SUPERPLASTIC FORMING OF CERAMIC INSULATION

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INTRODUCTION

The forming of ceramics is generally difficult because the melting points of ceramics are relatively high and, consequently, the temperatures required to thermally activate plastic deformation in ceramics are also high. In addition, the propensity for grain boundary separation in ceramics is well known. In the 1950s, extensive efforts were made in the western world to hot fabricate ceramics using conventional metallurgical processes such as extrusion, rolling, and forging [1-5]. (It is of interest to note that there was also some evidence indicating that a comprehensive Soviet activity was underway to improve ductility and fabricability in complex ceramics and ceramic composites [6,7].) The goal was to produce near-net-shape parts in order to avoid the expensive machining of ceramics. A number of structural oxides, including CaO, MgO, SiO_2, ZrO_2, BeO, ThO_2, and Al_2O_3, were studied [2]. As a result of this work, an improved understanding of ceramic deformation was developed but certain problems, and in particular the requirement for relatively high forming temperatures, still existed. For example, the temperature required for hot forging Al_2O_3 was found to be about 1900°C which is extremely high from a practical standpoint. Subsequently, the concept of thermomechanical processing of ceramics was more or less abandoned.

Recently, two major technical advances have changed this picture. First, ceramic powder processing technology has been greatly advanced and the quality of ceramic powders is greatly improved. High-purity ceramics of submicron grain size and more consistent microstructures are routinely prepared. Secondly, fine-grained superplasticity in metallic alloys has been extensively studied and has already found commercial applications [8]. Because of these two advances, together with the observation that fine-grained ceramics generally possess the microstructural prerequisites for superplasticity in metals, the concept of hot forming, and in particular, superplastic forming of ceramics has become an area of intense study.

Since about 1986, many fine-grained polycrystalline ceramics have been demonstrated to be superplastic. These include yttria-stabilized tetragonal zirconia polycrystal (Y-TZP) [9,10], Y-doped Al_2O_3 [11], β-spodumene glass ceramics [12], ZnS [13], and Al_2O_3-reinforced Y-TZP (Al_2O_3/YTZ) [14,15] and SiC-reinforced Si_3N_4 (SiC/Si_3N_4) [16] composites. Among these materials, a 3 mol% yttria-stabilized tetragonal zirconia polycrystal (3Y-TZP) was the first true polycrystalline ceramic demonstrated to be

superplastic. The material is brittle at low temperatures, but becomes ductile at T > 1000°C. Its tensile ductility depends critically on temperature rather than on strain rate at low temperatures. Tensile elongations of only 5 and 60% were measured at 1000 and 1200°C, respectively, but elongations of greater than 100% were routinely obtained at temperatures above 1350°C. A maximum value of elongation-to-failure of 800% has been recorded in a sample tested at 1550°C and a strain rate of 8.3 x 10^{-5} s^{-1}. A direct comparison of this superplastically deformed sample with an untested sample is given in Fig.1. It is evident that macroscopic necking does not occur until near the final fracture of the test sample. Superplastic flow in 3Y-TZP has been characterized as a diffusion-controlled process and the strain rate, $\dot{\epsilon}$, can be expressed as [17]

$$\dot{\epsilon} = A \cdot \frac{1}{d^3} \cdot \left[\frac{\sigma}{E} \right] \cdot \exp\left[-\frac{510,000}{RT} \right] \tag{1}$$

where σ is the flow stress, E is the elastic modulus, d is the grain size, A is a material constant, R is the gas constant, and T is the absolute temperature.

