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Experimental studies on joinability of zircaloy and SiC/SiC composite with titanium powder

Hisashi Serizawa[1] | Naofumi Nakazato[2] | Yuji Sato[1] | Masahiro Tsukamoto[1] | Joon-Soo Park[3] | Hirotatsu Kishimoto[2]

[1]Joining and Welding Research Institute, Osaka University, Osaka, Japan

[2]College of Design and Manufacturing Technology, Muroran Institute of Technology, Muroran, Japan

[3]Organization of Advanced Sustainability Initiative for Energy System/Material (OASIS), Muroran Institute of Technology, Muroran, Japan

Correspondence
Hisashi Serizawa, Joining and Welding Research Institute, Osaka University, 11-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan.
Email: serizawa@jwri.osaka-u.ac.jp

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Abstract

As for the enhanced accident tolerant fuel cladding for the advanced light-water reactor, silicon carbide fiber reinforced silicon carbide matrix composite (SiC/SiC composite) is expected as a potential replacement of the current Zircaloy fuel cladding due to its many superiorities, where it is a key issue to develop the endcap seal of SiC/SiC composite cladding. In this research, based on the calking method, the fiber laser irradiation method was employed for sealing the endcap of SiC/SiC composite tube by Zircaloy tube, where the titanium micro-powder was packed in the slit manufactured on the outer surface of SiC/SiC composite tube. By applying the laser beam irradiation to the outer surface of Zircaloy tube circumferentially, the mechanical calking joint was successfully fabricated. In addition, it is revealed that the good adhesion between Zircaloy and SiC/SiC composite was partially produced as the result of the generation of all proportional solid solution between titanium and zirconium. Moreover, the examinations about the slit shape influences indicate that the narrow hook slit seems to be the best type of silt for holding the titanium powder during the laser irradiation, where the width of slit should be narrower than that of laser irradiation line.

KEYWORDS

composites, joints/joining, lasers, silicon carbide

1 | INTRODUCTION

In order to improve the reliability of light-water reactor, various designs of the enhanced accident tolerant fuel (ATF) cladding for the advanced light-water reactor fuels have been proposed instead of the current used Zircaloy/UO2 fuels [1–3]. Silicon carbide fiber reinforced silicon carbide matrix (SiC/SiC) composite is expected as one of ATF claddings due to many superiorities: high-temperature thermos-mechanical and chemical stability, radiation tolerance, and so on[4–6]. In addition, SiC/SiC composite is stable at temperature higher than 2000 oC and would not melt under loss of coolant accident conditions. Although there have been various studies about the enhanced ATF claddings, the endcap seal of SiC/SiC composite is still a key problem to

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be solved[1–3,7–10]. The final goal for ATF is to use SiC/SiC composite (or monolithic silicon carbide) as the endcaps. However, SiC/SiC composite cladding with Zircaloy endcaps seems to be one possible design because the clear definition about severe accident may change potential fuel pin design.

As for the joining method of Zircaloy and SiC/SiC composite, the brazing was successfully conducted for SiC/SiC composite claddings[9,10], which was employed for Halden Boiling Water Reactor irradiation test at Halden in "SCARLET (SiC Fuel Cladding/Assembly Research, Launching Extra-Safe Technology)" project organized by Prof. Kohyama.[11] However, the melting temperature of brazing filler metal is generally much lower than those of target materials joined and there is a strong demand to establish the joint between Zircaloy and SiC/SiC composite without the brazing filler metal.

Recently, as a result of research and development about the joining technologies, the high-power fiber laser has been developed as one of the high-quality heat source[12] and the authors have examined the applicability of fiber laser irradiation for joining between Zircaloy and SiC/SiC composite tubes by using 4 kW fiber laser.[13] The results indicate that, although the mechanical calking joint of two tubes was successfully produced due to the difference in the coefficient of thermal expansion between SiC/SiC composite and Zircaloy[14,15] based on the calking method or shrink fitting, the airtightness was insufficient for the endcaps of SiC/SiC composite cladding. In addition, the applicability of 500 W diode laser instead of 4 kW fiber laser was examined and it was revealed that the good adhesion between Zircaloy and SiC/SiC composite was partially produced as the result of formation of all proportional solid solution between titanium and zirconium[16], which is the main component of Zircaloy, by inserting titanium micro-powder between Zircaloy and SiC/SiC composite tubes[17]. However, the airtightness was still insufficient for the endcap seal of SiC/SiC composite tube because all the proportional solid solution was not uniformly generated along circumferential direction. Moreover, the examinations about the 500 W diode laser irradiation and the insert method of titanium micro-powder suggest that the wide width laser irradiation seems to be inadequate because the larger shrinkage of Zircaloy would enhance the debonding between two tubes and the hook slit is considered to be the best cutting method for holding the titanium powder during the laser irradiation.[18]

