Reducing Hazard in Spent Fuel Removal Using Colloidal Silica Gel – 22274

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ABSTRACT

Nuclear site decommissioning involves the retrieval and handling of radioactive waste. Waste removal from nuclear reactors and/or storage facilities, such as spent fuel pools and storage silos, represents a potential hazard in terms of radiation exposure for the workforce and the surrounding environment. This study explores the suitability of colloidal silica grouting around radioactive waste to reduce radiation exposure during retrieval operations. Previous work on colloidal silica gel has proved its potential to form low-permeability hydraulic barriers, and to inhibit the diffusion of radionuclides through the gel, making it a promising material for spent fuel recovery applications. This work provides experimental evidence that colloidal silica hydrogel can maintain its integrity upon exposure to temperatures typical of the nuclear waste stored within pools and silos, both in standard conditions (up to 60 °C), and during loss of cooling/loss of coolant accidents (>100 °C).

INTRODUCTION

Nuclear power plants have a designed lifespan of 30 to 60 years. At the end of their operating life, nuclear sites have to be decommissioned following internationally recognized approaches [1], aimed at the clean-up of radioactivity and the progressive dismantling of the plant.

Decommissioning operations involve the retrieval and handling of radioactive waste. Waste removal from nuclear reactors and/or storage facilities, such as spent fuel pools and storage silos, represents a potential hazard in terms of radiation exposure for the workforce and the surrounding environment. This may be due to the accidental release of airborne radioactive particulates during waste recovery and transport, or to the loss of radioactive debris upon retrieval due to waste corrosion and degradation. The development of innovative techniques to reduce hazard in spent fuel removal operations is a critical aspect of site decommissioning.

This study explores the suitability of colloidal silica grouting around radioactive waste prior to removal, to reduce radiation exposure during nuclear waste retrieval operations. Colloidal silica is an aqueous suspension of silica (SiO_2) nanoparticles, with average particle size <100 nm. The creation of siloxane bonds (Si – O – Si), typically triggered by the addition of an electrolyte accelerator, leads to the formation of a solid-like network of silica nanoparticles in the form of a hydrogel. Previous work on colloidal silica gel has proved its potential to form low-permeability hydraulic barriers against fluid migration ([2];[3];[4]), and to inhibit the diffusion of radionuclides through the gel [5], making it a promising material for spent fuel recovery applications. However, such applications require evidence that colloidal silica hydrogel can maintain its integrity upon exposure to temperatures higher than ambient, typical of the nuclear waste stored within pools and silos. These might be in the range 40-60 °C in standard conditions, but could potentially exceed 100 °C during loss of cooling/loss of coolant accidents.

In this study, an experimental investigation was carried out to simulate colloidal silica grouting operations around objects at temperatures higher than ambient, up to a maximum of 120 °C. The effect of the temperature of the grouted object, and of the silica grout properties, on the performance of the gel was explored by carrying out a) microstructural analyses by x-ray imaging, to detect the presence and spatial distribution of temperature-induced cracks within the gel, and b) mechanical tensile and shear strength tests on gel samples at different silica concentrations.

The results of the experimental campaign carried out in this study confirmed the potential of colloidal silica grouting for spent fuel removal applications. In addition, critical parameters for design of the silica grout mix, to optimize performance upon exposure to temperature, are identified and discussed.

MATERIALS AND METHODS

Colloidal silica

Colloidal silica grout was prepared in this study by mixing a colloidal silica suspension (MasterRoc® MP320 Part A, supplied by BASF) with a NaCl electrolyte accelerator, at a 5:1 colloidal silica-to-accelerator ratio by volume. To assess the effect of different grout properties on the hydrogel's performance upon exposure to temperature higher than ambient, grout mixes at two silica concentrations were prepared and tested in this study, namely 34%wt and 17%wt. The molarity of the NaCl solutions used as the electrolyte accelerators were designed to obtain a gel time at room temperature of 10 minutes for both grout mixes.

Grouting experiments

Grouting experiments were performed in this study to simulate grouting operations around objects at temperature higher than ambient. The experimental setup is schematically shown in Figure 1. Exposure to high temperature was provided by means of a stainless-steel cylinder with an embedded thermocouple and resistive cartridge heater, both connected to a Proportional Integral Derivative (PID) temperature controller. Grouting operations were carried out by pouring colloidal silica grout (turning into a hydrogel 10 minutes after grout mixing) on the heating element, within an acrylic cylindrical container.

Three grouting experiments were performed to assess a) the effect of the temperature of the heated element, and b) the effect of the concentration of silica in the hydrogel, as shown in TABLE I. In experiment 1, a hydrogel at 34%wt silica concentration was exposed to 60 °C for 7 hours. In experiments 2 and 3, hydrogels at silica concentration of 34%wt and 17%wt respectively were exposed to 120 °C for 7 hours. Evaporation cracks were prevented by covering the gel's surface with a thin layer of water. Temperature-induced cracking was visually assessed during the experiments, and 3D-imaged via X-ray Computed Tomography (X-CT) at the end of the experiments.