In the area of superplastic forming, so far, several parts have been successfully formed from fine-grained 3Y-TZP. For example, Kellett et al. [18] demonstrated that submicron-sized (~0.23 μm) 3Y-TZP powders can be extruded superplastically, with an 8 to 1 reduction in area ratio (or a true strain of -2.2), at 1500°C to near full density; this is illustrated in Fig.1. Panda et al. [19] and Yamana et al. [20] have applied a sinter-forging technique to form bulk ZrO_2 stabilized by various amounts of Y_2O_3 from fine powders to full density at 1400°C. Recently, Wakai et al. [21] successfully bent a 3Y-TZP sheet sample to a large strain at 1450°C in air using SiC tools. Wu and Chen [22], on the other hand, demonstrated that a 2Y-TZP containing 0.3 mol% CuO can be biaxially stretched into hemispheric dome shapes at 1150°C, using a tungsten punch. This low temperature forming was a direct result of the decreasing melting point of the grain boundary glassy phase caused by the CuO addition. It is worth noting that Kellett and Lange [23] have successfully hot forged a 20 vol% ZrO_2/Al_2O_3 to 60% reduction in thickness at 1500°C.

The diffusion bonding technique has also been demonstrated successfully in Y-TZP and its composites. For example, Nagano et al. [24] and Wakai et al. [25] successfully performed diffusion bonding between Al_2O_3/YTZ composites and 3Y-TZP at the superplastic temperatures, i.e., 1450-1550°C, for the parent materials. The bond strength was found to depend on the bonding temperature and the compositions of the parent materials (similar and dissimilar). A maximum bond strength of greater than 1300 MPa was recorded in a 20%Al_2O_3/YTZ composite diffusion bonded at 1470°C [24].

The purpose of this paper is not to discuss general superplasticity in ceramics *per se*, but to present the first results of studies involving the free, biaxial, gas-pressure deformation of the superplastic 3Y-TZP and 20%Al_2O_3/YTZ composite. Additionally, the purpose is to address the technological implications of this

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superplastic forming technology and its potential applications to the forming and shaping of various ceramics, particularly ceramics used for thermal insulation, such as zirconia and its composites.
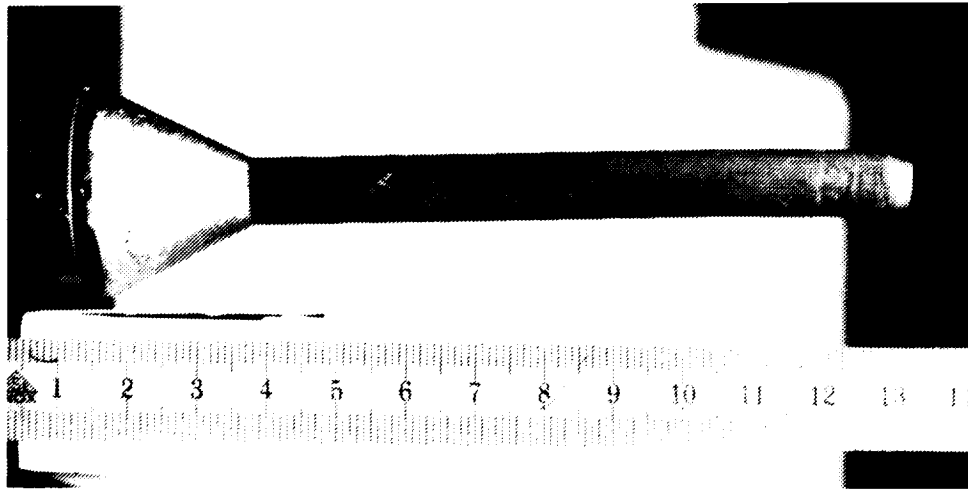

Fig. 1   Fine-grained Y-TZP extruded at 1500°C with a reduction in area ratio of 8. (from Ref. 18)

