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Strength and Reliability of Porous Ceramics Measured by Sphere Indentation on Bilayer Structure

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ABSTRACT

The importance of porous ceramics has been increasingly recognized and adequate strength of porous ceramics is now required for structural applications. Porosities of porous ceramics act as flaws in inner volume and outer surface which result in severe strength degradation. The effect of pore structure, however, on strength and reliability of porous ceramics has not been clearly understood. We investigate the relationship between pore structure and mechanical properties using a sphere indentation on bilayer structure, porous ceramic top layer with soft polymer substrate. Porous alumina and silica were prepared to characterize the isolated pore structure and interconnected pore structure, respectively. The porous ceramic with 1mm thickness were bonded to soft polycarbonate substrate and then fracture strengths were estimated from critical loads for radial cracking of porous ceramics during sphere indentation from top surface. This simple and reproducible technique provides Weibull modulus of strength of porous ceramics with different pore structure. It shows that the porous ceramics with isolated pore structure have higher strength and higher Weibull modulus as well, than those with interconnected pore structure even with the same porosity.

Key words : ***Bilayer, Porous ceramics, Strength, Reliability, Hertzian indentation***

1. Introduction

Porous materials have useful properties such as low density, high permeability and large specific surface area and the advantages of ceramic materials are high wear resistance, hardness, thermal shock and corrosion resistance. Porous ceramics, combination of porous metals or polymers with ceramics for synergistic properties, have been investigated through past two decades in various fields such as bio-ceramics, environmental ceramics, and structural ceramics. The porous ceramics have to keep structural reliability for practical use but it has not yet been made clear whether porous ceramics have low reliability due to low strength. It is widely believed that fracture of brittle dense ceramics at far below their theoretical strength is due to the presence of defects. Moreover, when pores or flaws are added intentionally in ceramics, they influence their mechanical properties.1-6) Gibson and Ashby7) have suggested open-cell and closed-cell models to explain the mechanics of foams. However, the mechanical properties of porous ceramics are not fully understood except simple cellular solids and it seems to be a worthwhile to investigate.

Previous research has demonstrated that elastic modulus depends on pore structure rather than porosity8) and Hertzian indentation can be used to measure the elastic modulus and to reveal the contact damage of porous ceramics.9) In this study, we investigated the relationship among strength, reliability10,11) and pore structure using bilayer flexural strength test developed from Hertzian indentation. Various reports have been published on the strength of porous ceramics using biaxial strength test, because it can be used to avoid tensile loaded edges.

The Weibull distribution has been widely used to describe the fracture behavior of the ceramics. The Weibull distribution of strength is based on the weakest-link theory, which means the most serious flaw controls the strength. The reliability of porous ceramics with homogeneous pore distribution is considered to be an important subject. A number of specimens are required to get a Weibull diagram, therefore bilayer flexural strength (uniquely simple method and reproducible specimens) is introduced in this study. Bilayer flexure strength test has come to occupy an important position in laminate structures that are formed with brittle outer-layers (typically hard ceramics) to shield soft or compliant supporting under-layers (metals, polymers, or even soft ceramics).12-16) However, no application of bilayer flexure strength test to porous ceramics has ever been reported in the literature.

In the present work we prepared porous ceramics/polycarbonate bilayer systems, and porous alumina and silica were selected as model porous ceramics to reveal the effect of pore structure on the strength and reliability. Fugitive spherical PMMA was used to fabricate porous alumina of isolated

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pore structure with controlled porosity. PMMA was profitable because spherical pore was more convenient to study the effect of pore structure, shape, and size than irregular pore that was induced by naphthalene, starch or carbon in the past. Porous silica was fabricated with controlled sintering temperature which allowed to have an interconnected pore structure.

2. Experimental Procedure

2.1. Preparation of Porous Ceramics with Controlled Pore Structure

Starting powders were prepared comprising a mixture of alumina (AKP-50, Sumitomo, Tokyo, Japan) and PMMA (Aldrich, Milwaukee, WI, USA) in 2-distilled water. The slurries were ball-milled without milling balls for 5 h and then oven-dried at 60°C for 24 h. Green bodies were formed in two kinds of shapes which were rectangular specimens (length 30 – 32 mm, width 5 – 6 mm, thickness 2 – 3 mm) for four-point flexural strength test and plate specimens (length 20 mm, width 10 mm, thickness 0.8 – 1.2 mm) for bilayer flexural strength test. The specimens were prepared by Cold-Isostatic Pressing (CIP, Autoclave Engineers, Erie, PA, USA) at 200 MPa and then sintered at 1600°C for 5 h using box furnace in air atmosphere. Processing method was removal of fugitive material added to the body, and alumina and starch were body and fugitive material, respectively. In sintering process, burn-out of PMMA occurred during sintering. When PMMA burned out, heating rate was 50°C/h to prevent evaporated PMMA combustion gas from remaining in matrix. Mixtures of 1 μm and 10 μm size silica were prepared to fabricate porous silica. To prepare the specimens for Scanning Electron Microscope (SEM, XL30, Philips Electronics N. V., Eindhoven, The Netherlands) observation, standard fracture-surface observation techniques were employed. Densities were measured by Archimedes method and porosities were calculated from the bulk density and the theoretical density. Elastic modulus measurement technique used was impulse excitation of vibration method (ASTM E1876-1).[17] Although pulse-echo method is widely used, impulse excitation method for highly porous ceramics is more convenient and on small-sized specimens.

