher ablation rate of 500 nm/pulse , clearly indicating that W deposited by PVD is more difficult to ablate than bulk tungsten. More experiments should be performed to better understand this surprising result.

The removal of tungsten coating in one spot was monitored all along the process by emission spectrometry within the spectral range of 503 nm 514 nm that contains lines of Cu, Cr and W elements. The detection of the W CuCrZr interface was therefore possible by following the evolution of the spectral radiance of these elements with the increasing number of pulses, as illustrated in Fig. 4. While the spectral radiance of W decreased slowly all along the ablation process, it can be seen that the spectral radiance of the substrate elements increased rapidly to reach almost 20% of the total spectral radiance after 150 pulses

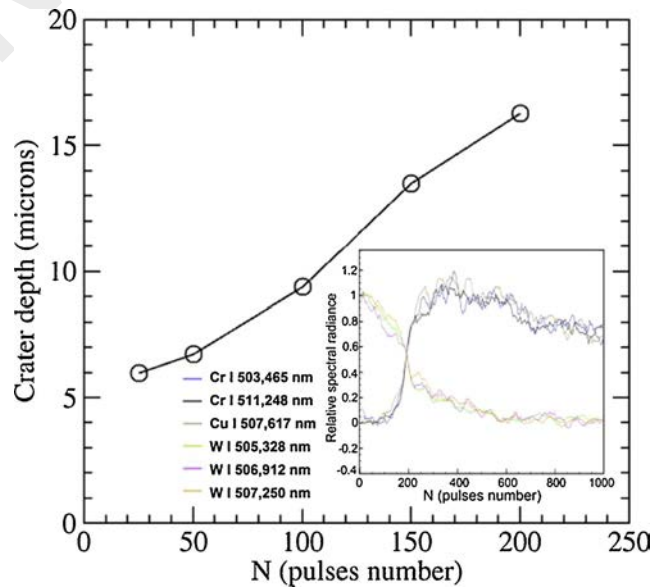


Fig. 4. Dependence of ablation depth on pulses number and evolution of the spectral radiance of Cr, Cu, W elements in the 503–514 nm range during the ablation of one spot.

and 50% after 200 pulses. This rather smooth transition between the signal associated with the coating and the substrate brings therefore a high sensibility as for the determination of the accurate location of the interface. Nevertheless, it can be safely stated that the copper substrate was reached after 200 pulses at least, which corresponds to an ablation depth of about $17 \mu\text{m}$.

In the configuration mentioned above, the lens was located at a

Table 1

Ablation craters diameter and ablation rate obtained for CuCrZr and W coating deposited by PVD according the lens-sample surface distance.

Material	Lens-sample surface distance (mm)	Ablation craters diameter (μm)	Ablation rate (nm/pulse)
CuCrZr	388	–	500
W deposited by PVD	388	200	90
	348	600	40
	328	800	13

distance of 388 mm from the sample surface and the saturated diameter of the crater was 200 μm . By approaching the lens to the surface and, consequently, by adjusting the impact surface area of the laser and its energy distribution, it was possible to modulate the ablation mode. As reported in Table 1, the diameter of the craters in the W layer deposited by PVD was found to increase up to 800 μm when the distance between the lens and the sample surface was reduced to 328 mm. The ablation rate also decreased from 90 nm/pulse to 13 nm/pulse due to the reduced ablated depth.

Although the ablation can be stopped whenever the first spectral lines of the copper substrate are detected, it was important to evaluate the ablation rate of CuCrZr. A specific sample preparation was therefore proposed to ablate the specimen from the back surface. The results showed a laser ablation rate of 500 nm/pulse, much higher than that of tungsten coating. Severe damages of the copper substrate can therefore occur if the laser removes more than the tungsten thickness, revealing therefore the importance of using spectrometry to properly detect the W/CuCrZr interface.

7. Discussion and conclusion

In summary, it was shown that chemical etching is a very good technique to selectively etch tungsten of complex shaped and large CuCrZr components without damaging any steel connection tubes. The 17 μm W layer was successfully removed after 8 h. This process is very well suited for ex situ applications. Nevertheless, the treatment of activated PFCs may require a specific recycling treatment for the effluents because of the activated residues generated in the acid solution. Sandblasting could be another ex situ technique able to remove large areas of W coatings but more experiments using smaller particles size or lower pressure should be conducted to properly evaluate its potential.

If the criteria of repairing PFCs in an in situ environment is

required, laser ablation would be the most suitable solution due to its ability to operate in a vacuum environment and the ease to integrate its technology onto a robotic arm. Laser ablation rates of tungsten deposited by PVD varied from 13 nm/pulse to 90 nm/pulse by defocusing the laser beam, with crater diameters ranging from 800 μm to 200 μm , respectively. Although these etching rates are relatively low and would allow local ablation only, a modulation of the process can be considered (coarse then fine ablation as the tungsten copper interface is approached). Nevertheless, this process must be monitored by spectrometry (LIBS) to avoid damaging the CuCrZr substrate which ablation rate was shown to be about 5 times higher than for tungsten deposited by PVD.

This study only focused on the first stage of the repair procedure for actively cooled PFCs. Our work will now aimed at fabricating and depositing a new W layer on the etched surface, using the same experimental conditions that were used in 2015 during the fabrication of WEST PFCs. The properties of the new coating will be assessed in order to make sure they stand the required specifications. In the search of a suitable technique to repair tungsten coatings, it is also essential to evaluate the necessity of removing the whole coating thickness. The deposition of a new W layer directly on top of the old one could be considered here. In that case, laser ablation or plasma etching could be used to prepare and clean the damaged surface by eliminating impurities or residues that may have come during the operation of the tokamak.

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