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Glasses for laser joining of zirconia ceramics

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Abstract

Laser joining of ZrO_2 ceramics using glasses and glass ceramics as sealing components requires optimized systems. The ternary systems SiO_2–BaO–B_2O_3 and BaO–SrO–SiO_2 were selected as a basis for development of suitable glass compositions for the laser joining process. Additives such as CaO, TiO_2, Al_2O_3, and MgO were used to control the crystallization processes and hence the thermal expansion coefficients during glass synthesis. The glass viscosity, the strength of the ceramic-glass-ceramic joint, and the joint tightness are other important glass properties which were optimized for the laser process. For glass G018-345, this yielded strengths of up to 225 MPa (Weibull modulus of $m = 8.6$) and He leak rates of up to 4.3×10^{-5} mbar l s^{-1}. Because of the varying viscosities obtained, the optimized glass systems could be used selectively in a temperature range of 700–900 °C.

Keywords: Laser joining; Glass sealant; Mechanical strength; Thermal expansion; ZrO_2 ceramics

1. Introduction

Current trends in materials development are showing an increasing demand for functionalized components and an associated need for joining materials of different compositions. The joining technology that leads to the best joint depends mainly on the material parameters and the compatibility of the parts to be joined.[1–3] In this paper, the joining of yttria-stabilized zirconia (YSZ) using laser radiation is described. A diode laser with combined beams of wavelengths 808 nm and 940 nm was used as the laser source. In comparison with conventionally used furnace technology, laser technology offers the decisive advantages of extremely short process times and pinpointed energy input. Development and optimization of laser-compatible and thermally stable glasses are examined in detail. The ZrO_2 material used in the experiments was selected due to its popularity as a design and functional material and its high development potential, as well as its selective oxygen ion permeability.[1,4] YSZ is used as a solid electrolyte in the production of SOFCs and as a functional layer in oxygen sensors.[1] The production of zirconia housings for sensor and display applications using laser joining has also been investigated.[5]

Extensive work was performed in the recent past to develop special sealing-glasses for joining zirconia ceramics. Melting was usually performed in a furnace, yielding borosilicate glasses with a B_2O_3:SiO_2 ratio of approx. 0.55. BaO was added to adjust the thermal expansion coefficients, La_2O_3 was added to alter the viscosity, crystallization was suppressed through addition of Al_2O_3.[6,7] For other SOFC glass applications using the system RO–Al_2O_3–SiO_2 (R = Ba, Ca), the BaO/CaO ratio varied between 6 and 8.[8] Other work dealt with glass systems in which specific crystallized components formed during joining. The relatively long process times of the furnace-based technology led to a near-equilibrium defined glass microstructure in the seam. Glass ceramics are preferential to conventional glasses for SOFC applications due to the higher joint strengths that can be achieved with them.[9,10] Thus, for example, addition of MgO to a commercially available glass from Schott AG significantly increased the strength of the joint due to the formation of crystalline phases such as $BaAl_2Si_2O_8$, SiO_2, and $MgSiO_3$.[9] The same effect was achieved through specific conditioning of selected BaO–Al_2O_3–SiO_2 glass compositions through addition of CaO and B_2O_3.[10] The main component that crystallized for this composition was $BaSiO_3$. Similar observations were made in investigations on the same base system modified with MgO and

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B_2O_3.[11] In this case, the crystalline components $Ba_2Si_3O_4$ and $Ba_5Si_8O_{21}$ as well as small amounts of crystalline $BaMgSiO_4$ and $Ba_2MgSi_2O_7$ were formed. The compositions described here yielded joints exhibiting high strengths in combination with ZrO_2 as well as chemical stability and high density in the specific application conditions typically found in high-temperature fuel cells.

In general, the ceramic microstructure adjacent to the joint seam is not changed during joining. Unlike welding, soldering requires introduction into the joint region of a glass that is compatible with both components to be joined.[3,12–14] Within the scope of the work presented here, the developed sealing-glasses were optimized to achieve the following:

- Good compatibility with the two components to be joined;
- Adequately high absorption of laser radiation by the sealing-glass;
- Suitable flow properties of the molten glass in the joint gap and very good wetting of the ceramic material;
- Specific thermal expansion coefficient of the glass adapted to that of the ceramic material;
- Joining temperature during the laser process at least 100 K above intended application temperature.

