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Low Energy Surface Activation of Zirconia Based Restorations

ABSTRACT

Purpose: to evaluate the influence of low energy surface activation technique on the biaxial flexure strength of zirconia frameworks.

Materials and methods: zirconia discs were prepared by cutting CAD/CAM zirconia blocks. Sintered discs were airborne particle abraded using one of the following particles: 30 µm alumina particles, 50 µm alumina particles, or modified round edges 30 µm alumina particles at low pressure. Scanning electron microscopy, x-ray diffraction analysis, surface roughness , and biaxial flexure strength tests were performed (n=20). Fractured specimens were fractographically analyzed (α=0.05).

Results: Low energy surface activation resulted in 7% monoclinic crystallographic transformation, increasing surface roughness from 0.05 to 0.3 µm and in significant increase in biaxial flexure strength (1718 MPa) compared 30 µm (1064 MPa), 50 µm (1210 MPa), and as-sintered specimens (1150 MPa).

Conclusions: low energy surface activation of zirconia specimens improved the biaxial flexure strength of zirconia frameworks without creation of surface damage.

Clinical implications: by controlling particle size and shape of alumina, the flexure strength of zirconia restorations could be increased using low pressure particle abrasion.

Keywords

Surface Activation
Low Pressure Particle Abrasion
XRD
Transformation

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INTRODUCTION

The introduction of zirconia to the dental field opened the design and application limits of all-ceramic restorations. Today accurate and precise long span zirconia restorations are fabricated using CAD/CAM technology with high reliability and success rate. Zirconia is a dynamic material on an ultra-structural level. On a crystal level, tetragonal monoclinic transformation and reverse transformation occur when the material is subjected to mechanical or chemical activation in the form of stress concentration or change in surrounding chemical environment. One of the characteristic advantages of zirconia is its unique crystal transformation where the partially stable tetragonal crystals transform into the more stable monoclinic form. This transformation is accompanied by volumetric expansion (almost 4% expansion) leading to the creation of a compressive filed resulting in interruption of propagating cracks.[1]

Partial stabilization of transformation activity could be achieved by addition of small amounts of doping agents as yttrium, calcium, or magnesium. Controlled addition of doping agents preserves the high temperature tetragonal phase at room temperature.[2] The material still preserves limited capacity to transform to the more stable monoclinic phase thus increasing the fracture toughness of the material. In dentistry, 3 mol Y-TZP is the most widely used partially stabilized zirconia type for fabrication of all-ceramic restorations.

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Through the fabrication process, zirconia frameworks are subjected to different forms of surface damage; one during airborne particle abrasion and the other during internal fit corrections using diamond points. The produced surface damage has a pronounced effect on the mechanical properties of the damaged framework. Previous studies indicated significant reduction in flexure strength after airborne particle abrasion in addition to reduction in the survival probability of the damaged material as indicated by reduction in the calculated Weibull modulus that indicates a decrease in the reliability of strength data.[3,4] It has to be noted that the amount of surface damage has two important parameters; the percent of transformation activity that accompanies surface damage and secondly, the depth of the created surface scratches and grooves. While the first could be reversed using controlled heat treatment, the second remains to be a site for stress concentration. Thus airborne particle abrasion increases the number and distribution of surface defects which is directly related to the fatigue life of the abraded material. Today, several manufacturers do not recommend particle abrasion of their products to avoid unintentional weakening of the material.

However, there remains a need to increase the surface roughness of zirconia frameworks in order to improve bonding and veneering procedures. Due to its high chemical stability, zirconia does not respond to various chemical treatments as phosphoric and hydrofluoric acid etching. Unlike glass ceramic restorations, increasing energy and wettability is not an effective method in establishing a strong and durable bond strength to zirconia. Previous studies used low pressure particle abrasion as non destructive method used to increase surface roughness of zirconia. The parameters of particle size, particle shape, particle type, pressure, nozzle distance, and blasting time control the amount of energy delivered to the surface crystals.[5,6] Proper control of these parameters could result in controlled surface activation of tetragonal monoclinic transformation reaction without the introduction of marked surface damage.

