

Impact of water ingress on deuterium release, oxidation, and dust generation in beryllium plasma-facing components

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ABSTRACT

Beryllium samples from the JET ITER-like wall limiter tiles with either co-deposits or surface cracks caused by melt damage, were immersed into boiling water for 4 h 15 min to simulate and assess the impact of coolant water ingress into a tokamak on the state of Be components. Microscopy of the water-treated surfaces and the lack of residue in the water revealed that no thermomechanical damage (cracking or exfoliation) occurred to the samples during the exposure. Ion beam analysis showed no measurable release of deuterium from the samples. Combined ion beam analysis and Raman spectroscopy indicated only some degree of surface oxidation, but no thick oxide films were formed.

1. Introduction

All in-vessel components (IVCs) of the ITER tokamak will be actively water-cooled. Therefore, an accident involving a pipe rupture and subsequent water leak into the vessel cannot be completely excluded [1,2]. This would lead to the interaction of water at the base coolant temperature (between 70 °C (inlet) and 126 °C (outlet) in Integrated Blanket ELMs and Divertor Primary Heat Transfer System (IBED PHTS) of ITER [3]) with plasma-facing components (PFCs). Such interaction with co-deposited layers present on PFCs might lead to their fracture and delamination of co-deposits due to thermomechanical stresses, thus creating a risk of dust generation [4]. Another potential source of dust is a near-surface cracking of PFCs themselves, and/or propagation of pre-existing cracks resulting in the subsequent material disintegration into smaller fragments (debris) [5,6].

Generation of dust constitutes a manifold safety concern. In the worst-case scenario interaction of water with hot dust might lead to an explosion of the released hydrogen gas and, the consequential vessel damage [7,8]. Mobilization of tritium-containing and neutron-activated dust might then escape the damaged vessel thus posing a severe radiological hazard in the environment [9]. Even under a regular tokamak operation, dust deposition may potentially damage PFCs, e.g. dust deposition on high-heat flux components would lead to a localized heating which would require modification of an operational scenario to

avoid damage. Dust also degrades performance of in-vessel diagnostics, such as mirrors [10,11]. In addition, particles tend to accumulate in remote parts of the vessel from where it is difficult or impossible to remove them. This increases the overall tritium retention [12].

The other concern is a possibility of releasing retained tritium (T) from the PFCs, which would subsequently interact with water [13]. The formation of T-contaminated water would create serious radiological environmental hazards [14,15].

PFCs in ITER will be made of beryllium (Be) in the first wall panels and tungsten (W) in the divertor. The JET tokamak, operated since 2011 with the ITER-like wall (JET-ILW), is a uniquely suitable test bed for studies of ITER-relevant processes of plasma-wall interaction and plasma-facing materials [16]. JET-ILW uses ITER mix of plasma-facing materials, featuring bulk Be poloidal limiters, upper dump plates and other main chamber protection components, while the divertor consists of bulk W and W-coated CFC (carbon fibre components). PFCs are regularly retrieved from JET vessel during scheduled shutdowns, and subsequently cut into samples (cm size) to facilitate a variety of ex-situ analyses.

The considerations described above have provided motivation and determined methodology for the present work. The main objective is to assess the impact of boiling water on beryllium PFCs with co-deposits in the event of water ingress.

Be PFCs of ITER are mimicked by samples cut out of the castellated

¹ See list of authors of Overview of JET results for optimizing ITER operation by J. Mailloux et al, Nuclear Fusion 62 (2022) 042026.

limiter tiles from JET ILW. These included samples affected by the plasma impact: surface with co-deposits and/or melt damage. Such samples are relevant for testing of the worst-case scenario, as both co-deposits and pre-existing damage make them likely prone to cracking or delamination in the event of interaction with water. A series of experiments was conducted in which JET Be samples were immersed into the boiling water at 100 °C, this temperature being in the middle of the range expected for the coolant water in ITER [3]; subsequent analysis of these samples was focusing on changes in the surface morphology, deuterium content and oxidation state using microscopy and elemental analysis tools.