EXPERIMENTAL PROCEDURE

The materials used in this study were a fine-grained 3Y-TZP and its composite containing 20 wt%Al_2O_3 (Al_2O_3/YTZ); both were obtained as 50 mm diameter discs, 1.5 mm in thickness, from Nikkato Corp., Japan. The microstructure exhibited an equiaxed grain morphology. The grain size of 3Y-TZP was about 0.3 μm, while it was about 0.5 μm (both zirconia and alumina grains) in the composite. Data regarding the microstructure, superplastic flow properties, grain growth behavior, and cavitation characteristics of these two materials (under uniaxial tension conditions) have previously been reported [10,26,27]. Building upon information from these previous studies, a gas-pressure forming apparatus was constructed with the capability to operate at temperatures of as high as 1700°C at gas forming pressures of as high as 2.5 MPa. A two-part die was designed and incorporated to impart a clamping force to the periphery of the ceramic sheet permitting an unconstrained diaphragm diameter of 38 mm. Deformation of the diaphragm was accomplished through the controlled application of Ar as a forming gas. An ambient pressure of 3×10^4 Pa of Ar was maintained during all forming experiments. In order to accomplish the on-line sensing of the progress of the diaphragm deformation, a high-temperature contact LVDT was specially constructed and integrated into the forming apparatus.

RESULTS AND DISCUSSION

Generally, gas-pressure biaxial forming uses one of two experimental techniques to achieve the desired deformation- constant applied pressure or constant applied stress. The former technique of utilizing constant

applied gas pressure has been used for the study of other superplastic alloys [28,29]. With this method, both the flow stress and strain rate in the deforming shell change continuously during the experiment. Thus, it is difficult to interpret data from these tests in terms of basic materials properties. From a practical point of view, however, the constant applied gas-pressure technique is probably more applicable to a manufacturing environment than is the constant applied flow stress test. In contrast to this technique, an alternative method of constant applied flow stress in the deforming shell has also been used for forming Ti-based alloys [30]. The principal advantage of this technique is that it allows for the direct comparison of biaxial deformation behavior with data obtained from uniaxial tension testing. (Most studies of superplastic deformation involve the application of a constant strain rate or constant flow stress in uniaxial tension or compression.)

In the present study, experiments were conducted under conditions designed to approach that of a constant applied flow stress in the deforming shell. The deformation process was accomplished through a consideration of the pressure-curvature relationship for a spherical shell (as shown in Fig.2):

$$\sigma = \frac{P\,r}{2\,t} \qquad (2)$$

where σ is the flow stress, P is the applied gas pressure, r is the instantaneous radius of curvature, and t is the instantaneous shell thickness. It is noted that during the course of an experiment, P is varied as a function of r (and therefore t) in order to achieve a predetermined value of σ in the shell. The thickness t was assumed to vary uniformly over the deforming shell and to decrease steadily in a manner which was calculated assuming a constant volume. (Although expedient for the design of this initial series of experiments, the uniform thickness assumption is not rigorously correct.) The height of a deforming hemispherical cap may be related to its radius through a consideration of the cap geometry, as schematically shown in Fig.2. A hemispherical cap of apex height h and having a base radius of r_o may be considered as a section of a sphere of radius r and included angle 2α. The included half-angle α may be determined by measuring the height h of the cap:

$$tan\left(\frac{\alpha}{2}\right) = \frac{h}{r_o} \qquad (3)$$

The radius of curvature r may then be determined as

$$r = \frac{r_o}{sin\alpha} \qquad (4)$$

By preselecting a shell stress for a particular experiment, the values of Ar gas pressures required to compensate for the changing dome height and shell thickness during deformation could be adjusted accordingly.
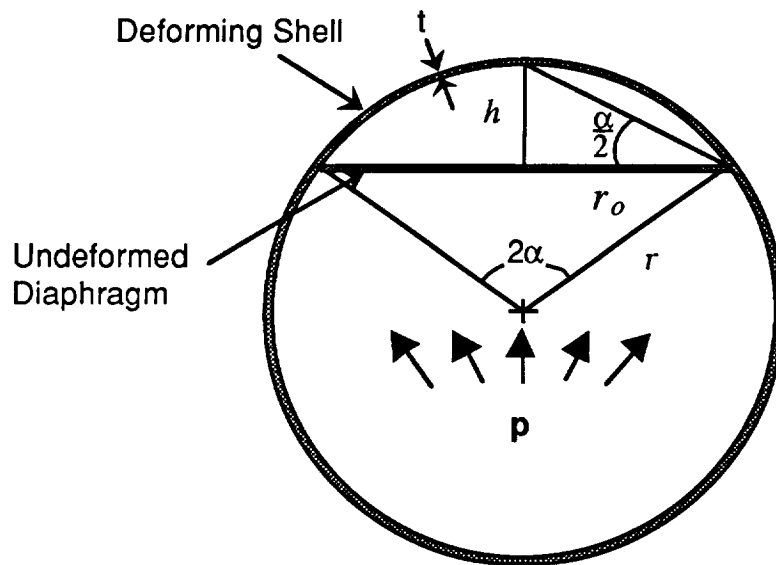
Fig.2 A hemispherical cap of height h and basal radius r_o may be considered to be a section of a sphere of radius r and included angle 2α.