However alumina particles used to perform low pressure particle abrasion have sharp edges which still induce sharp scratches on the surface acting as potential sites for stress generation. Modification of particle shape could solve this problem. The aim of this study was to investigate the influence of low energy surface activation of zirconia using modified low pressure particle abrasion technique. The hypothesis presented in this study was that low energy surface activation would increase biaxial flexure strength of zirconia and reduce surface damage associated with conventional alumina particles.

MATERIALS AND METHODS

PREPARATION OF TEST SPECIMENS

Zirconia discs (20 mm x 0.5 mm) were prepared by cutting CAD/CAM zirconia blocks (Lava zirconia, 3M ESPE, Seefeld, Germany). Before sintering (1350°C for 2 h), the discs were highly polished using 1200 grit silicon carbide paper and ultra fine diamond paste (0.5 µm). The discs ultrasonically cleaned and were divided into four groups based on the performed surface treatment (n=20).

SURFACE TREATMENT

1- Low pressure surface activation:

30 µm alumina particles were ground to remove sharp edges and sharp line angles. 100 g of the original powder were placed in a glass beaker and large marble beads were used to crush the particles in a rotating motion. After 8 h, a 40 µm fine stainless steel sieve was used to filter the powder and remove any larger fragments. The selected powder was washed using 90% ethanol to remove fine dust and dried at 70°C for 4 h. The process was repeated until 2 kg of the modified powder was obtained.

The modified powder was added to an electronically controlled particle abrasion unit. A special attachment unit was used to hold the specimens at 6 cm distance from the blasting nozzle and maintain 20° angle to the streaming line. Pressure was set to 1 bar and the specimens were airborne particle abraded for 12 sec. Only one side of the discs was low energy surface activated. Half of the specimens were heat annealed (heating at 1150°C for 1 h) to activate reverse transformation reaction (monoclinic to transformation).

The kinetic energy of an object is the energy which it possesses due to its motion and is defined as the work needed to accelerate a body of a given mass from rest to its finale velocity. The accelerated particle maintains this kinetic energy unless its speed changes or if it stops by impacting another object releasing its kinetic energy to the struck object. The kinetic energy of a non-rotating object is described using Equation 1:

Kinetic energy= $\frac{1}{2} mv^2$

Where m is the mass of particle and v is its speed. Reducing particle size from 50 µm to 30 µm reduces both its mass and projectile speed and thus the total energy delivered to the struck surface is reduced to almost half. The delivered energy/unit area of the created new surface E required to produce abrasion fragments increases rapidly at first as the corners and edges of the blasting powder are removed.[7]

2 - Airborne particle abrasion:

The prepared zirconia discs were airborne particle abraded using two sizes of alumina particles:

- 30 µm standard unmodified alumina particles at 1 bar using similar procedure as previously described.
- 50 µm alumina particles at 1.5 bar simulating standard laboratory procedure.

CHARACTERIZATION OF THE CREATED SURFACE

Specimens were gold sputter coated, and examined using scanning electron microscopy (SEM) to study surface changes (XL 30, Philips, Eindhoven, the Netherlands). The central surface of the specimens was examined using x-ray diffraction analysis (XRD; M18XHF-SRA, Mac Co., Japan) to detect changes in the crystallographic content before and after heat treatment for all specimens. As-sintered zirconia surface was used as a base line. XRD data were obtained with a diffractometer using nickel-filtered CuK radiation. The tetragonal/monoclinic zirconia ratio was determined using the integrated intensity (measuring the area under the diffractometer peaks) of the tetragonal (101) and two monoclinic (1_11) and (111) peaks. The obtained volume fractions were individually normalized. Although the X-ray penetration is too deep to be considered as a real surface analysis, it allowed precise evaluation of the transformation activity.

Surface roughness (Ra peak valley parameter) was measured using contact profilometry (Marsurf PS1, Mahr GM6H, Gottingen, Germany). The specimens were placed with the treated surface facing the travelling needle of the device and the central region of every specimen was measured three times. The travelling needle covered 3mm stroke distance on the surface.

BIAXIAL FLEXURE STRENGTH

20 specimens of each group were loaded in a special biaxial flexure loading device ensuring adequate support of the discs with the treated surface in tension. The biaxial flexure strength (BFS) was calculated using Equation 2:

$$BFS = \frac{3F(1+\upsilon)\cdot[1+2\ln(D_s / D + \{1 - D^2/(2\cdot D_S^2)\}\cdot(D_s^2/D^2)\cdot(1-\upsilon)/(1+\upsilon)}{4\pi T^2}$$

where F is the fracture load, ν is the Poisson ratio, T is the specimen thickness and Ds and D are the diameters of the support ring and of the tested disc, respectively.