2.2. Measurement of Strength and Weibull Modulus by Bilayer Structure

If the thickness of upper-layer is thin, flexural stress becomes dominant, and radial cracks initiate at the tensile undersurface. The maximum tensile stress at the center of the undersurface of upper-layer is determined from the theory of plates on elastic foundations.

$$P_R = B\sigma_c d^2/\log(CE_c/E_s) \quad (1)$$

Where, P_R is critical load for radial cracking, B and C are dimensionless constants, E_c/E_s is modulus ratio of upper-layer and substrate and d is thickness of upper-layer. From Fig. 1, it shows the schematic illustration of flexural strength measurement of porous ceramics with bilayer indentation configuration. Porous alumina and porous silica were subjected to top-surface contact loading from tungsten carbide indenting spheres (radius of 1.98 mm, 3.57 mm) mounted under the crosshead of a mechanical loading machine (Model 4400R, Instron Corp, Canton, MA, USA). The crosshead was lowered until the sphere contacted the top surface of the specimen and the displacement was then controlled to a constant load rate of 0.01 mm/s to a specified peak value. Several tests were performed on every specimen over a load range of 100 – 1000 N.

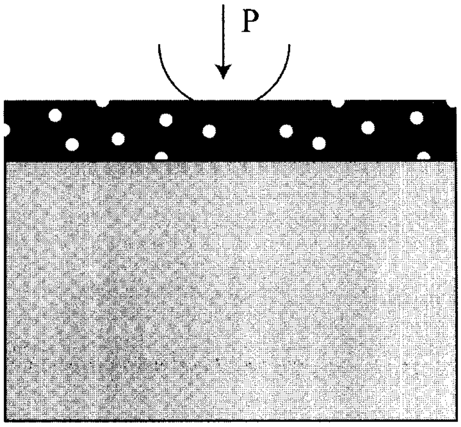


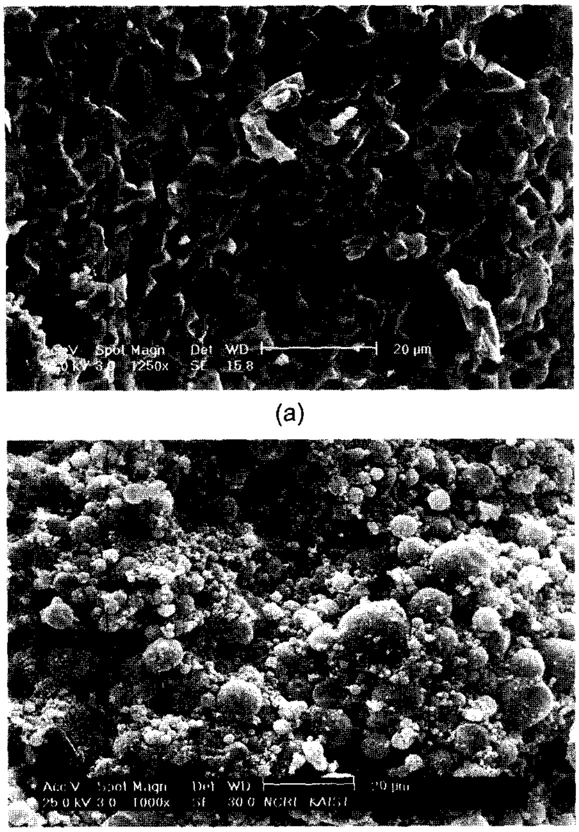
Fig. 1. Schematic illustration showing flexural strength measurement of porous ceramics with bilayer indentation configuration.

In Weibull diagram, Failure probability, P_f, obtained by bilayer flexural strength is plotted against strength and the slope shows Weibull modulus clearly. P_f was not expressed as a function of ln [ln 1/(1 – P_f)], because it could not show direct relationship between slope and Weibull modulus.