In laser-based joining, the energy required for melting the glass and heating the joint edge regions is introduced via the laser radiation in the joint area. Interaction between the laser and the ceramic material hence determines the quality of the joint. Nonoxide ceramics are practically nontransparent to the laser wavelengths of the diode laser. This means that the laser radiation is absorbed on the surface of the ceramic component and energy transport into the body of the ceramic occurs via heat conduction. The high thermal conductivity of nonoxide ceramics counteracts the thermally induced stresses, thereby preventing micro- and macrocrack formation.[12]

In contrast, oxide ceramics such as ZrO_2 are partially transparent for the specific wavelengths of diode lasers. This results in absorption of the laser radiation as well as the thermal energy into the volume of the ceramic body and is the reason why oxide ceramics, despite having low thermal conductivities, can be heated using laser radiation in short times without allowable thermal stress being exceeded. However, the partial transparency of oxide ceramics is strongly dependent on temperature. Measurements of optical properties as a function of temperature revealed a linear increase in laser absorption up to approx. 750 °C followed by an exponential increase in absorption at higher temperatures.[14] This fact must be taken into account during use of the laser technology.

2. Materials and methods

2.1. Materials

A commercial ZrO_2 ceramic material (ZN101B) from CeramTec GmbH was used for joining. The material was isostatically pressed and sintered at 1500 °C.

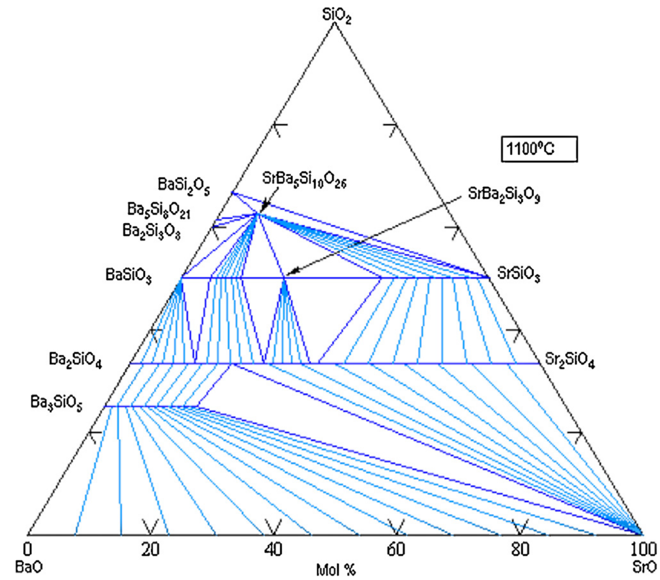

Fig. 1. Base system BaO–SrO–SiO_2 (Source: Sci. Glass).

Three glasses were developed by Schott AG for use as sealing-glasses in a laser joining process. The thermal expansion coefficients of the glasses were adapted as closely as possible to that of the zirconia ceramic material without increasing the joining temperature to >1100 °C. All of the glasses investigated showed crystalline phases in the glass matrix after joining, enabling the thermal expansion coefficients to be suitably adapted and adequate thermal stability of the joint to be achieved.

The first glass developed (G018-339) had a composition in the ternary system SiO_2–BaO–B_2O_3. Earlier work performed on the base system SiO_2–BaO–B_2O_3 showed crystallization of the phases sanbornite ($BaSi_2O_5$) and cristobalite (SiO_2). Cristobalite must be avoided due to its anisotropic expansion. The anisotropic thermal expansion of cristobalite is caused by a phase transformation in the temperature range between 200 °C and 300 °C. This phase transformation affects the thermal expansion behavior strongly and is undesirable due to a possible formation of cracks. Thus, CaO was added to the glass system to suppress the formation of cristobalite in the glass. TiO_2 was also introduced into the glass composition as a nucleation aid to favor crystallization of the desired phase, sanbornite.

The ternary system BaO–SrO–SiO_2 was selected as another base system. This system includes, among other features, a eutectic point at which the crystalline phase $SrBa_2Si_3O_9$ precipitates (Fig. 1). This phase has a relatively low melting point at approx. 1100 °C as well as high SrO and BaO contents, suggestive of a high thermal expansion coefficient. The SrO, BaO, and SiO_2 contents were selected to match the eutectic composition.

The glass G018-340 was synthesized through addition of Al_2O_3, MgO, CaO, and B_2O_3. These additive components were selected to reduce the glass formation temperature as well as the very strong crystallization tendency.

The third glass, G018-345, was yielded from application-specific adaptation of its predecessor. The composition was changed minimally (MgO concentration increased and Al_2O_3