The discs were immersed under inert oil, to prevent effect of moisture during fracture process, and loaded at a crosshead speed of 0.1 mm/min using a universal testing machine (Instron 6022, Instron Limited, High Wycombe, UK). Load deformation curves were collected using computer software and a calibrated digital micrometer. Twenty discs were tested for each surface treatment (n=20).

STATISTICS

One way analysis of variance (ANOVA) and Bonferroni *post hoc* tests were used to analyze the data. Based on sample size (n=20), selected level of significance (α=0.05), and medium effect size, the selected test had adequate power to detect differences in biaxial flexure strength.

RESULTS

There were significant differences in biaxial flexure strength between the tested groups (F=103, P<0.001) as low energy surface activation had significantly higher BFS (1770 ± 56 MPa) compared to as-sintered specimens (1131 ± 93 MPa), heat treated control specimens (1150 ± 72 MPa), 30 µm particle abraded specimens (1064 ± 37 MPa), and 50 µm particle abraded specimens (1210 ± 88 MPa).

Regarding surface roughness, 50 µm particle abraded specimens (Ra=0.45 µm) and 30 µm abraded specimens (Ra=0.21 µm) had significantly higher Ra values (F=57.7, P<0.001) compared to as-sintered specimens (Ra=0.11 µm) and low energy surface activated specimens (Ra=0.19 µm). Previous data are summarized in table 1.

Table 1. Biaxial flexure strength, surface roughness, and percent of monoclinic phase of tested specimens.

Surface treatment	BFS (MPa ± SD)	Ra (µm)	% monoclinic phase
Low pressure particle abrasion	1770 ± 56	0.19 ± 0.03	7.4
30 µm particles	1064 ± 37	0.21 ± 0.04	3.1
50 µm particles	1210 ± 88	0.45 ± 0.04	3.9
As-sintered control	1131 ± 93	0.11 ± 0.01	0
Heat treated control	1150 ± 72	0.12 ± 0.02	0

X ray diffraction analysis revealed 100% tetragonal crystal structure of as-sintered and heat treated specimens while the low energy surface activated specimens had 7.4% monoclinic crystals *(Figure 4)*. 30 μm and 50 μm particle abraded specimens had lower amounts of monoclinic phase; 3.1% and 3.9% respectively. No evidence of cubic or deformed monoclinic crystals was detected. Scanning electron microscope images revealed homogenous zirconia grains (0.5 μm) for all specimens *(Figure 1a)*. Low energy surface activated specimens demonstrated increased in micro-roughness of the treated surface and a pitted structure without the creation of sharp scratches *(Figure 1b)*, while surface damage in the form of scratches, indentations, and sharp grooves were observed for 30 μm and 50 μm particle abraded specimens *(Figure 1c)*.

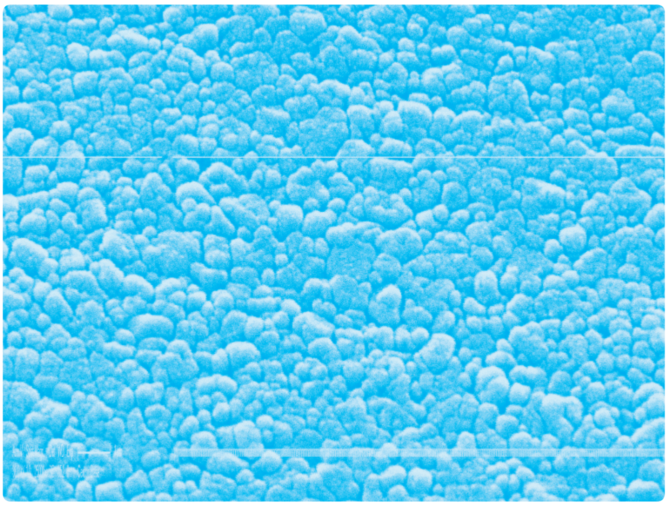

Figure 1a: SEM image, 1000x, demonstrating surface structure of low energy surface activated zirconia specimens

