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Near Net Shape Production of Monolithic and Composite High Temperature Ceramics by Hot Isostatic Pressing (HIP)

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Abstract

Improved properties of high temperature ceramics in general are achieved by compositional and processing research and development—compatible with sintering and forming needs. Pressure can be used to increase the driving force for densification and with hot isostatic pressing (HIP) the form can be closely controlled, even of complex shaped parts, like turbine wheels. Recent development within the EUREKA-AGATA hybrid electric car gas turbine project shows that improved high temperature material properties can be achieved, while at the same time fabricating components like combustion parts and turbine wheels, to near-net-shape. For such components a highly uniform green powder body is desired. Combined with a type of encapsulation during HIP, which does not create shear stresses at the surface of the green body during the shrinkage/sintering to full density, but at the same time prevents penetration into the body, optimal near-net-shape results can be achieved. Recent studies, e.g. by TEM, have confirmed that some encapsulation glass constituents can form new compounds with silicon nitride, at the very surface, which appear to help develop these desired characteristics. Non-homogeneous and non-isotropic ceramics, like fiber reinforced composites, may be fabricated using rigid, shape controlling tools on one or several sides. Particularly for large (and curved) panels such use of a hot isostatic press can be an advantage. © 1999 Elsevier Science Ltd. All rights reserved.

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1 Introduction

Very strong interatomic bonds generally characterize high temperature (HT) ceramics. This can give the desired properties, in use, such as, resistance to chemical attack, high hardness, stiffness and strength, also at elevated temperatures and for prolonged periods of time. However, a consequence of the strong bond characteristic is that the ceramic components are often very difficult to fully densify and to machine to the desired final shape.

Pressure has traditionally been resorted to in order to help sintering and densification problems. Processing and compositional development could, however, in most cases provide acceptable solutions without the use of pressure. Such solutions should of course be used, if the production cost to manufacture the product with specified properties and form can be reduced. The traditional, uniaxial hot pressing (HP) has consequently, for cost reasons, in most cases been replaced, except, for example, in the production of whisker reinforced cutting tools. The very limited ability to make products with the required form is a major drawback. Production costs were, for example, reported to be cut by a factor of 10,[1] by a change from HP and finish machining of high-quality silicon nitride bearing balls, to glass-encapsulated HIP (hot isostatic pressing) of balls formed as green bodies.

Products of HT ceramics must, like any other component, satisfy two sets of specifications, namely fulfilling a set of *property specifications* (mechanical, chemical, thermal....) and fulfilling a set of *form specifications* of its exterior (shape with required tolerances, surface finish...etc, at different locations). In some cases, like for panels, HP can satisfy both property and form specifications, but more often only property requirements can be met and the form has to be achieved by finish machining.

HIP has in many cases been found to provide unique solutions for the densification and forming

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of ceramics. Important for HT ceramics is not only the 5–10 times higher pressure level than in HP but also the inherent ability of the HIP process to produce components with complex and precisely defined shape.

The purpose of this paper is to discuss HIP techniques for monolithic and composite HT ceramics and to present recent applications for silicon nitride turbine wheels and for combustion components of silicon nitride and oxide/oxide fiber CMC (ceramic matrix composites).

2 HIP for Near-Net-Shape (NNS)

The HIP process is schematically illustrated in Fig. 1. An inert gas (usually argon, for cost and availability reasons) is used to transmit pressure to the boundary of the porous body to be processed. The gas must be prevented from penetrating into any voids of the body that are to be sealed. Metals, ceramics or, most commonly, glasses may be used for such gas impervious encapsulation.

The gas pressure, typically 100–300 MPa, (even above 2000°C and in large size industrial equipment), is applied perpendicularly to—and with the same magnitude—on all accessible surfaces of the body. It provides a multiplication of the driving force for densification,[2] relative to the forces developed by the common sintering mechanisms related to surface tension on pore surfaces. In contrast to these, the contribution by the gas pressure to "effective pressure in particle contacts" (cf. Fig. 1), is *not* influenced by particle size.