2. Experiment

The study was carried out with single castellation cut out of the wide poloidal (WPL) limiter tile 4D14 (referred to as Sample 608), and the upper dump plate (UDP) tile 3A8 (Sample 427). The tiles were retrieved from the JET vessel in 2016 after the third ILW campaign (ILW-3). Locations of the castellation in the corresponding tiles are shown in Fig. 1a–b. The limiter sample was taken from the deposition-dominated zone on the outer wing of the tile and features surface deposit. SEM inspection indicated that the appearance and surface structure of this deposit is uniform across the extent of the sample. The UDP sample originated from the region affected by melting and solidification in connection with disruptions.

The exposure to water was performed in an apparatus shown in Fig. 1c. A flask containing 250 ml of deionized water was placed in the heating mantle. A water-cooled reflux condenser was attached at the top of the flask. As water evaporates from the flask during boiling, it condenses and flows downwards along the inner tube walls back into the flask. This way, boiling can be sustained indefinitely as water is not lost to evaporation, and therefore prolonged immersion of samples in the boiling water is possible.

The experimental procedure consisted of the following steps. First water in the flask was brought to the boiling point. When the water started boiling, a single sample (kept at room temperature until this point) was dropped into the flask through one of the side ports. By

regulating power of the heating mantle, the sample was kept in the boiling liquid for 4 h 15 min, i.e. the total exposure time. Extended duration of exposure was chosen to represent a worst-case scenario and potentially generate largest possible damage. Specific time was determined by technical limitations of the used setup. Afterwards the sample was extracted from the flask and dried in air, after which post-exposure examination of this sample was performed. The water was then evaporated from the flask, and a possible residue on the bottom of the flask was collected using adhesive carbon tabs.

Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) studies were performed on the samples' surfaces before and after the experiment using a TESCAN Mira3 XMH microscope with Oxford Instruments X-Max 80 EDS detector. Adhesive tabs were also examined by these microscopy methods.

Ion beam analysis (IBA) of the Be samples was carried out using a 2.5 MV Van de Graaff accelerator at the Laboratory of Accelerators and Radiation Technologies, Lisbon, Portugal. The accelerator is equipped with a chamber dedicated to fusion research, where Be- and tritium-containing samples are handled. Rutherford backscattering (RBS) and nuclear reaction analysis (NRA) were performed using ^3He ions at an energy of 2.3 MeV in order to determine D and O concentrations in the investigated sample. NRA was based on proton and alpha-particle detection from the $\text{D}(^3\text{He},\text{p})^4\text{He}$ reaction. Measurements were done before and after the exposure to water in several locations on the samples in order to monitor the changes in D and O contents. The quantification of D and O was performed using NDF code [17].

Raman spectroscopy was performed using WiTec Alpha 300 ARS confocal Raman microscope. 532 nm excitation, 59 mW, x2.5 magnification, 10 accumulations per point with a 4 s integration time. It was used to detect possible presence of BeO on the surface of the samples, i.e. to verify whether exposure to water resulted in oxidation.

3. Results and discussion

Prior to the treatment in the boiling water, SEM imaging of the samples' surfaces was done, in order to compare the state of individual surface features before and after the exposure and study possible

