

**ORIGINAL ARTICLE**

# **Microstructure Evaluation of the Interface between Dental Zirconia Ceramics and Veneering Porcelain**

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## **Synopsis**

The application of zirconia ceramics for the dental restorations and dental implants has increased. However the weak performance of the veneering porcelains and low adhesion to the zirconia framework were reported. In this study, the micro-structure, elemental distribution and crystal phase around the interface between zirconia and veneering porcelain were estimated using SEM-EDS (energy dispersed spectrometry) and micro-XRD (X-ray diffraction). The specimen fired at 940°C for 1 minute showed clear interface in the SEM and the elemental distribution images of Zr and Si. With extending firing period up to 384 hours, slight diffusions of Zr and Si, which are the major components of zirconia and porcelain were suggested. In micro-XRD, no phase transformation was observed in the zirconia and porcelain, thus extension of firing period would not affect the crystal phase around the zirconia/porcelain interface. Therefore, the extension of firing period was expected to slightly improve the zirconia/porcelain bonding.

**Key words:** *zirconia ceramics, veneering porcelain, microstructure*

## **Introduction**

In recent years, the application of zirconia ceramics for the dental restorations (e.g. framework of fixed partial dentures (FPDs) and dental implants) has increased because of its high mechanical toughness, esthetics and biocompatibility [1]. In the early 1990s yttrium oxide partially stabilized tetragonal zirconia polycrystal (Y-TZP) was introduced to dentistry as a core material for all-ceramic restorations and has been made available through the CAD/CAM technique. Due to a transformation toughening mechanism, Y-TZP has been shown to have superior mechanical properties compared to other all-ceramic systems [2,3]. For example, Y-TZP

demonstrated a flexural strength of 900-1,200MPa in vitro, which is higher than that of dental alloys [2].

For the actual application for dental restorations the zirconia is veneered with dental porcelain in order to improve its estheticity. Thus, the long-term stability of both Y-TZP framework and veneered porcelain are required. In the clinical studies within 1 to 3 years the Y-TZP core ceramic exhibited a high stability as a framework material and no fractures of the zirconia framework could be observed [4-8]. However the weak performance of the veneering porcelains and low adhesion to the zirconia framework were reported [5,7,8]. Especially minor

chip-off fractures of the veneering porcelain were described as the most frequent reason for failures of zirconia restorations. Chip-off fracture rates at 15% after 24 months [6] 25% after 31 months [8] and 8% and 13% after 36 and 38 months [5,7], were reported. Therefore the bonding strength would be most important for the long-term stability in the clinical usage. Concerning to the bonding strength and thermal cycling effect between zirconia cores and veneering porcelain was reported [9]. For further improvement of their bonding strength, the observation and estimation and estimation of micro-structure of their interface is required.

In this study, the micro-structure, elemental distribution and crystal phase around the interface between zirconia and veneering porcelain were estimated using a scanning electron microscope (SEM), energy dispersive X-ray spectroscopy (EDS) and micro X-ray diffraction (Micro-XRD).