The result of one series of experiments to demonstrate the formability of the Al_2O_3/YTZ sheet is shown in Fig.3. Interrupted tests illustrating the progressive deformation of the Al_2O_3/YTZ at various strain levels are shown at an applied shell stress of 13.8 MPa for tests conducted at 1475°C. (Similar results were also obtained for the 3Y-TZP sheet.) A forming temperature of 1475°C was selected as a compromise between higher temperatures which would result in excessive grain growth rates, and lower temperatures which would result in unacceptably low strain rates. All shells in Fig.3 were noted to deform to symmetrical spherical sections with no evidence of inhomogeneous flow or fracture resulting from the imposed clamping pressure.
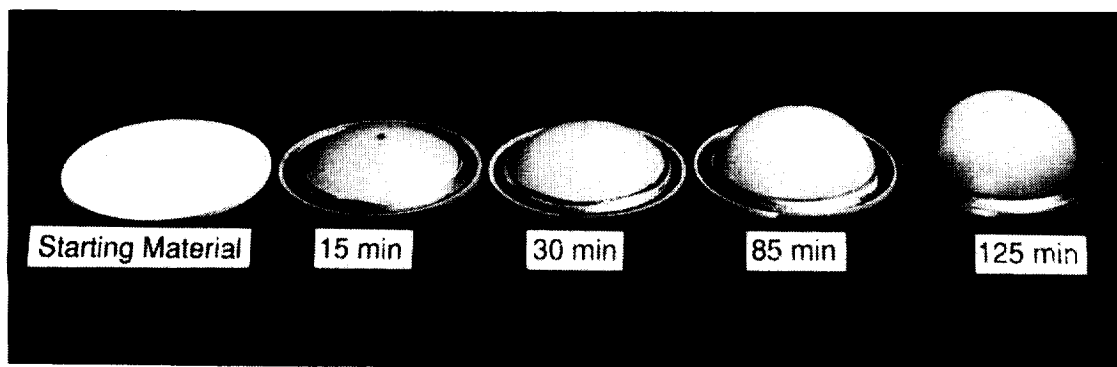

Fig.3 Progressive deformation of 20% Al_2O_3/YTZ at 1475°C and 13.8 MPa flow stress.