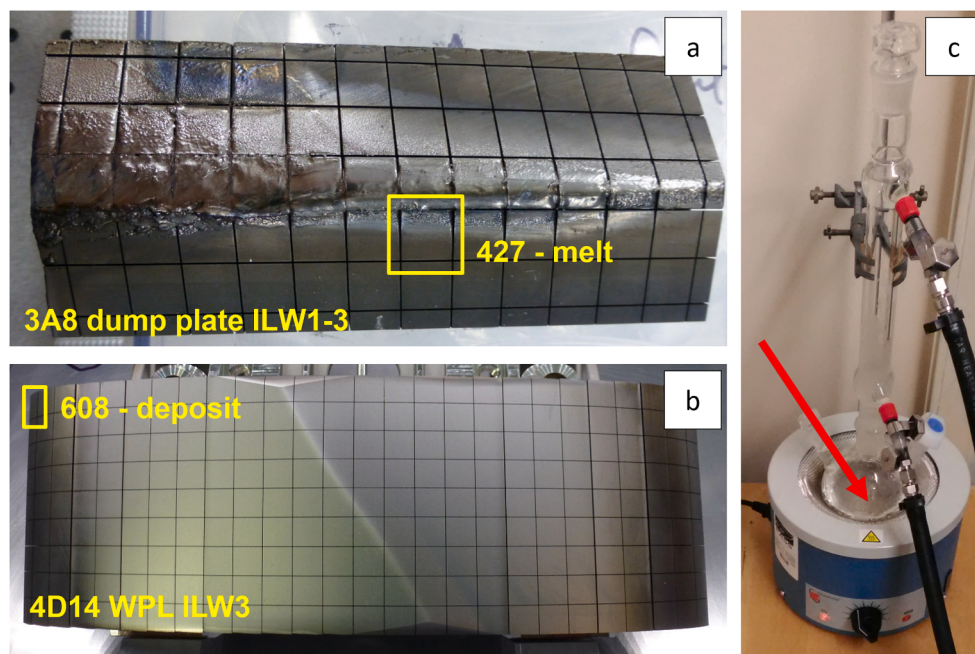


Fig. 1. Optical images of the JET tiles and locations of the castellation that were exposed to boiling water: (a) Tile 3A8 UDP (Sample 427, with melt damage and crack network); (b) Tile 4D14 WPL (Sample 608, with Be co-deposit); (c) apparatus for samples' treatment in water; the arrow indicates the location of a sample during exposure.

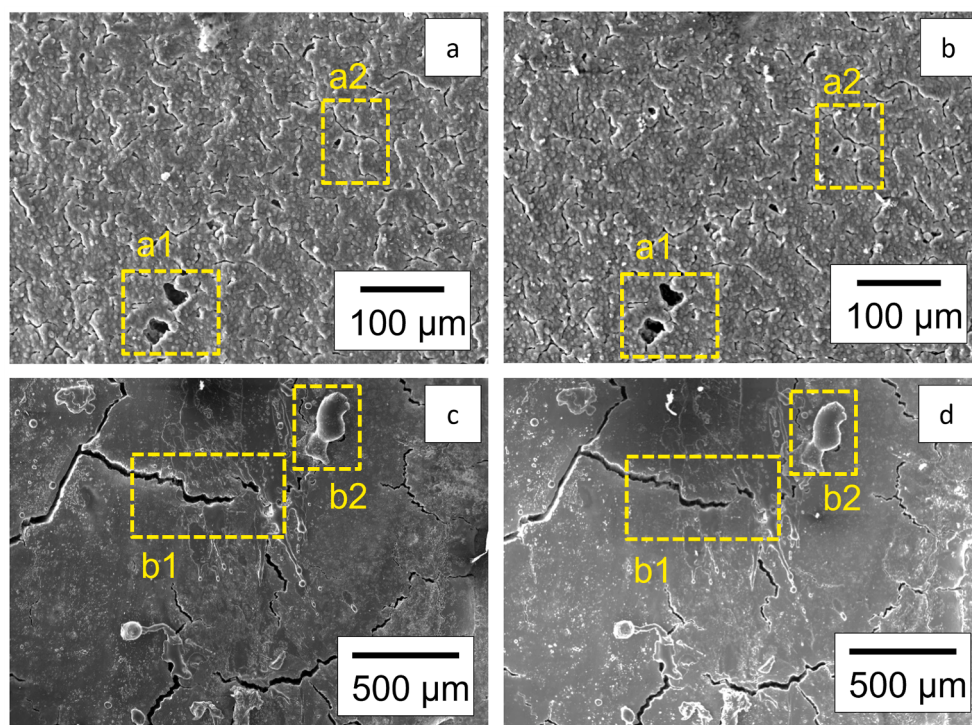


Fig. 2. SEM surface images of surfaces: (a) Sample 427 before exposure to water; (b) 427 after exposure; (c) 608 before exposure; (d) 608 after exposure. Labels a1, a2, b1, b2 denote the same features in the images taken before and after the exposure.

changes. An example of such comparison for some of these features is presented in Fig. 2 for the samples with a co-deposit and a melt damage. The same regions of the upper and side surfaces of the sample are shown, and exactly the same specific features are highlighted. It is evident that no changes have occurred on the examined surfaces as a result of exposure – neither during water immersion, nor during subsequent drying. In particular, no further development of the pre-existing cracks occurred. It can also be seen that there was no macroscopic structural damage such as cracking (i.e. no generation of new cracks in addition to pre-existing ones) or flaking and subsequent delamination of the co-deposit. The latter agrees with results of previous studies on JET limiter samples with co-deposited layers exposed to hot water and water vapour [18].