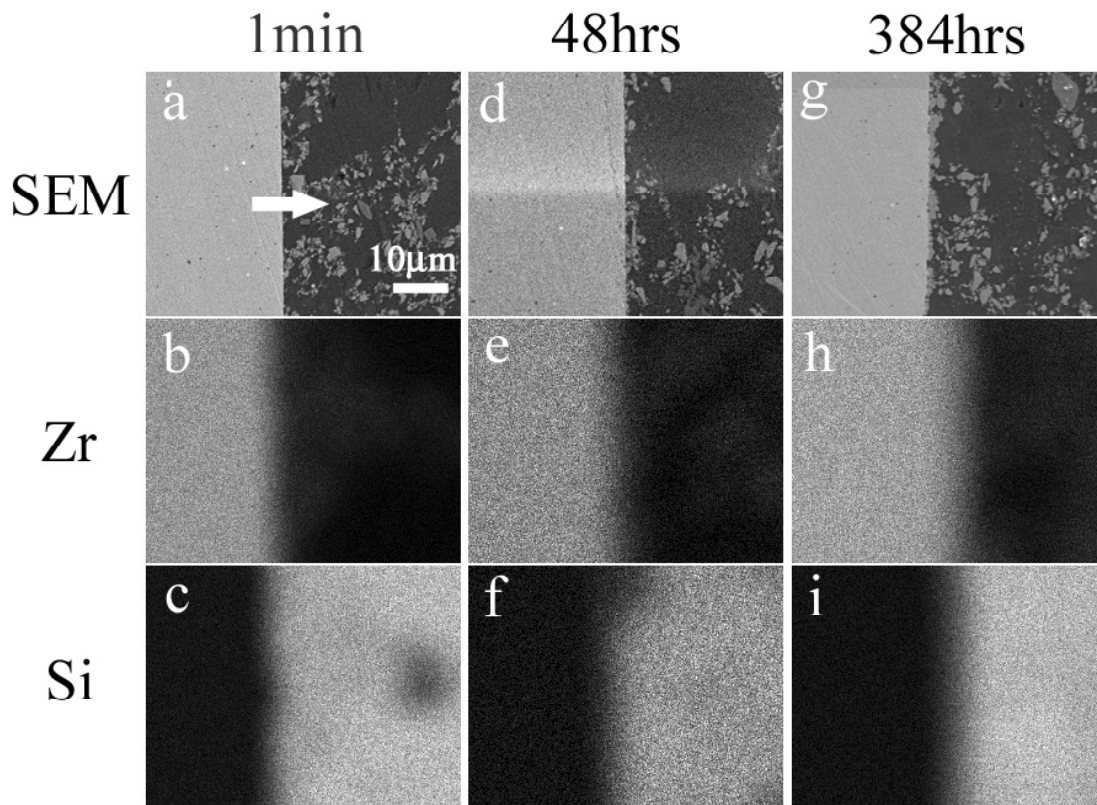
## Materials and Methods

### 1. Preparation of zirconia substrate

0.6g of yttria stabilized zirconia (TZ-3YB-E, Tosoh corporation, Tokyo, Japan) was pressed into disk shape using cylindrical mold and sintered at 1350°C for 2 hours in air with following the manufacturer's instruction. The sintered zirconia disks were 8mm in diameter and 2mm in thickness and density was 6.01g/cm<sup>3</sup>. The obtained disks were polished with emery paper (#2000) and cleaned with sonication.

### 2. Preparation of veneering porcelain

The veneering porcelain (Vintage ZR A3B, Shofu Inc, Kyoto, Japan) was layered in 1mm thickness and fired at 940°C for 1 minute with following the manufacturer's recommendation. Parts of specimens were extended the firing period up to 384 hours. The porcelain veneered zirconia was cross-sectioned, polished and applied for the following observations.



**Fig. 1** SEM images and elemental distribution (Zr, Si) images.

### 3. SEM and EDS analyses

The interface between zirconia and porcelain was analyzed using a scanning electron microscope (SEM) (S-2380, Hitachi, Tokyo, Japan). The elemental distribution around the interface was estimated using an energy dispersive X-ray spectroscopy (EDS) (Genesis, EDAX Japan, Tokyo, Japan). The elemental distribution images of interface was acquired with the resolution of 256 x 200 pixels with integration time for 200  $\mu$ sec per point. The acceleration voltage was 15kv.

### 4. Micro-X-ray diffraction analyses

The crystal phase distribution of around the interface between zirconia and porcelain was analyzed using micro X-ray diffractometer (JDX-3500, JEOL, Tokyo, Japan) with 30 $\mu$ m collimator in diameter. Measurement was carried for 5 points in 50  $\mu$ m step across the zirco-

nia/porcelain interface. Diffraction spectrum was integrated with the position sensitive proportional counter (PSPC) for 30 minutes in each point.

### Results

Fig. 1 shows the SEM images and elemental distribution (Zr, Si) images of the interface between zirconia and porcelain with different firing periods (1min, 48hrs, 384hrs). The specimen fired at 940 $^{\circ}$ C for 1 minute showed clear interface in the SEM image(Fig.1(a)). The elemental distribution images (Fig.1(b)and(c)) were also clearly separated into a zirconia part (Zr-rich area) and a porcelain part (Si-rich area), and slight transition layer was observed. The specimens with extended firing period for 48 and 384 hrs also showed clear interface in SEM images (d, g) and no significant change in the transition layer was observed in the Zr(e, h) and Si(f, i)