In HIP generally, all the voids, particularly large ones,[3] can be efficiently reduced in size and/or appearance in most materials. Ceramics can, therefore, with the use of encapsulation, be densified at relatively low temperatures. A very high uniformity of properties as well as freedom from directionality can, if desired, be obtained. The absence of shear forces inside the body (on a scale substantially larger than particle size) and of any

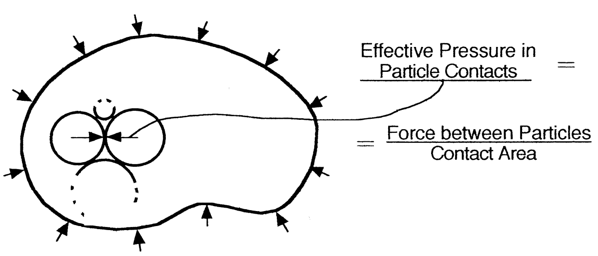


Fig. 1. Key components of the HIP process. A porous body (e.g. of powder particles) is subjected to an isostatic pressure (p) acting on an impervious gas barrier at its boundary at a temperature (*T*) for a time (*t*). As a consequence the "effective pressure in particle contacts" increases with (*p*) which at typical pressures greatly enhances densification and form-stability of the processed part.

die friction, combined with a pressure level up to an order of magnitude higher than in HP, makes HIP an even more powerful densification method. Even ceramics extremely difficult to sinter can be fully densified. High-purity silicon nitride powders without any sintering additives at all is one example, ceramic/ceramic composites with high loading of particles, whiskers or long fibers are another example.

A comprehensive discussion of different alternatives for HIP of ceramics can be found elsewhere.[4–6] Here only the HIP method, which we consider most suitable for HT ceramics, that is encapsulated (sometimes called direct) HIP, will be further discussed.

There are two main routes to produce dense products by encapsulated HIP; depending on if the green body for the product is mainly isotropic or not.

- An (on a meso-scale) isotropic green body can, by encapsulated HIP be fully densified with virtually no change of shape, just of scale.
- A grossly non-isotropic green body (e.g. a flat or curved panel of long fiber CMC) can, by encapsulated HIP, using rigid support be densified to an acceptable shape over large surfaces.

2.1 High-precision processing starting with isotropic green bodies

Encapsulated HIP can be used to produce parts with very accurate shapes. In order to obtain high precision of the final product, directional and spatial uniformity of the green powder body is needed as well as precision of size and shape of its boundary. In an *ideal HIP process, a pure isostatic pressure, only, should be exerted on the body boundary*, and the stress state in the body should *only* emanate from that and from sintering mechanisms. Gravity-less conditions and an impervious pressure gas seal at the body boundary, *creating no shear stresses*, would satisfy these criteria. Such demands on the process may seem unrealistic, but can nevertheless nearly be satisfied.

Too rigid encapsulation or gravity forces could cause distortion. Metal containers are usually rather rigid, particularly of alloys that could be used at the required temperatures for HT ceramics, and heavy distortion would result. If too rapid heating of the part forms a density front in the powder body while under pressure, similar effects appear. Large changes of shape may develop[7] but can be avoided by tailored heating rates.

With methods like injection molding for formation of the green powder body, (which can give very high uniformity and precision) and with glass powder encapsulation (which is flexible enough not to restrain the shrinkage of the ceramic powder body), it appears that almost ideal conditions can be achieved.[8]

A recent Ph.D. thesis[9] throws light on how glass can interact with a ceramic green body surface. Figure 2 shows a TEM image of crystalline boron nitride between grains of silicon oxynitride at the surface of a silicon nitride powder body. The origin of this BN is assumed to be a reaction between the boron oxide containing encapsulation glass and silicon nitride particles. Thermodynamic calculations show that BN can be thermodynamically stable already at the temperature of pressure application (1180–1200°C in this model experiment) and that silicon oxynitride would also be formed. If such boron nitride forms in former pore channels when the glass starts to penetrate the small pores, the flow will be hindered. A low viscosity glass, which would create only insignificant shear stresses on the surface of the ceramic body, could be used without risk for gross glass infiltration.