Adhesive carbon tabs were used to collect residue from the bottom of the flasks following the exposure. Three tabs were used per exposed sample to probe three different parts of the flask's internal surface. If exposure to water resulted in macroscopic flaking or delamination of surface deposits in sample 608, or cracking and exfoliation of fragments in sample 427, these particles would have remained on the flask bottom after water has been evaporated. Such residue would have been collected by the tabs. SEM imaging, coupled with EDX elemental mapping, was done on the tabs to identify any Be-containing particles that could originate from the water-exposed samples. Entire surfaces of each tab were investigated. No Be particles were found.

The observations that no damage was seen on the sample's surfaces and no delaminated Be particles were found clearly indicate that bulk Be (including Be with pre-existing pattern of cracks) and co-deposit are not susceptible to macroscopic mechanical damage when exposed to boiling water, despite inherent brittleness of Be [19]. It is a positive result from the point of view of the ITER future operation. It suggests that water ingress is not likely to cause massive formation and/or mobilization of dust from co-deposited films or bulk Be components even if they have been pre-damaged by off-normal events such as disruptions or runaway electrons.

Fig. 3 shows D and O contents measured with IBA across the samples;

approximate locations of the beam spots are shown in the insert. A comparison of results obtained in similar locations before and after the exposure reveals that exposure to water didn't lead to a measurable change in the D content (Fig. 3a, c) in all measured points. This implies that no significant amount of D was released from the samples during the experiment. Again, from the point of view of ITER operation, the observation that negligible amount of D was lost from the sample as a result of exposure is a positive result. Deuterium release from JET samples can be used as a proxy for the release of T in ITER. A significant loss of D during the exposure would indicate that ITER PFCs, which in the nuclear phase would retain similar amounts of D and T, would release significant amounts of tritium when in contact with water. As a result, a significant amount of T-contaminated water would be generated. No loss of fuel species in the reported experiment indicates that no significant T loss would occur either. Therefore, it may be expected that under water ingress Be wall components will not generate large amounts of highly tritiated water.

In Fig. 3b, d it can be perceived that the oxygen concentration been increased somewhat as a result of the exposure to water. That increase of the surface oxygen might possibly be associated either with the enhanced surface oxidation or the adsorption of water in the surface layer. Surface roughness may play a role in the latter. Raman spectroscopy was performed to ascertain whether a surface oxide is present, and the to characterize it. Comparative studies were done on the unexposed reference Be coupon and on the water-treated samples. Fig. 4 shows the recorded Raman spectra for those two types of surfaces. All spectra feature a noticeable peak at the wavenumber $\sim 450\text{ cm}^{-1}$, which is a characteristic Be peak [20]. On the other hand, BeO peaks are expected to be present at the wavenumbers $\sim 675\text{ cm}^{-1}$ and 725 cm^{-1} [21]. Such peaks are absent in all investigated samples, thus indicating only negligible amount of oxide present on the surface following the exposure to water.

Both IBA and Raman data demonstrate that a thick oxide layer was not formed. SEM and EDX surface imaging and elemental mapping also support this conclusion, as no extra features have been identified on the