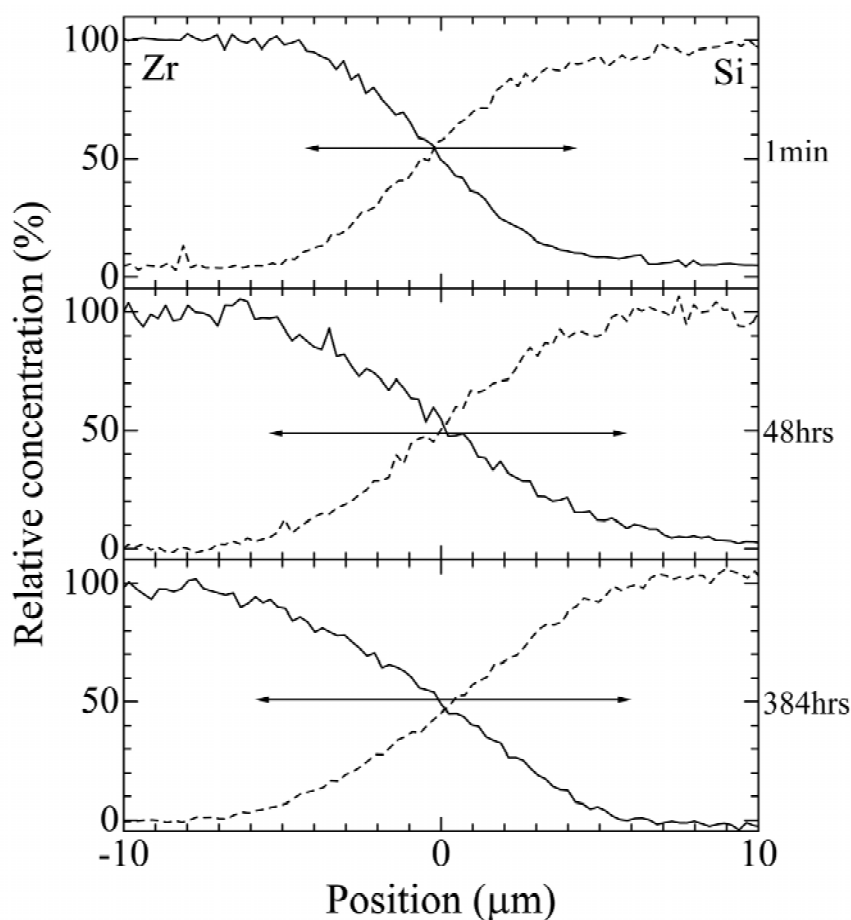


Fig. 2 Line analysis of the interface between zirconia and porcelain.

distribution images. Thus, remarkable changes with extension of firing period could not be observed in the elemental distribution images.

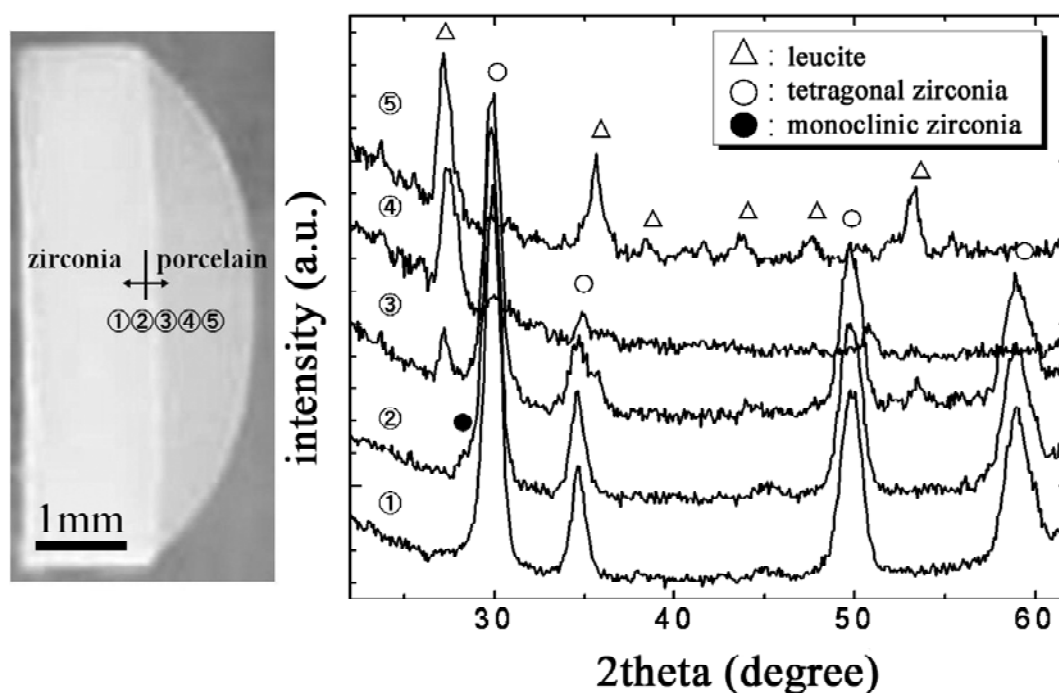
Detailed elemental distributions around the interface were estimated with line analysis of SEM/EDS. Fig. 2 shows the line analysis of the interface between zirconia and porcelain along the arrows in Fig 1. The thickness of the transition layers of Zr and Si concentration were observed as 10 $\mu$ m approximately. The decrease of Zr concentration in the transition layer was similar to that of Si concentration. The layer thickness was slightly increased with extending the firing period from 1minute to 384 hours. Therefore, the mutual diffusion of Zr and Si, which are the major components of zirconia and porcelain was suggested.