In this research, three types of hook silt were employed for packing titanium micro-powder and the influence of titanium powder packing method on joinability between Zircaloy and SiC/SiC composite tubes were examined, where 500 W fiber laser was used instead of diode laser in order to prevent the effect of wide width laser irradiation.

2 | EXPERIMENTAL PROCEDURE

The materials used were NITE (Nano Infiltration and Transient Eutectic-phase Process) SiC/SiC composite[5] and Zircaloy-2 tubes, whose outer and inner diameters are 12.0 and 10.0 and about 12.3 and 10.6 mm, respectively. The composition of matrix in SiC/SiC composite is almost stoichiometric, and its structure is highly crystalline, while the chemical composition of Zircaloy-2 is shown in Table 1. Because the coefficient of thermal expansion of Zircaloy is larger than that of SiC/SiC composite[6,14,15], the inner face of Zircaloy tube and the outer surface of SiC/SiC composite tube were machined so that SiC/SiC composite tube was fitted into Zircaloy tube. Then, by applying the laser irradiation on the outer surface of Zircaloy tube, SiC/SiC composite tube is calked thermally.

Based on our previous studies[17,18], a flat cuttings were conducted for the inner surface of Zircaloy and the outer surface of SiC/SiC composite as shown in Figure 1. Where thickness of Zircaloy and SiC/SiC composite tubes was reduced to be about 0.35 and 0.85 mm, respectively. In addition, the slits were machined on the outer surface of SiC/SiC composite tube and the titanium powder was filled into the slits as shown in Figure 2, where a clearance between two tubes was set to be about 25 μm in order to prevent the dispersion of titanium powder during the laser irradiation because the particle size of titanium powder is smaller than 45 μm. Figure 3 shows the three types of slits tested in this research and those are denoted as "Wide Hook Slit 1", "Wide Hook Slit 2," and "Narrow Hook Slit" according to the width of slit.

A 500 W diode laser, which was employed in our previous studies,[17,18] is used for the laser cladding, and the shape of beam spot at a focal position is a rectangle whose size is $2600 \times 300\ \mu m^2$,[19] so that the wide width laser irradiation may enhance the debonding between Zircaloy and SiC/SiC composite tubes. Then, in this research, a 500 W fiber laser was employed instead of the diode laser where the shape of fiber laser beam spot at a focal position was a circle whose diameter was 200 μm. Figure 4 shows the photographs of the experimental equipment, where the position of laser and rotational apparatus were fixed and the two tubes were rotated with a constant speed during the laser irradiation. The focal position was set the outer surface of Zircaloy tube. In

TABLE 1 Chemical composition of Zircaloy-2. (unit: wt%)

	Cr	Fe	Ni	O	Sn	Zr
Zircaloy-2	0.104-0.107	0.168-0.179	0.068-0.072	0.121-0.125	1.38-1.45	Bal.

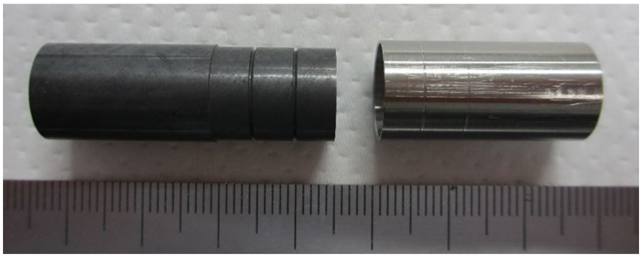

FIGURE 1 Photographs of Zircaloy and SiC/SiC composite tube after flat cutting with slits

