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Experimental and numerical study of crack healing in a nuclear glass

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ABSTRACT

An experimental study of thermally or water-induced crack healing in an inactive borosilicate glass, chemically analogous to that used in France for the vitrification of nuclear waste was carried out. Partial welding of glass plates was observed after annealing in air at 425 °C (77 °C below T_g) when at least 20 MPa compressive stress was applied, while annealing at 450 °C under 20 MPa led to a complete disappearance of the interface. Closure of indentation-induced cracks was observed during annealing at 400 °C in an ESEM as a result of viscous relaxation of residual stresses but it did not constitute a sufficient proof of crack healing. DCDC specimens were thus pre-cracked in an ESEM and then either annealed at various temperatures (350–490 °C) in secondary vacuum or in air, or left in water at 70–90 °C, sometimes under a compressive stress normal to the crack face. The specimens were then reloaded in the ESEM and the crack opening displacements under a given load were compared to those measured during pre-cracking. The cracks were bridged by an alteration layer over a distance from the crack tip which decreased as loading increased. The restraining effect of these bridges on crack opening was assessed via finite element simulations, using interface elements. The tensile strength of the bridging layer was estimated as 27–39 MPa after vacuum annealing at 400 °C, 11–20 MPa after 15 days in water at 90 °C and 44–78 MPa after 11 days in water at 70 °C under 5 MPa normal compression. Partially healed cracks did not resume propagation from their former crack tip, but due to branch cracks re-initiated a few hundred microns behind it which grow avoiding the healed area. This behaviour was explained using finite element simulations.

1. Introduction

1.1. Context of the study

In a French reprocessing plant, large blocks of vitrified nuclear waste for underground disposal are prepared by pouring a mixture of crushed high-activity waste and molten borosilicate glass into steel canisters. Upon cooling, cracks may develop due to thermal gradients. This problem has been investigated in previous studies (Dubé et al., 2010; Doquet et al., 2013). However, since the radionuclides constitute an internal heat source and since several canisters are initially stored in the same room, the temperature inside the glass blocks remains high during a long period (above 400 °C during the first day and above 350 °C during three years, according to Barth et al., 2012), so that thermally-induced healing of the cracks can be expected, at least partially. An experimental and numerical study of thermally-induced crack healing in an inactive borosilicate glass, chemically analogous to the industrial product was therefore undertaken.

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Another issue to address is that of long-range behaviour: even though the glass block temperature will eventually drop below 90 °C (due to radioactive decay), water will at a time be able to reach the glass blocks. In addition, the galleries may collapse, so that the glass block will also be submitted to a lithostatic compression.

All this might trigger subcritical growth of some of the pre-existing cracks, more precisely those submitted to a sufficient mode I loading. Many studies in the literature have already been devoted to this topic. At the same time, the new solicitations might also heal those among the cracks that will be submitted to a normal compression or even to a weak mode I loading below the subcritical crack growth threshold. Contrary to the former "damaging" scenario, the latter has hardly been considered so far. That is why an investigation of water-induced and mechanically-assisted crack healing at 70 to 90 °C was also undertaken.

Before describing this study and its results, a survey of the literature devoted to crack healing phenomena in glasses, at high or low temperatures will first be presented.

1.2. Literature on thermally-induced crack healing in glasses around T_g

Thermally-induced healing of indentation-induced cracks in transparent soda-lime glass annealed at 650 °C has been observed by Hrma et al. (1988). The process was attributed to "capillarity-driven viscous flow" and the adimensional variable $\frac{t\mu}{\eta}$ – where t stands for the annealing time, μ for the shear modulus and η for viscosity – was considered as a controlling parameter.

Hirao and Tomozawa (1987) annealed various types of glass near their glass-transition temperature, in air or vacuum, and found that annealing in moist air is much more efficient for strength recovery of indentation-precracked specimens. They attributed this recovery to viscous flow-induced crack tip blunting, made easier by moisture since, as shown by Bartolomew (1983), water absorption reduces the viscosity of glass. Crack tip blunting suppresses the stress singularity at the crack tip and thus explains the strength recovery.

This phenomenon was later confirmed by Atomic Force Microscope (AFM) measurements by Kese et al. (2006).

Girard et al. (2011) reached similar conclusions as those of Hirao and Tomozawa concerning the lowering of viscosity by water and the enhancement of crack healing.

Holden and Fréchette (1989) also concluded that a certain degree of moisture is necessary for crack healing in soda-lime-silica glass annealed at 550 °C and that a compressive stress assists self-welding of contacting glass plates during annealing, provided that it is not so high that it prevents moisture from reaching the interface, if the stress is applied before the temperature rise.

This argument seems questionable, since, according to the kinetic data from D'Souza and Pantano (2002), water was probably already adsorbed on the surface of their specimens (kept in air before annealing). Due to surface roughness, which leaves empty space for water in-between the contacting surfaces, a compressive stress might even be suspected to trap water instead of preventing its access.

Wilson and Case (1997) investigated thermally-induced healing of indentation-induced cracks in borosilicate glass by direct ESEM observations during short *in situ* annealing. They reported crack tip regression or crack pinch-off at discrete points along the crack. They varied the initial moisture content in the ESEM chamber during annealing (Wilson and Case, 1999) and concluded that a higher moisture allows crack healing at a lower temperature (as low as 370 °C in borosilicate glass for a moisture level RH = 64%).

Similar *in situ* observations were also performed by Méar et al. (2011) who found that healing starts when the temperature approaches the softening temperature, T_s, for which the viscosity of the glass drops to 10^{11} Poise. In the glasses used to seal solid oxide fuel cells that they investigated, T_s was 60 to 65 °C above T_g (for which the viscosity is 10^{13} Poise).

A linear relationship between the time necessary for complete crack healing and glass viscosity was deduced from optical monitoring of indentation-induced cracks during high temperature annealing of silicate glass by Sing and Parihar (2009). They developed a predictive model based on a three-stage process: (1) crack tip blunting and some other morphological changes, which results in cylinderisation of the crack, (2) filling of the cylindrical cavity, which results in spheroidisation and (3) filling of the spherical cavity, which results in a crack-free surface. They also considered the softening temperature of the glass as a threshold temperature for thermally-induced crack healing.

1.3. Literature on low-temperature crack healing in glasses

Cheeseman and Lawn (1970) achieved partial healing of Hertzian cone cracks in an unspecified type of glass, by low temperature annealing (200 °C) in nitrogen and even at room temperature, under the mere effect of a compressive stress.

Widerhorn and Townsend (1970) reported a more complete room temperature healing of cracks induced in soda-lime-silica glass by mechanical shock in a superdry nitrogen atmosphere than that of cracks grown in air. They attributed this difference to a reduction in chemical activity of fracture surfaces by adsorbed H_2O and O_2.

Michalske and Fuller (1985) measured the energy release rate, G_r, corresponding to re-opening of "healed" cracks (as they call cracks kept closed for 5 min at room temperature in various environments with controlled moisture) in vitreous silica and soda-lime silica glass. They suggested that hydrogen bonding of water molecules adsorbed on crack surfaces is sufficient to pull crack surfaces together and cause crack closure in silica or soda lime glass while a finite load is still being applied. Due to its relatively long range, hydrogen bonding would be the first interaction to take place, while the cracks are progressively closing. For soda-lime glass, they report the highest G_r in the driest conditions, which they attribute either to bridging between SiO^- groups by surface-adsorbed cations, or to the formation of bridging siloxane bonds by the condensation of surface silanol groups.