3. Results and Discussion

3.1. Microstructure and Porosity

Porosity was controlled as a function of the amount of fugitives. Increasing the amount of fugitives from 0% to 5% by 1.25%, the porosity increased linearly from 1.33% to 19.40%. Target porosity range was planned to be less than 20% to avoid pore clustering that can bring about additional effects. Fig. 2(a) shows fracture surface SEM image of porous alumina with isolated pore structure. Well-defined pores were formed in alumina matrix. It shows that homogeneous spherical pores surrounded by a dense alumina matrix and spherical pores had the same size with those of PMMA particles in alumina matrix. We fabricated porous silica with inter-connected pore structure by sintering of silica at different temperature, which was used to compared with the strength and reliability of isolated pore structure. Porous silica includes interconnected pore structure as shown in Fig. 2(b). Well-defined interconnected pore structure was fabricated up to 30% porosity successfully.



(a)

(b)

Fig. 2. SEM images of the fracture surface of specimens (a) porous alumina with 4.7% and (b) porous silica with 18%.

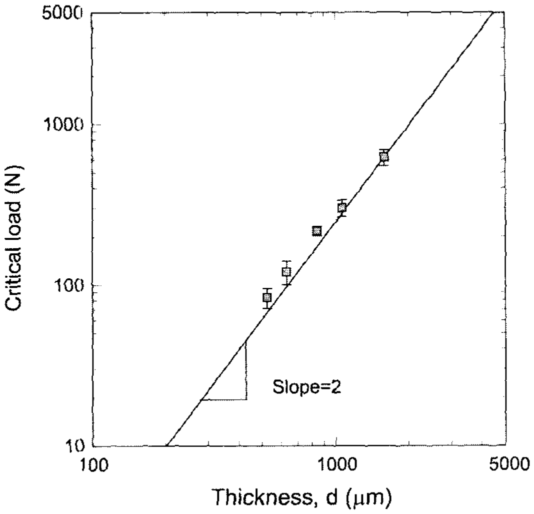


Fig. 3. Critical loads for porous ceramics/polycarbonate bilayer as function of thickness d.

3.2. Strength and Reliability of Porous Ceramics

The measurements of critical loads as function of thickness were carried out with the apparatus previously described. We may expect that the critical load data of the radial cracking can be used to confirm the d^2 dependency of critical load for radial cracking, P_R. In Fig. 3, the slope is the same as derived in Eq. (3) and the points for critical load do not deviate from the slope line. Based on the facts that have been clarified so far, we reasoned that the strength of porous ceramics also can be obtained from bilayer flexural strength test. Making good use of the merits of bilayer flexural strength test, multiple data acquisition from one specimen is necessary. In general, the length of radial crack growth was greatly affected by the thickness of specimen. However, the difficulties of detection of critical load also increased as the thickness of specimen increased, as the determined optimal thickness of specimen is about 1 mm.

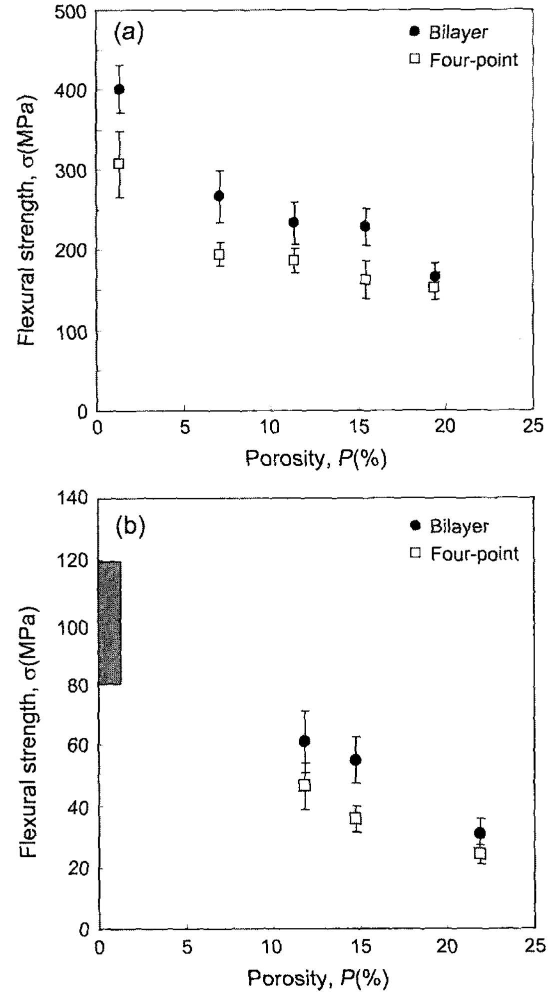


Fig. 4. Comparison of bilayer flexural strength and four-point flexural strength of (a) porous alumina and (b) porous silica (Shaded area at left indicates failures from natural flaws).