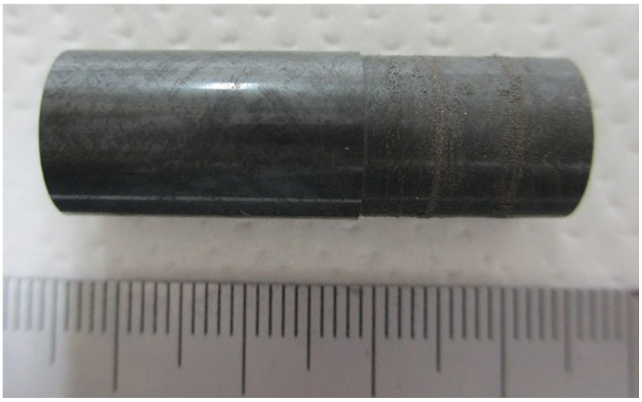

FIGURE 2 Photograph of SiC/SiC composite tubes with titanium powder

(A) Wide Hook Slit 1 **(B)** Wide Hook Slit 2 **(C)** Narrow Hook Slit

FIGURE 3 SEM images of three types of slit

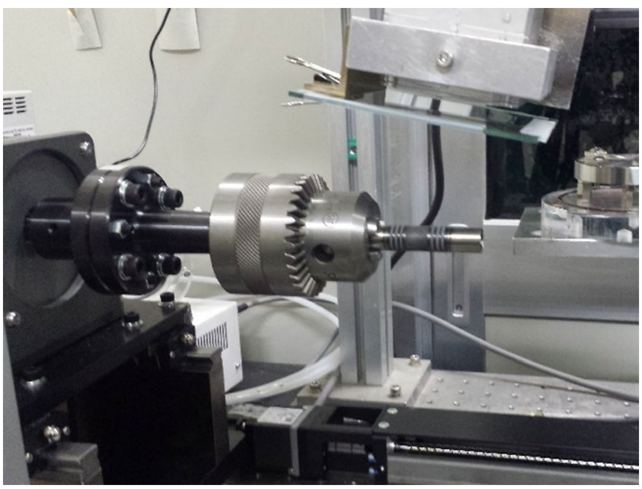

FIGURE 4 Photographs of experimental equipment for laser irradiation

addition, in order to prevent any oxidization of materials, an argon gas was used for the shielding gas whose flow was 40 L/min.

Although the final target value of airtightness for SiC/SiC composite cladding with Zircaloy endcaps is less than 2×10^{-10} Pa m^3/s, the simple bubble leak test was employed according to the following test method in order to identify the airtightness easily. The joint was dropped into the water, and the air pressure, which is larger than water pressure, was applied into the tube by using the air compressor. Then, the airtightness was evaluated through the visual examination of bubbles.

3 | RESULTS AND DISCUSSIONS

Figure 5 shows the overview of Zircaloy-SiC/SiC composite tubes with Wide Hook silt 1 after the laser irradiations. In addition to the two circumferential lines for the slits, two laser irradiations were applied at the interface between Zircaloy and SiC/SiC composite tubes in order to prevent the dispersion of titanium powder. The rotational speed and laser irradiation time for the prevention of titanium powder dispersion were 60 rpm and 1.2 sec, respectively, while those for the slits were 60 rpm and 3.2 s respectively. Those laser irradiation conditions were decided according to our previous studies.[17,18] Although the smoke was not observed apparently during the laser irradiations to the slits and the mechanical calking joint of Zircaloy and SiC/SiC composite tubes was successfully produced, the air leak test of this calking joint indicated that this joint is still insufficient for the endcaps of SiC/SiC composite cladding.

The calking joint was cut to the half haves along the joint length after embedding in the resin, and the scanning electron microscope (SEM) images of the joint interface are shown in Figure 6. Although the hook slit was employed for holding the titanium powder deeply, the gap between Zircaloy and SiC/SiC composite remained after the laser irradiation obviously. As shown in Figure 6C, the width of laser irradiation to Zircaloy is narrower than the width of Wide Hook Silt 1 and the outer Zircaloy tube seems to bend into the wide slit, so that the crack is observed at the center of irradiation position of Zircaloy. However, because the good adhesions were generated at the edges of hook slits as shown in Figures 6B and D, these gaps might be eliminated by reducing the width of hook slit. In order to analyze a generation mechanism of the adhesion between two tubes shown in Figure 6D, the elemental analysis was conducted using energy dispersive X-ray spectrometry (EDS) and the results are summarized in Figure 7 where the backscattered electron image (BEI) of area analyzed was also shown in this figure. From this figure, it is revealed that only the titanium layer was attached