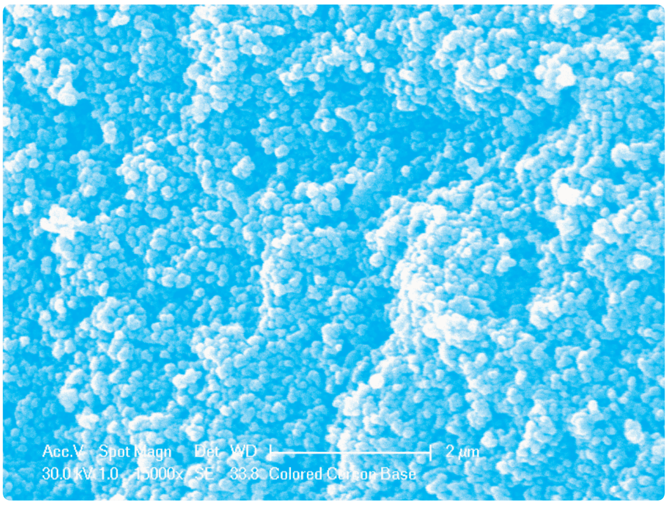

Figure 1b: SEM image, 15000x, demonstrating surface of low energy surface activated zirconia specimens

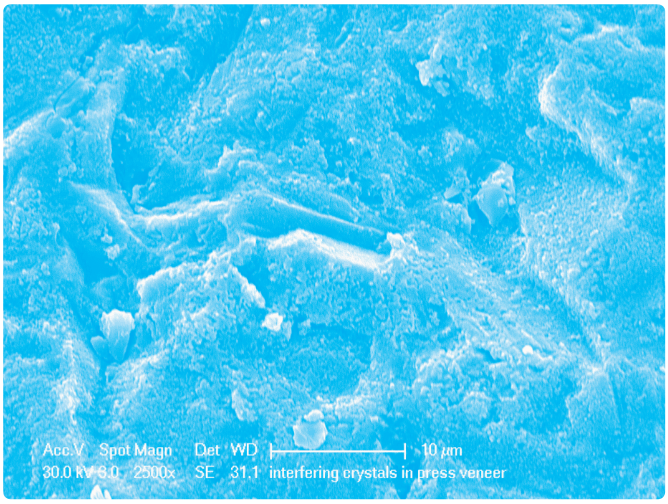

Figure 1c: SEM image, 2500x, demonstrating surface sharp cracks and surface damage of airborne particle abraded zirconia specimens

Fractographic analysis reported subsurface fracture origin for low energy surface activated specimens and critical crack located at the surface fracture for 30 μm and 50 μm particle abraded specimens *(Figure 2a & b)*. Scanning microscopic images revealed also roundation of the particle edges compared to the original sharp edges observed in the original particles *(Figure 3a & b)*.

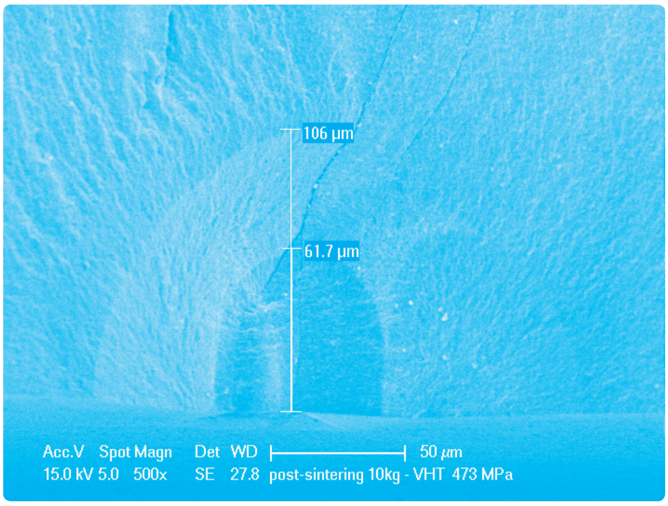

Figure 2a: SEM image, 500x, demonstrating subsurface fracture origin of low energy surface activated specimens indicating presence of compression phase in the region of critical crack