Fig. 4(a) shows the comparison of bilayer flexural strength and four-point flexural strength of porous alumina. It should be pointed out that porous alumina was fabricated to characterize isolated pore structure. We measured the strength of porous ceramics by bilayer configuration and by four-point flexural configuration as a function of porosity such as 1.3, 7.1, 11.3, 15.4, and 19.4% bilayer flexural strength was higher than 4-point flexural strength. The factors affecting it could be minimized edge effect as like biaxial flexural strength test. Once flaw was added, strength degradation is drastic but as porosity increased, strength decreased gradually. Comparison of bilayer flexural strength and four-point flexural strength showed slight difference and it seemed that minimized edge effect started saturation at 19.4% porosity. Fig 4(b) shows the comparison of bilayer flexural strength and four-point flexural strength of porous silica and shaded area at left indicates failures from natural flaws. Although porous silica was prepared to characterize interconnected pore structure, porous alumina with isolated pore structure and porous silica with interconnected pore structure have similar strength degradation trends. Bilayer flexural strength was higher than 4-point flexural strength at 11.8, 14.7, 11.3, and 15.4% porosity. So, it is believed that edge effect could be greatly reduced by using bilayer flexural strength test independent of pore structure.

Weibull diagram of bilayer flexural strength for porous alumina is shown in Fig. 5(a). Weibull modulus difference between 1.3 and 7.1% porosity was relatively high, but Weibull modulus did not show a linear relation with porosity when porosity was higher than 7.1%. It is obvious that porous ceramics had low strength than fully dense ceramics, but the porosity increase did not coincide with reliability decrease and it means that porous ceramics could have high reliability regardless of low strength. In Fig. 5(b) Weibull diagram of flexural strength for porous silica is shown. The difference between fully dense silica and porous silica was higher than the difference in case of porous alumina. Porous silica had lower reliability than porous alumina and the reason for this is they had different pore structure. When the specimen had isolated pore structure, higher elastic modulus and yield point was obtained than that when it had interconnected pore structure in previous research.[9] Also it is believed that strength of porous ceramics with interconnected pore structure was more sensitive to porosity than that of isolated one. It was due to the fact that inter-connected pore structure had minimum contact area between particles and only necks were formed between particles. This study shows that Weibull modulus was not strongly related to porosity, but pore structure. If we change the substrate material and upper-layer porous ceramics with other materials of different elastic modulus and isolated pore structure, having different pore size, respectively, some unclear points about the application of bilayer flexural strength test to porous ceramics will be answered. This is the subject for a future study and will be discussed in a separate paper.

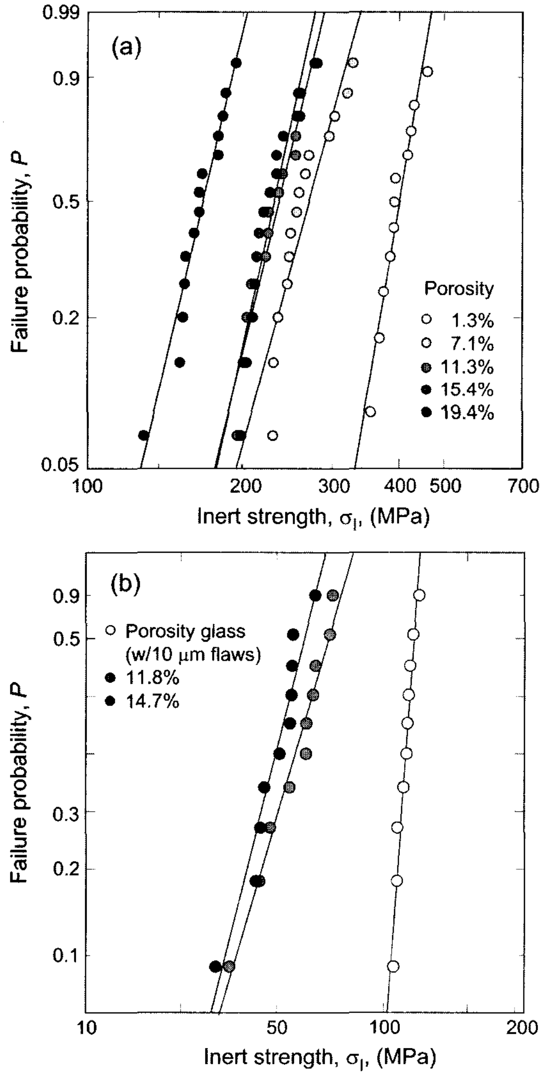


Fig. 5. Weibull diagram of flexural strength for (a) porous alumina and (b) porous silica.

4. Conclusions

The strength of porous ceramics bilayer systems (porous ceramic/soft dense substrate) were obtained from Hertzian indentation, which allowed uniquely simple and reproducible experimentation. Porous alumina and porous silica were prepared to characterize the isolated pore structure and interconnected pore structure, respectively. We have measured the strength of porous ceramics by bilayer configuration and by four-point flexural configuration. Porous ceramics with isolated pore structure has higher Weibull modulus(m), which represents higher reliability, and higher strength than porous ceramics with interconnected pore structure at the same porosity.

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