FIGURE 5 Photograph of Zircaloy-SiC/SiC composite joints with Wide Hook Slit 1 after fiber laser irradiation

with SiC/SiC composite and all proportional solid solution was successfully produced between the titanium layer and Zircaloy. In addition, Figure 6 indicates that only the outer surface of Zircaloy irradiated is melted and the reaction between the titanium powder and zirconium occurred at only the inner surface of Zircaloy because the melting temperature of titanium is lower than that of zirconium.[16] Namely, the most of Zircaloy tube seems to keep the original microstructure and then its physical properties might not be changed after the laser irradiation. Then, it can be concluded that the width of slit should be narrower than that of laser irradiation line.

As for the narrower slit to the laser irradiation line, two types of silt which are Wide Hook Silt 2 and Narrow Hook Slit were tested. Because the titanium powder packed in the hook silt was partially disappeared after laser irradiations to the slit despite the additional laser irradiation for the prevention of titanium powder dispersion as shown in Figure 6, the previous two irradiations were omitted in the following irradiation tests. Both the cases of Wide Hook Slit 2 and Narrow Hook Slit, the two laser irradiations were applied to the slits circumferentially where the rotational speed and laser irradiation time were 60 rpm and 3.2 s respectively. Figure 8 shows the overviews of Zircaloy-SiC/SiC composite tubes with Wide Hook slit 2 and Narrow Hook Slit after the laser irradiations. Although there are not any cracks or voids along the irradiation lines and the mechanical calking joints were successfully produced, the air leak tests indicated that these joints are still insufficient for the endcaps of SiC/SiC composite cladding. However, the number of bubbles in the case of Narrow Hook Slit was

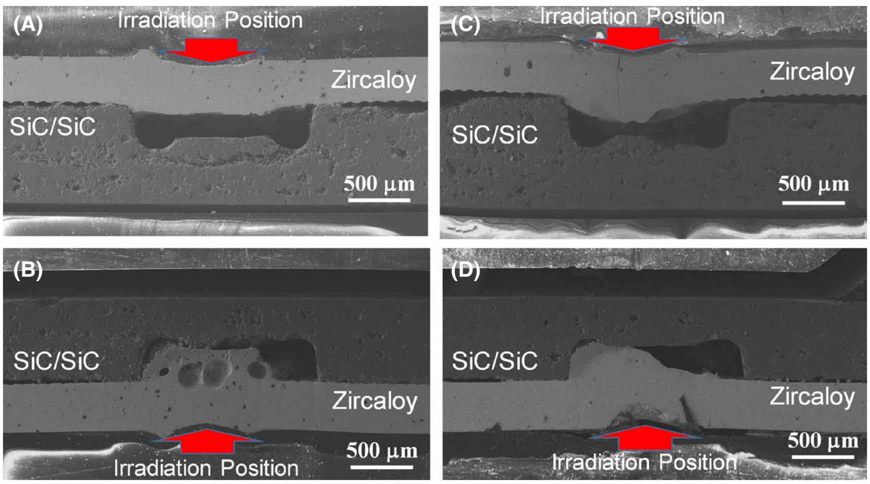

FIGURE 6 SEM images of joint interface of Zircaloy-SiC/SiC composite joint with Wide Hook Slit 1 after fiber laser irradiation