Fig. 3 shows the XRD spectra of the zirconia/porcelain interface of a specimen fired at 940°C for 384 hours. Tetragonal zirconia and leucite represented in the zirconia and porcelain, respectively. The spectrum at the interface (No. 2) showed quite small peak assigned to monoclinic zirconia, except for the tetragonal zirconia and leucite. However, no significant phase trans-

sition was observed around the zirconia/porcelain interface.

### Discussion

In the present study, we paid attention to interface between zirconia ceramics and porcelain and evaluated their microstructures or changes of a crystal phase in various conditions by using SEM-EDS and Micro-XRD. In the SEM observation, zirconia/porcelain interface was clearly divided to both layer and no mutual diffusion was observed. In the elemental distribution images and line analysis of Zr and Si, the transition layer of both elements were observed. However, those don't directly suggest the mutual diffusion on the zirconia/porcelain interface. Because, the lateral resolution of SEM/EDS is larger than 1 $\mu$ m [10, 11] and that depends on the specimen and acceleration voltage. In the observation condition in this study, the penetration range of irradiated electron, which is similar to the range of the characteristic X-ray generation, was supposed as 2-3 $\mu$ m. That is the lateral resolution of the elemental distribution and line analysis. The thickness of the transition layer of Zr and Si



**Fig.3** XRD spectra of the zirconia/porcelain interface of a specimen fired at 940°C for 384 hours. Each measured point was shown in the picture.

concentration estimated in the line analysis is 10 $\mu$ m, approximately. Thus, considering the lateral resolution of SEM/EDS analysis, the observed elemental transition layer would be mainly caused by the low resolution of SEM/EDS analysis, and the actual elemental diffusion would be quite thin. The increase of the firing period up to 48 and 384 hours slightly increased the transition layer as shown in Fig.2. The SEM/EDS analysis of those specimens were carried under same conditions, thus the increase of the transition layer might be caused by the mutual diffusion of Zr and Si. However, the increase was quite small as few micron with 384 hours firing. Therefore, the mutual diffusion at the zirconia/porcelain interface might be hardly occurred under realistic firing condition. In the micro-XRD measurement, slight peak assigned to monoclinic zirconia was observed at the zirconia/porcelain interface. Concerning to the monoclinic phase formation, Ban et al. reported that polishing or sandblasting treatment would led the phase transformation [12]. Therefore, the slight peak of monoclinic in Fig.3(No.2) would be caused by the surface polishing before the porcelain veneering. No phase transformation or another phase formation was observed in the zirconia and porcelain. Thus, the extension of firing period would not affect the crystal phase around the zirconia/porcelain interface.

### Conclusion

The interface between zirconia and porcelain was clearly divided with the specimen fired at 940°C for 1 minute in the SEM image, and slight transition layer was observed in the elemental distribution images. With extending firing period for 48 and 384 hours, slight diffusions of Zr and Si, which are the major components of zirconia and porcelain were suggested. No phase transformation was observed in the zirconia and porcelain in the micro-XRD. Thus, the extension of firing period would not affect the crystal phase around the zirconia/porcelain interface. This study suggested that the extension of firing period was expected to slightly improve the zirconia/porcelain bonding.

### Acknowledgments

This work was supported by Grant-in-Aid for Challenging Exploratory Research No.21659449, from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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(Received: Jun 4/  
Accepted: Jun 24)

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