Fig. 2. TEM micrograph of textured, crystalline boron nitride formed near the original silicon nitride green body surface. The silicon nitride body has been facing encapsulation glass containing boron oxide during the HIP cycle. The apex of the wedge-shaped BN area points inwards. Adjacent grains now consist of silicon oxynitride.

In glass encapsulated HIP, the high gas pressure, when transmitted to the porous body via the encapsulation, makes the body extremely rigid and insensitive to external influence, like gravitational forces, which is particularly important in the initial stages of densification when bonds between particles are still very weak. The gas pressure which acts isostatically via the glass encapsulation on the boundary of the porous powder body has a very large influence on the *effective pressure in particle contacts* (Fig. 1). As a result, gravity induced tensile stress components, which could cause shape deformation, will become virtually negligible.[2] The powder body is in a typical glass powder encapsulated HIP kept in a very firm grip by the gas pressure acting on its boundary throughout its entire shrinkage and sintering process. The common vacuum packed plastic parcel of ground coffee may visualize the rigidity of the porous powder body. The ceramic powder is, however, much harder and the pressure difference over its boundary more than a thousand times higher (similar to the isostatic pressure at the largest depth found in the Pacific, the bottom of the Mariana Trench!).

2.2 Processing of panels of highly non-isotropic green bodies

The process discussed in Section 2.1 relies on the principle that a homogeneous green powder body without any directionality in properties is densified by just a reduction in scale but with no change in shape. An ideally "soft" encapsulation that still does not penetrate into the green powder body when high pressure is applied enables such an ideal HIP process. Often in practice there may, however, be density variations or some directionality in the body. A deviation from 'not changing the shape just changing the scale' will occur after full densification with a "soft" encapsulation. If the density profile or directionality can be consistently repeated then the fully densified products will also have a consistent shape. A desired product shape may then be obtained by adjusting the green body shape. This is a possible method also for products like cutting tool inserts of alumina with high loading of silicon carbide whiskers aligned in one plane.

A different principle must, however, be used for products like panels of long fiber CMC materials. Even if the positioning of long fibers in the matrix could be made with very high precision and symmetrical to both sides of a panel the probability is high that heavy distortion would occur. The panel would buckle and fiber breakage take place.

A method using rigid support tools has successfully been used for fabrication of combustion tiles with silicon carbide or single crystal oxide fibers and mullite or alumina matrix.[10,11] In these applications solid graphite tools were used on both sides of the tiles, molybdenum foil was used for sealing and the assembly was glass encapsulated and HIPed. Tiles with good shape and similar behavior in mechanical testing as tiles processed by HP at the same temperature and pressure, were obtained. Cost projections for the production of tiles by HIP were favorable in comparison with other available processes, like HP.

This HIP method for long fiber CMC using rigid tools should be advantageous also for very large and curved panels, and if it is desired to use higher pressures than those available in HP. Higher pressure offers the possibility of reducing the temperature, in order to avoid undesirable reactions.

3 Some Recent Applications for HIPed HT Ceramics

3.1 Components for the EUREKA-AGATA program

The goal of the European EUREKA project, EU 209, known as AGATA (Advanced Gas Turbine for Automobiles), is—to develop three critical ceramic components—a catalytic combustor, a radial turbine wheel and a static heat exchanger—for a 60 kW turbo-generator in an hybrid electric vehicle. These three components, which are of critical importance to the achievement of low emissions and high efficiency, have been designed, developed, manufactured and tested as part of a full-scale feasibility study, with a view to the eventual application of the concept in the automotive industry. AGATA is a joint project conducted by eight commercial companies and four research institutes in France and Sweden.