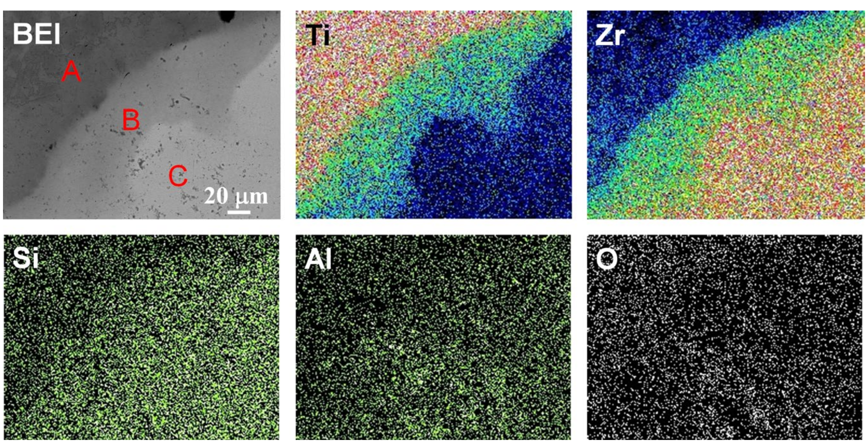

FIGURE 7 BEI and EDS results of adhesion part between Zircaloy and SiC/SiC composite with Wide Hook Silt 1 after fiber laser irradiation shown in Figure 6D

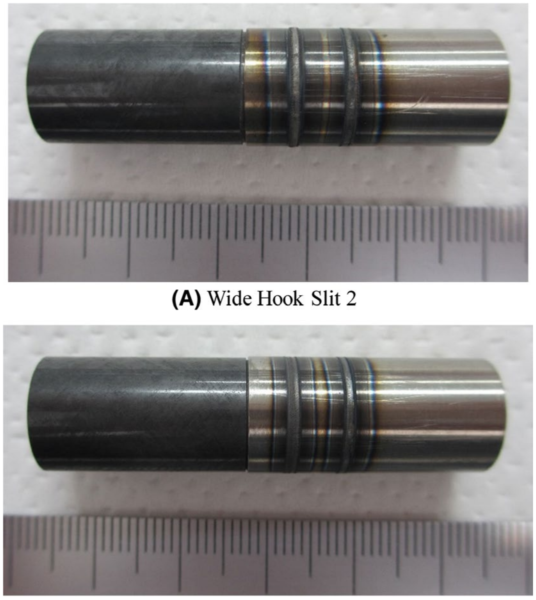
(A) Wide Hook Slit 2
(B) Narrow Hook Slit

FIGURE 8 Photograph of Zircaloy-SiC/SiC composite joints with Wide Hook Slit 2 and Narrow Hook Slit after fiber laser irradiation

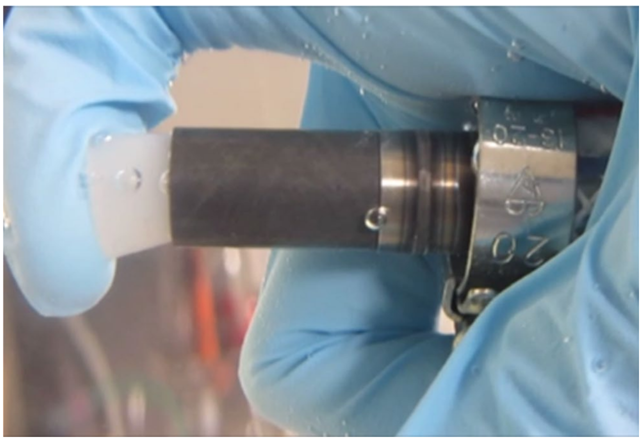

FIGURE 9 Photograph of airtightness test for Zircaloy-SiC/SiC composite joint with Narrow Hook Slit after fiber laser irradiation

much smaller than that in the case of Wide Hook Slit 2 and the bubbles were observed at only one point as shown in Figure 9.

In order to examine the formation of all proportional solid solution in the better calking joint with Narrow Hook Slit, this joint was cut to the half haves and Figure 10 shows SEM images of the joint interface. From this figure, it is found that the good adhesions were produced at the edges of hook slits as shown in Figure 10A, C, and D, although the titanium powder remains unreacted as shown in Figure 10A-C. The enlarged SEM view and BEI of Figure 10A are shown in Figure 11. From this figure, it is revealed that the adhesion part can be divided into three regions as same as Figure 8 and these regions seem to be Zircaloy, all proportional solid solution of zirconium and titanium, and the titanium, respectively. Finally, it can be concluded that the Narrow Hook Slit would be the best type of slit for SiC/SiC composite for holding the titanium powder during the laser irradiation where the width of slit should be narrower than that of laser irradiation line.