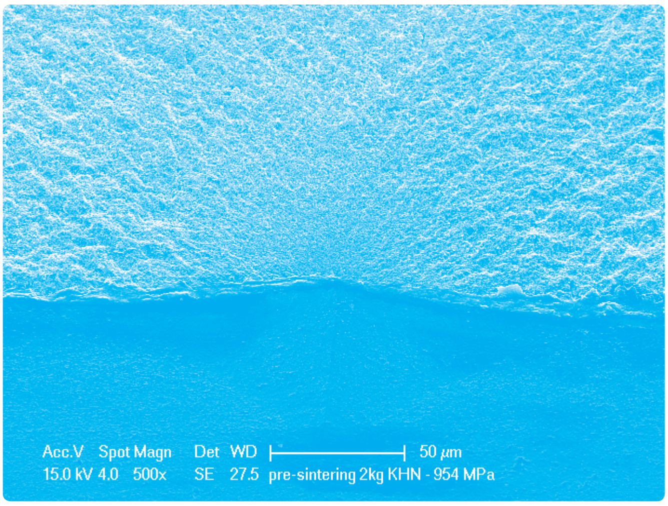

Figure 2b: SEM image, 500x, demonstrating smaller fracture origin of airborne particle abraded specimens as tetragonal monoclinic transformation did not contribute to crack resistance mechanism

Figure 3a: SEM image, 250x, demonstrating alumina particles shape before modification

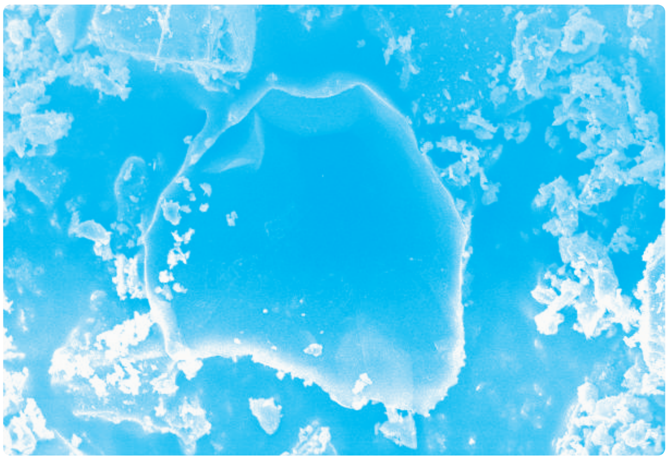

Figure 3b: SEM image, 300x, demonstrating alumina particles shape after modification. Notice roundation of particle edges

DISCUSSION

Partially stabilized zirconia (3mol Y-TZP) has a precise ratio of the stabilizing element which is sufficient to maintain tetragonal phase at room temperature but not sufficient enough to prevent tetragonal monoclinic transformation upon mechanical stimulation resulting in marked increase in fracture toughness compared to fully stabilized zirconia.[8] As monoclinic crystal phase is slightly larger than the tetragonal phase, it tends to occupy more volume in the bulk of the material resulting in compressing propagating cracks; thus enhancing resistance to crack propagation. Several studies evaluated the fracture toughness of different zirconia frameworks and reported a value ranging between 4.1 and 13.6 MPa.m^0.5 using different methods.[9-12] This partial state of stability allows tetragonal monoclinic transformation of surface crystals when subjected to mechanical stimulation in the form of grinding, airborne particle abrasion, or surface friction.

Tetragonal monoclinic surface transformation was observed after airborne particle abrasion of zirconia frameworks[13] and after micro-indentation using diamond points.[14] The detected percent of monoclinic phase ranged between as low as 0.3 and 10% and increased after artificial aging.[8,15-19] Strained or as called rhombohedra crystals were also reported.[20] The accompanying volumetric expansion (4%) resulted in grain pull-out and increase in surface roughness. Surface transformation was a factor responsible for delamination of the veneer ceramic and bond disruption with adhesive resins.[21]