Silicon nitride ceramics play an important role both in the development of the catalytic combustor and the radial turbine wheel. AC Cerama, Sweden has developed a HIPed Si_3N_4 material designated CSN 101. This material has been selected for the catalytic combustion afterburner as well as for the radial turbine wheel. Mechanical properties of the CSN 101 Si_3N_4 have been found to be fully comparable with the best available high temperature Si_3N_4 materials.[12]

3.1.1 Catalytic combustor afterburner

A catalytic combustor provides an attractive, alternative means of achieving even leaner air/fuel mixtures and lower combustion temperatures than the best technology available today, the lean premix, pre-vaporization (LPP) concept. The aim is to further reduce thermal NO_x emissions. The concept of the catalytic combustor is developed further in the redefined AGATA project.

The main emphasis of the technical development program is on ceramic structural components and catalytic combustion. All of the ceramic components operate without cooling at 1350°C. The combustor housing is manufactured as rings by Aerospatiale, France, and made of SiC-whisker-reinforced Si_3N_4 by HP.

The exit cone is manufactured by AC Cerama using glass encapsulation and HIPing of the CSN 101 grade Si_3N_4. This component is at the prototype scale manufactured by cold isostatic pressing, followed by green machining, glass encapsulation and HIP. In a future larger production volume such a part could be formed by injection molding. The outer and inner surfaces of the combustor component are used with as-HIPed surface finish and only the contact surfaces are machined after HIP. Two afterburner cones are shown in Fig. 3, prior to delivery to Aerospatiale for catalytic section assembly. The rings and cone are joined by polished conical contact surfaces and the combustor is held together by externally mounted springs. The full scale testing of the combustor has been successfully completed. The afterburner cones showed no evidence of oxidation and withstood transient tests and steady state tests at 1350°C.

3.1.2 Turbine wheel

A fruitful cooperation between designers and ceramic manufacturers was established, generating a beneficial influence on design and material development activities. Interaction of this nature is of the utmost importance in designing and manufacturing a ceramic turbine wheel capable of operating at a high rotational speed and a turbine inlet temperature of 1350°C. CSN 101 Si_3N_4 is the material chosen for the radial turbine wheel. AC Cerama has developed a technique of combining two green-forming methods in a two-piece approach. The hub section of the wheel is formed by cold isostatic pressing (CIP) and green machining, while the blade-ring is injection molded, Fig. 4.

Fig. 3. Ceramic combustor exit cones of HIPed Si_3N_4, (CSN 101). The inner and outer surfaces are used with as-HIPed surface finish.

Following binder burnout, the hub and blade ring are green-joined, glass encapsulated and HIPed, the advantage being that binder burnout is achieved much more quickly and easily with two smaller components, than when the component is injection molded in a single piece. AC Cerama has also evaluated injection molding of the wheels as one piece. After some initial problems with large internal defects this route is now also a feasible way of producing good quality wheels, Fig. 5. Volvo Aero Turbines did the aerodynamic and mechanical design of the wheel. The rotor has been designed for high efficiency and with a fairly high tip speed. In a wheel efficiency test 84% was measured using a wheel of the final design. The overall geometry and blade thickness distribution has been selected in cooperation with the ceramic manufacturers. Efforts have been put into reduction of surface stresses at the blade roots. Maximum stress, which is around 290 MPa, occurs during the cold start transient. The ceramic wheel will be joined to the metal shaft by brazing developed by CEA/CEREM, France.