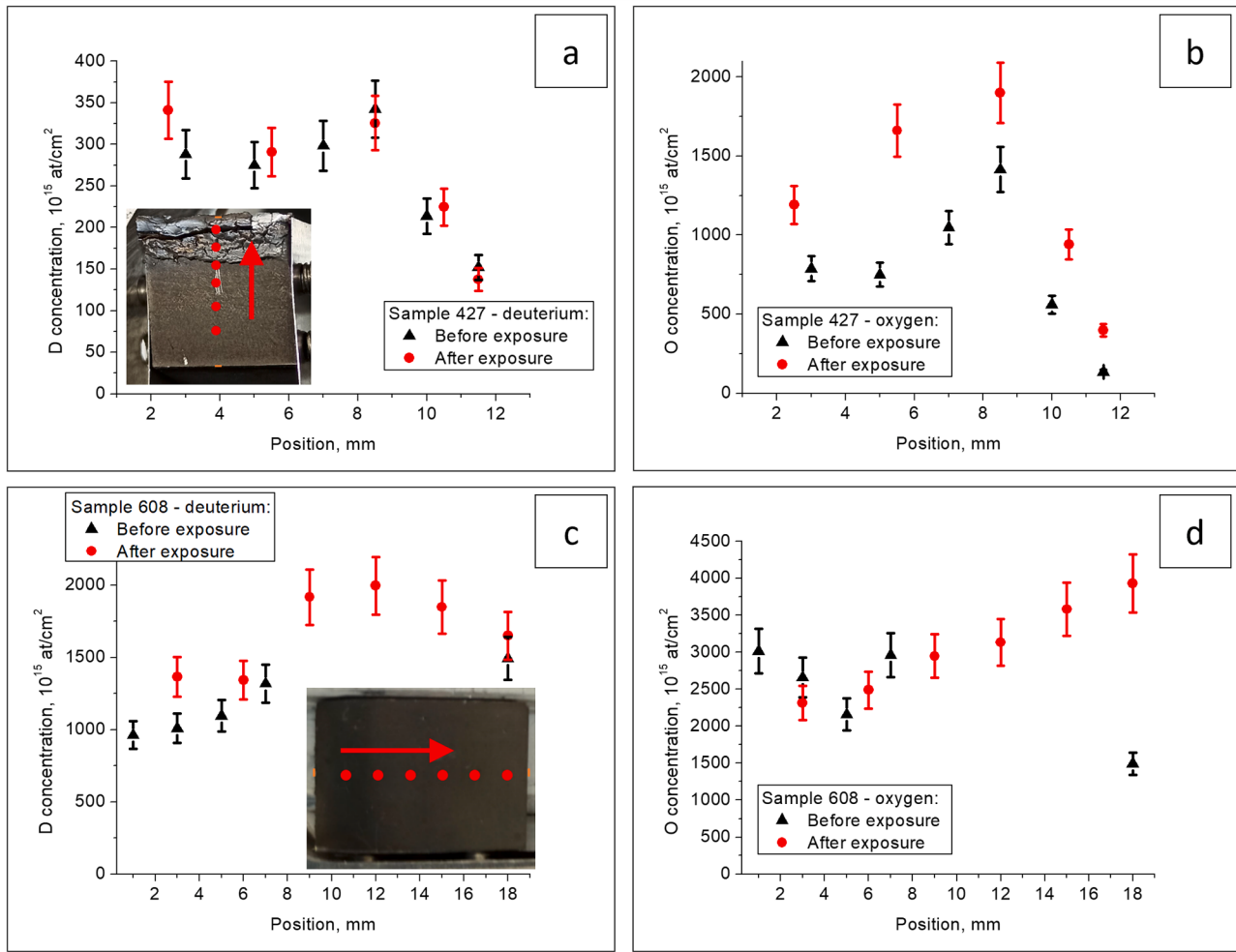


Fig. 3. Results of the IBA measurements across samples (a) deuterium in Sample 427 (with melt damage and crack network); (b) oxygen in 427; (c) deuterium in Sample 608 (with Be co-deposit); (d) oxygen in 608. Inserts – approximate measurement locations on the corresponding samples; the arrows represent the direction of x-coordinate.

examined surfaces after the exposure. All results indicate that exposure of Be PFCs to boiling water, even for a prolonged time, does not lead to either their strong oxidation or permanent degradation of surface properties.

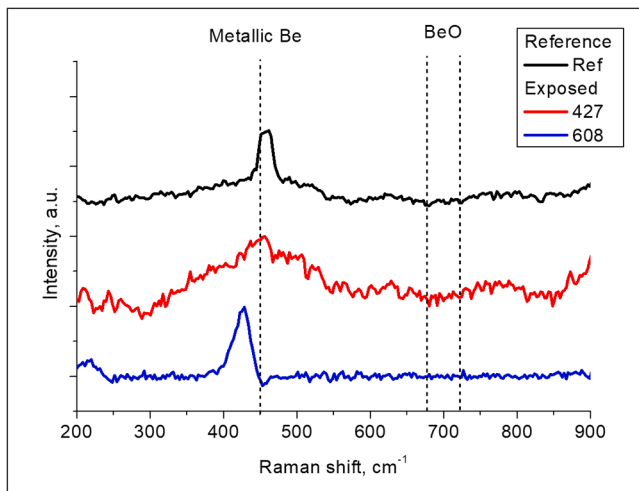


Fig. 4. Raman spectra of exposed and reference Be samples.