Experiments were also conducted with Al₂O₃/YTZ at 1475°C for different applied shell stresses. The apex height as a function of forming time for three applied stresses is shown in Fig.4. Since these experiments were carried out under constant-stress conditions, the curves in Fig.4 are similar to those observed during the creep of most metal alloys. (Strictly speaking, the constant-stress condition is only an approximation. This is a result of the fact that Eq.{1} is based on the assumption of uniform thickness which, in actuality, is not rigorously correct.) In terms of formability, all the above applied stresses produce good-quality hemispherical domes; applied stresses only affect the forming rates.
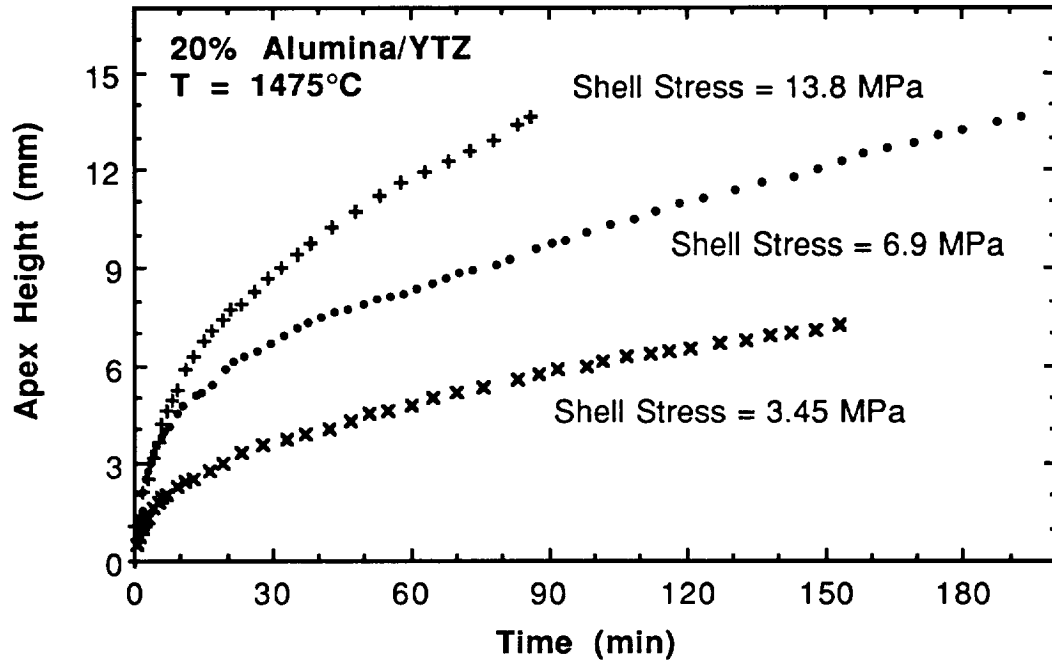

Fig.4   Apex height as a function of forming time for 20% Al₂O₃/YTZ at 1475°C and at various shell stresses.

To demonstrate further the versatility of the above gas-pressure forming technique, we also conducted experiments to form superplastic ceramics into shapes other than a simple hemisphere. One of the examples is given in Fig.5, which illustrates schematically the design of a forming die and the conceptual forming operation. Experiments were subsequently carried out with 3Y-TZP samples at temperatures between 1500, 1550, and 1575°C and at an applied gas pressure of 0.28 MPa (about 6.9 MPa nominal shell stress) to verify the above forming process. Shown in Fig.6 are three 3Y-TZP samples illustrating the progressive deformation of the 3Y-TZP gas-pressure formed at various strain levels. These samples fully demonstrate the excellent superplastic formability of 3Y-TZP sheet.

(It is worth noting that both deformed 3Y-TZP and Al₂O₃/YTZ samples appear dark. This is a result of the fact that the experiments were carried out in a vacuum-inert gas atmosphere and it is known that 3Y-TZP

darkens in vacuum, i.e., an oxygen-depleted environment. The expected white color can be easily recovered by annealing at 1200°C in air for 0.5 h.)
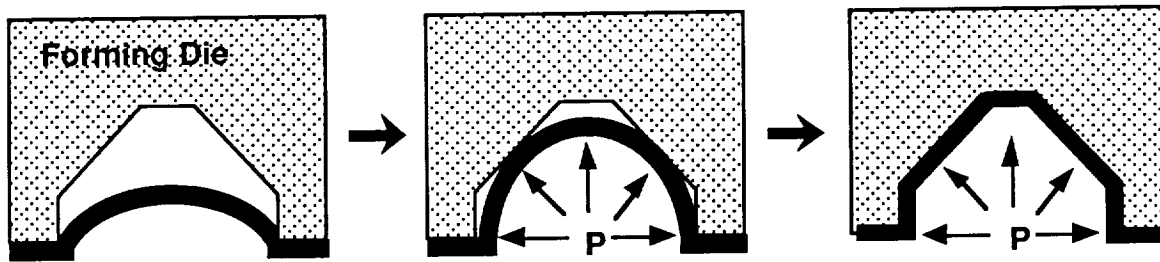