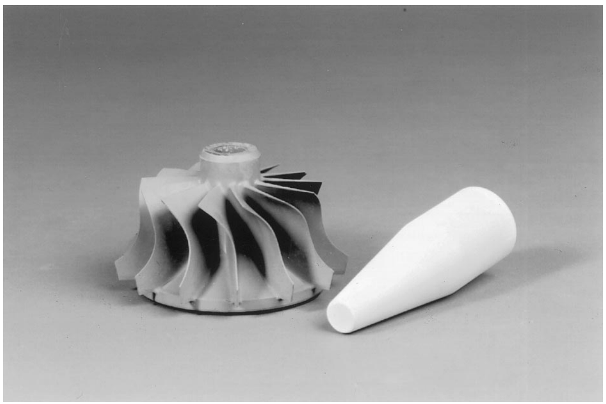
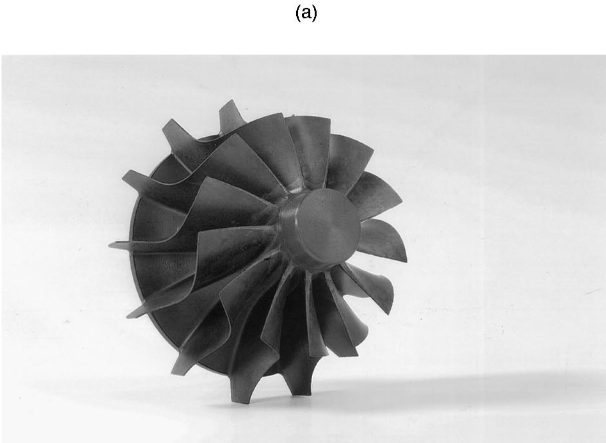


Fig. 4. Injection molded blade ring and cold pressed hub green bodies, prior to glass encapsulation and HIP.

(a)



(b)

Fig. 5. (a) and (b). Two views of turbine wheels made by AC Cerama, CSN 101 grade, HIPed Si_3N_4. Only the back face and stub shafts need to be finish machined.

Both spin discs and wheels of the final design have been cold-spin burst-tested at room temperature in vacuum. Hot-spin testing will be performed in a gas turbine rig under actual pressure, temperature and aerodynamic loading conditions. These tests will take the form of start/stop, low-cycle fatigue tests.

3.1.3 Properties of the HIPed silicon nitride (CSN 101) for afterburner cone and turbine wheel

The Si_3N_4 grade CSN 101 was developed by AC Cerama to meet the demands of high strength and fracture toughness coupled with good creep resistance at 1350°C and good thermal shock resistance. It is a 3 wt% Y_2O_3 composition based on high purity Si_3N_4 powder and can be green formed either by injection molding or by CIP. The densification is then performed using the proprietary glass encapsulation and hot isostatic pressing (HIP) technique discussed in Section 2.1.

Typical properties of this material are given in Table 1. Tensile creep testing is under way. Some of the results are presented in Table 2.

4-point flexural stepped-temperature-stress-rupture (STSR) tests going from 1000°C up to 1400°C have been presented previously.[13] Both test bars with machined surface, with as-HIPed surface (glass encapsulation) and with as-HIPed surface using a novel interlayer between glass encapsulation and silicon nitride have been tested. It was clearly seen that the new interlayer technique gives material with high temperature properties as good as the bulk material and at the same level as the best silicon nitride materials reported in the literature.[14,15]

Long term oxidation tests both in static air and in a high velocity gas burner have been performed

Table 1. Properties of AC Cerama CSN 101 Si_3N_4

Density	3·22 g/cm
Hardness	19 GPa
Flexural strength, RT 4-p	955 MPa
Weibull Modulus, RT	28
Flexural strength, 1350°C, 4-p	630 MPa
Fracture toughness	6 MPa$\sqrt{m}$
Thermal expansion coeff.	3·7 10^{-6} K^{-1} (20–800°C)
Thermal conductivity, RT	40 W/mK

on bend specimens. After static oxidation exposure, a cracked glassy oxide layer covers the specimens. An average weight gain up to 0.26 mg/cm^2 was observed after the 1000 h run.