On the other hand, although the high radio frequency circumferential heating would be selected as one of the other circumferential heating methods, this method seems to be inadequate because the entire thickness of Zircaloy tube was thermally affected and the heated zone of the high radio frequency circumferential heating would be much wider than that of the laser irradiation.

4 | CONCLUSIONS

In order to examine the effect of titanium powder packing method on joinability between Zircaloy and SiC/SiC composite tubes, three types of hook slit were employed for packing titanium powder. The conclusions can be summarized as follows.

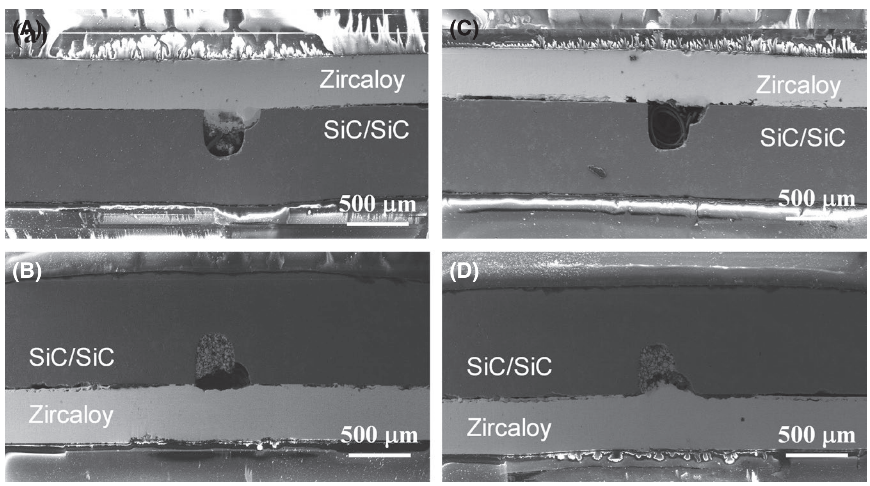

FIGURE 10 SEM images of joint interface of Zircaloy-SiC/SiC composite joint with Narrow Hook Silt after fiber laser irradiation

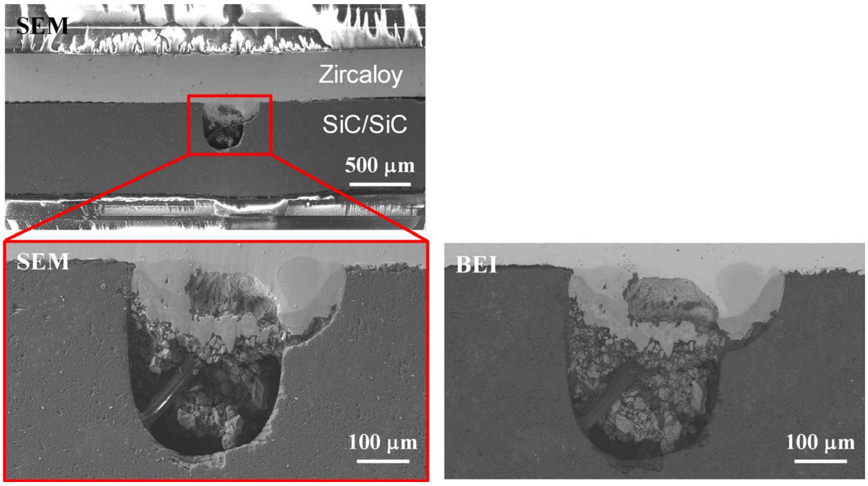

FIGURE 11 SEM image and BEI of joint interface in Zircaloy-SiC/SiC composite joint with Narrow Hook Silt after fiber laser irradiation shown in Figure 10(A)

1. Regardless of hook slit type, the mechanical calking joint of Zircaloy and SiC/SiC composite tubes was successfully produced.
2. Although all proportional solid solution between titanium and zirconium seems to be successfully formed as the result of laser irradiation, the width of slit should be narrower than that of laser irradiation line to prevent the unnecessary deformation in Zircaloy tube.
3. Although the airtightness of calking joint was still insufficient for the endcap of SiC/SiC composite cladding, the narrow hook slit would be the best type of slit for SiC/SiC composite for holding the titanium powder during the laser irradiation.

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ORCID

Hisashi Serizawa https://orcid.org/0000-0002-9890-7772

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