Fig.5 Schematic illustration of the gas-pressure forming process for superplastic ceramic sheet.
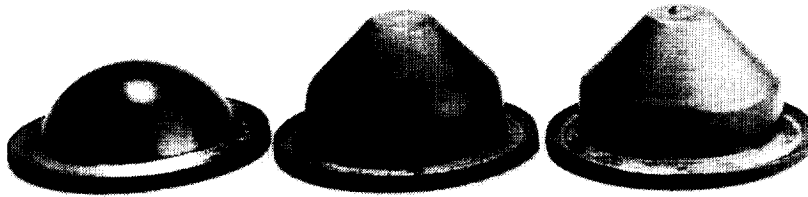

Fig.6 3Y-TZP samples progressively deformed, according to the diagrams shown in Fig.5, to various strain levels.

It is noted that the thickness of a gas-pressure formed sample is generally not uniform. This is because the center of a deforming diaphragm is in a stress state of equibiaxial tension (plane stress), while at the clamped periphery of the diaphragm there is a state of plane strain. When a diaphragm is deformed through the action of an applied gas pressure, the stress state varies between the apex and the periphery. As a result of this stress gradient, deformation occurs under a corresponding strain rate gradient. The degree of thickness variation is therefore closely related to the strain rate sensitivity of the deforming material. The development of an analytical model to describe the thickness distribution in gas-pressure formed samples is underway. Technologically, however, the problem of thickness nonuniformity has been encountered by the metal industries, and was overcome by a number of techniques, including a reverse forming operation [31].

PERSPECTIVE

Future hypersonic flight vehicles impose many distinct design restrictions in terms of vehicle size, weight, takeoff and landing characteristics, safety, and logistics. These requirements for hypersonic flight

created a unique challenge in the selection of fuels and thermal management. For example, the maximum speed for an aircraft, with a combined ramjet-turbojet propulsion, with the given range and weight requirements is most likely to be about Mach 5 [32]. The temperature of the ramjet combustor and engine nozzle wall for such a flight vehicle are expected to be near 4200°F; this is schematically shown in Fig.7.

HOT GAS (4200 F)

4000 F

915 F — Zirconia Tube Insulation

838 F — Strain Isolater

750 F — Cooling Panel (TI 6-4)

Honeycomb Panel (Sheet TI6242, Core TI 6-4)
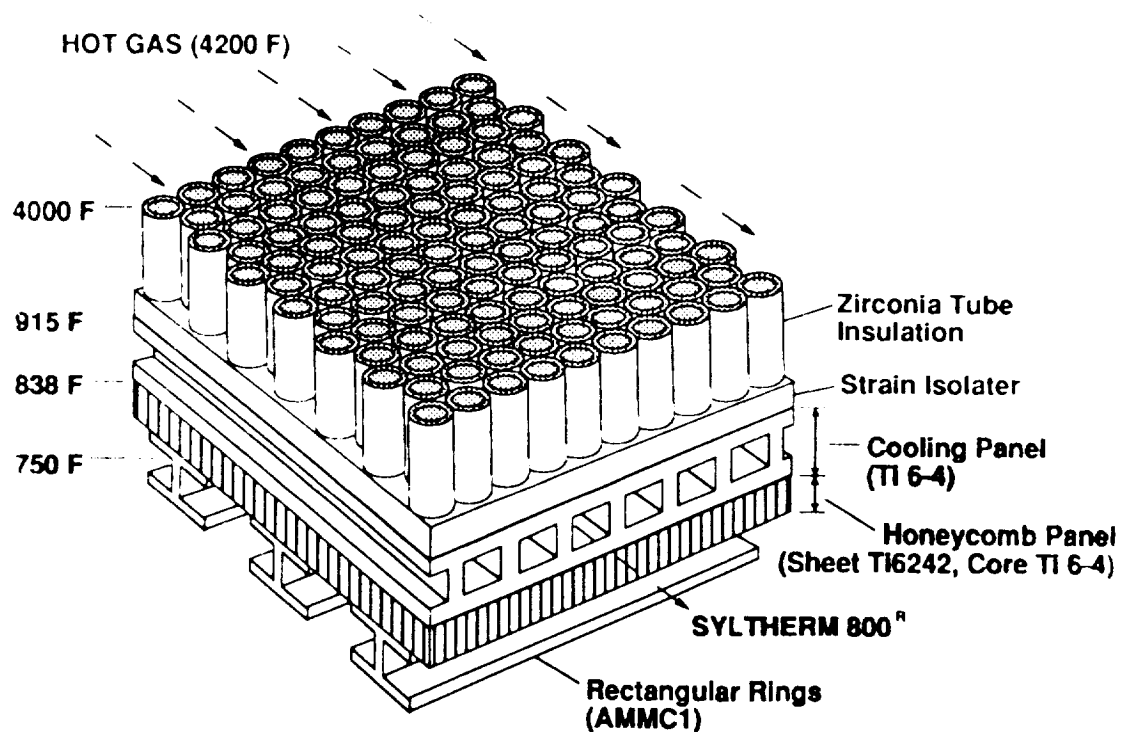
SYLTHERM 800ᴿ