Burner rig hot corrosion environment was performed at Kyushu National Industrial Research Institute, Japan. All burner rig exposed surfaces exhibited a powder like oxide surface layer easily removed by hand. An average weight loss up to -0.5 mg/cm^2 was recorded.. The residual 4-point bend strength, up to 1350°C is shown in Fig. 6 after the burner rig oxidation.

Table 2. Tensile creep rupture results of AC Cerama CSN 101

Bar 1: 1350°C, 50 MPa	> 1700 h (no failure)
Bar 2: 1350°C, 75 MPa	> 796 h (no failure, load increased)
Bar 2: 1350°C, 100 MPa	> 687 h (no failure)
Bar 3: 1300°C, 100 MPa	> 425 h (no failure, load increased)
Bar 3: 1300°C, 200 MPa	> 150 h (no failure)

3.2 HIPed long fiber combustor tiles

Gas turbine combustor concepts designed to give improved control of NO_x emissions require the use of hot uncooled walls. The main material properties needed in this application include mechanical and chemical stability at temperatures above 1400°C for over 10 000 h. Composites made from single crystal oxide fiber reinforced oxide with compatible high temperature stable–weak oxide interface are potential candidate materials to fulfil such requirements.

A consortium consisting of French, Swedish and British companies are addressing these needs in a Brite/EuRam supported programme.[16] Combustor tiles were made from single crystal oxide fiber reinforced oxide and tested up to 1000 h at 1400°C with good results.

Fabrication of the tiles by HIP started, as for the HP route, with filament winding, tape casting, cutting, stacking lamination (at 0 and 90° if so desired) and binder burn-out (see Ref. 11). A green composite, cut to shape, was placed in a recess with the same shape in a graphite block. A graphite block fitting the recess was slid in and sealed with BN slurry. The assembly was kept together, encapsulated in glass and HIPed at the desired temperature and pressure. Up to about 100 such assemblies could be processed in the same HIP cycle in AC Cerama's large press. The resulting tiles have properties and form fully comparable with HP parts. Particularly interesting with HIP is the potential for cost reductions and to make curved panels.

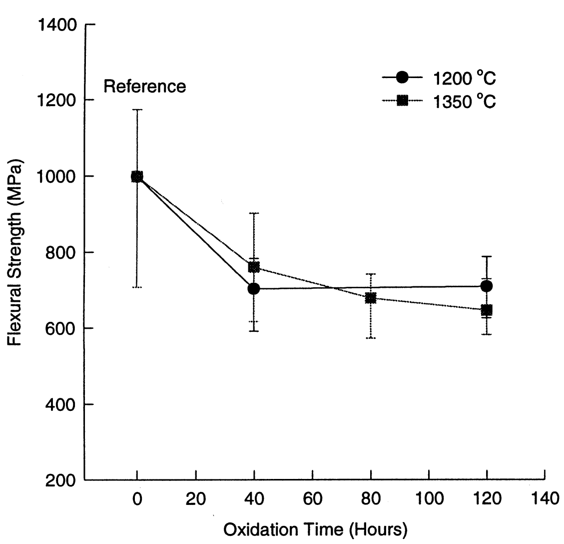


Fig. 6. Flexural strength after burner rig exposure (bars indicate the maximum and the minimum values).

4 Conclusions

HIP with glass encapsulation is suitable for fabricating several types of HT ceramics to near-net-shape. Integrated turbine wheels in the EUREKA-AGATA program have been processed to excellent HT properties and high precision in shape using injection molding and flexible glass powder encapsulation. Recent research throws light on how glass can interact at the surface of a green powder body and prevent even a very low viscosity glass from penetrating the powder body. Panels of highly non-isotropic long fiber CMCs can be processed to shape using rigid support tools and glass encapsulated HIP. Cost projections for the production of long fiber combustor tiles by HIP were favorable in comparison with other available processes, like conventional hot pressing. The feasibility to make curved panels is particularly interesting.

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