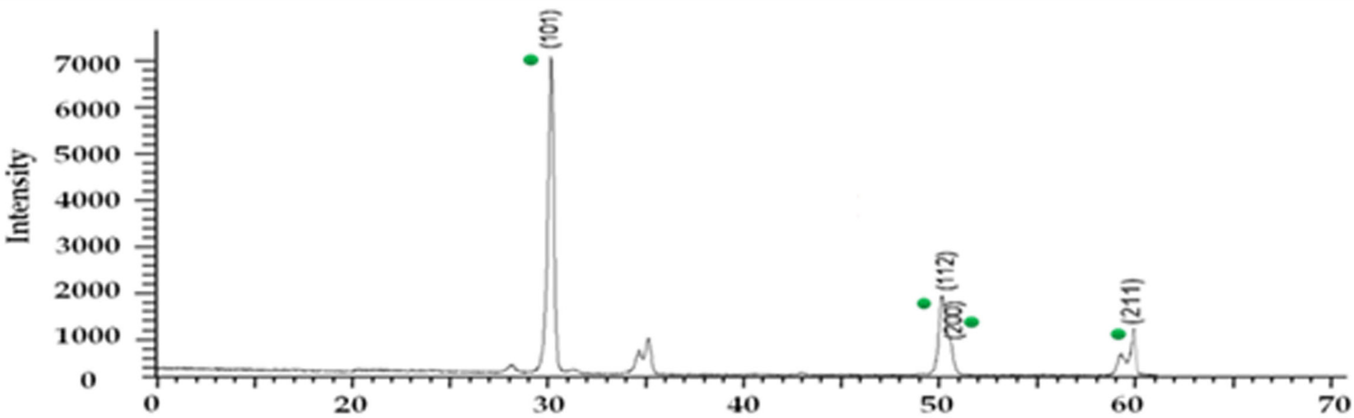

Figure 4: XRD of control specimens showing tetragonal crystal peaks

Similar observations were reported in the present study. Low energy surface activated specimens had significantly higher biaxial flexure strength compared to the other specimens (almost 50% increase in BFS). The amount of detected monoclinic crystals for these specimens was much higher (7.2%) compared to 30 µm and 50 µm particle abraded specimens which could explain the marked increase in biaxial flexure strength observed for these specimens. However, the higher content in monoclinic phase of low energy surface activated specimens is not the only factor responsible for the significant increase in flexure strength. Low energy surface activated specimens had a surface roughness value comparable to as-sintered and polished specimens indicating absence of marked surface defects associated with this surface treatment *(Figure 1b)*. Rotational grinding of the used particles resulted in removal of their sharp edges reducing the amount of surface damage. At the used pressure, distance, and time these particles were able to initiate monoclinic transformation without creation of marked surface damage which was observed with conventional alumina particles *(Figure 1c)*. The proposed hypothesis was accepted.

A strong correlation was established between degree of surface damage, flexure strength, and failure probability of zirconia specimens. In previous studies, coarse airborne particle abrasion using 120 µm alumina particles reduced flexure strength of zirconia to almost half (600 MPa) in addition to marked reduction in the calculated survival statistics (Weibull modulus) of these specimens. Despite a marked increase in monoclinic phase associated with coarse particle abrasion, the amount of induced surface damage resulted in stress concentration at deep and sharp surface cracks and scratches causing a marked reduction in mechanical properties of the abraded specimens. [3,22]

Fractographic examination of fractured low energy particle abraded specimens revealed that the critical crack was located few microns below the surface. This could be explained by creation of compression zone on the surface associated with monoclinic transformation *(Figure 2a)*. On the other hand, the critical crack of specimens abraded with conventional particles was located at the depth of sharp cracks located on the surface. *(Figure 2b)* [8,21,23,24]

Reverse transformation by annealing heat treatment resulted in complete reverse transformation of the monoclinic crystals created by low energy surface activation technique causing complete loss of the gained strength[20] back to the value of polished specimens (theoretical strength of zirconia). Such finding clearly attributes the marked increase in flexure strength to the high monoclinic content associated with low energy surface activation surface treatment.

Low pressure surface activation of zirconia restorations is a very simple procedure that requires utilization of modified alumina particles in standard airbone particle abrasion machines. The technique could contribute to modifying surface structure of zirconia and increasing its mechanical properties without compromising fatigue life by introducing surface defects.

CONCLUSIONS

Within limitations of this study, low energy surface activation of zirconia significantly increased its flexure strength without inducing marked surface damage. The technique could be used to modify the surface structure of sintered zirconia restorations in place of the more damaging particle abrasion and grinding techniques.

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