Again, from the point of view of ITER operation and Be PFC lifetime the result is positive. It suggests that not only exposure of Be components to boiling water is unlikely to lead to mechanical damage, as detailed above, but also no chemical modification (oxidation) should be expected. Taken together these findings suggest that water ingress would not lead to a considerable degradation of properties of Be PFCs, and hence in the event of such ingress affected components might not need to be replaced.

It should be stressed, however, that presented results should be taken with care, as several limitations of the study can be pointed out. Conditions used in the presented experiments are of course not fully representative of the conditions of water ingress event into a tokamak during plasma operation. In particular, water that enters the vessel from a ruptured cooling pipe, within which it is pressurized, will be moving at high speed. Therefore, in addition to a thermomechanical stress due to the high temperature, a mechanical load due to high-speed impact is to be taken into account. This impact could not be incorporated in the laboratory studies. Complicated chemical effects due to the presence of corrosion products in the cooling water are also possible [22].

Undesirable chemical effects, not related to the thermomechanical stability of Be components, are possible, such as generation of hydrogen in the reactions between hot Be and steam; presence of this hydrogen can then represent a danger of explosion. These effects were altogether outside of scope of the present work. Studies of steam oxidation of Be and subsequent hydrogen generation were reported [23,24], but were

never performed on actual tokamak materials and therefore don't include the effects of Be co-deposited layers.

Finally, during the D-T operation PFCs, will also be exposed to high energy neutrons. As neutron irradiation leads to embrittlement of Be [25], it stands to reason that Be components, due to increased brittleness, may become more susceptible to cracking as a result of thermo-mechanical stresses of any nature including the exposure to hot coolant water in the event of an accident.

In summary, the results presented here are promising, but further studies are required for full assessment of water interaction effects from the point of view of ITER operation. Promising avenues for further study on the topic can be suggested – in particular, the study of dynamic effects of impact of water moving at high speed with Be (bulk and deposits), the study of hydrogen generation in Be-steam interaction and investigation of interaction of boiling water with irradiated Be.

4. Conclusions

The effects of hot water interaction with damaged Be PFCs and co-deposited layers were studied to assess the consequences of an accident involving water ingress into the tokamak vessel. The motivation was related to the nuclear safety in the reactor (ITER) operation. To authors' knowledge, it is the first so comprehensive approach comprising both long-term exposure of Be tiles to water and analyses performed by a set of complementary ion, electron and photon-based methods. The results clearly indicate that neither material damage nor surface modification (i.e. no enhanced Be oxidation) and dust generation have occurred. This may suggest that in case of an accident involving water, Be tiles are not expected to be a major source of mobilizable dust. No damage and only negligible Be oxidation (if any) means that water ingress may not lead to a significant permanent deterioration of Be components, hence not necessarily they will have to be replaced. Very little, if any, D was released into water to form HDO and D₂O molecules. It may be tentatively stated that in the D-T phase of reactor operation, the tritium release from either bulk Be or Be co-deposits would be limited.

From the point of view of the future ITER operation, the described findings constitute a positive result and allow for some optimism regarding a range of safety aspects. However, it has to be stressed that more studies are needed done under conditions more closely reflecting the ITER nuclear environment: the presence of corrosion products in water, high speed flow of water and radiation damage in the tested material.

CRediT authorship contribution statement

Y. Zayachuk: Methodology, Investigation, Writing – original draft, Writing – review & editing. **N. Catarino:** Investigation, Writing – review & editing. **C. Smith:** Investigation, Writing – review & editing. **I. Jepu:** Methodology. **C. Ayres:** Methodology. **A. Widdowson:** Methodology, Conceptualization, Project administration, Writing – review & editing. **E. Alves:** Project administration. **M. Rubel:** Conceptualization, Methodology, Project administration, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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