Rectangular Rings (AMMC1)

Fig.7 Ramjet combustor and engine nozzle wall construction for a Mach-5 hypersonic aircraft with with a combined ramjet-turbojet propulsion. (from Ref. 32)

Zirconia is probably the only insulation material that is capable of withstanding such a high temperature. There is no other known insulation material which is designed to withstand exposure to gases at temperatures above 4000°F. The insulation in Fig.7 is proposed to consist of 1/8 inch O.D. zirconia tubes filled with a foam of the same material. This insulation design is a state-of-the-art concept.

Technically, zirconia tubes may be produced by conventional powder metallurgy and/or slip casting techniques which are primarily batch processes. This poses two major problems - product reliablity and cost. From both the cost and product reliability points of view, the process of superplastic extrusion of Y-TZP tubes using existing metallurgical facilities would be extremely attractive. In particular, this process has already been proven with fine zirconia powders [18]. Using the superplastic extrusion technique, zirconia tubes with given diameters can be produced in large quantities. The extruded tubes can be subsequently sliced into proper lengths for applications. In addition, since tube extrusion is a bulk process, product relaibility will be greatly improved and will certainly reduce the production cost significantly.

CONCLUSION

Superplasticity has been demonstrated in many fine-grained structural ceramics and ceramic composites, including yttria-stabilized tetragonal zirconia polycrystal (YTZP), alumina, and Al_2O_3-reinforced zirconia (Al_2O_3/YTZ) duplex composites and SiC-reinforced Si_3N_4. These superplastic ceramics obviously offer the potential benefit of forming net shape or near net shape parts. This could be particularly useful for forming complicated shapes that are difficult to achieve using conventional forming techniques, or require elaborate, subsequent machining. In the present study, we successfully demonstrated the following:

1) Superplastic 3Y-TZP and 20%Al_2O_3/YTZ composite have for the first time been successfully deformed into hemispherical caps via a biaxial gas-pressure forming technique.

2) No experimental difficulty was encountered in applying the required gas pressures and temperatures to achieve the results presented in this report. Thus, it is certain that higher rates of deformation than those presented in this study will be possible by using the current test apparatus at higher temperatures and pressures.

3) An analytical model incorporating material parameters, such as variations during forming in the strain rate sensitivity exponent and grain growth-induced strain hardening, is needed to model accurately and therefore precisely control the biaxial gas-pressure forming of superplastic ceramics.

Based on the results of this study, we propose to fabricate zirconia insulation tubes by superplastic extrusion of zirconia polycrystal. This would not only reduce the cost, but also improve the reliability of the tube products.

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