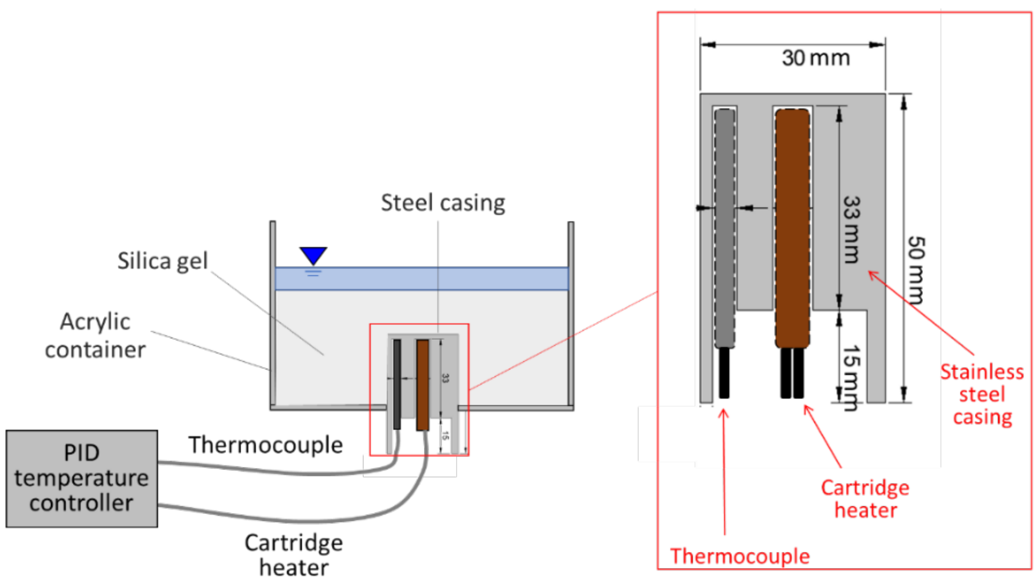

Fig. 1. Schematic view of the experimental setup for grouting experiments

TABLE I. Experimental conditions of grouting experiments

	Temperature of the headed element (°C)	Silica concentration (%wt)
Experiment 1	60	34
Experiment 2	120	34
Experiment 3	120	17

Mechanical tests

Mechanical testing upon different loading conditions were performed in this study on cylindrical samples (37 mm diameter, 74 mm height) to assess the effect of silica concentration (34%wt and 17%wt) on the gel's strength, and to help interpret the cracking behavior observed during grouting experiments. Cylindrical samples for mechanical testing were created by injecting colloidal silica grout within bespoke cylindrical molds, filled with 2-mm diameter glass-beads. After grout gelation had occurred, the grouted glass beads samples were extruded and cured at room temperature for 11 days prior to mechanical testing. The presence of glass beads in the gel's matrix facilitated the handling of the samples upon mechanical testing by providing additional resistance upon loading. Although the results of the mechanical tests refer to the overall glass beads/silica hydrogel system, they are qualitatively representative of the mechanical behavior of the hydrogel, as failure only occurred within the gel matrix in all tests (inter-granular failure).

The ability of the gel to accommodate deformation upon tensile loading, mimicking crack initiation upon water vapor formation, was assessed by performing splitting tensile tests in a loading frame apparatus at constant strain rate (0.1 mm/min). On the other hand, the stress-strain behavior of the gel upon shearing was assessed by performing Unconsolidated-Undrained (UU) triaxial tests within a triaxial cell apparatus, to explore the effect of silica concentration on the ability of the gel to be handled during retrieval and disposal operations.

RESULTS AND DISCUSSION

The results of the X-ray tomographies from the three grouting experiments are reported in Figure 2. Nikon software CT Pro 3D was used for the 3D image reconstruction, and Avizo 9.3 software was used for processing the reconstructed volumes. Here, the mid-sections of the reconstructed three-dimensional gel volumes after 7-hour exposure to temperature higher than ambient are shown. Three different phases were detected upon image processing, namely water, air, and silica gel.

At 60°C and silica concentration 34%wt (Figure 2a, Experiment 1), the hydrogel performed extremely well, with no significant cracking observed after 7-hours exposure time. After extrusion from the acrylic cylinder (Figure 3a), the gel remained intact and easy to handle, thus confirming the potential of colloidal silica for grouting around waste at temperatures lower than 100 °C.

At 120°C, gel cracking was observed (Figure 2b and 2c, Experiments 2 and 3). Extensive cracking was detected within the gel at higher silica concentration (SiO_2 34%wt, Figure 2b). Here, X-ray tomographies revealed the presence of large cracks propagating from the heated element outwards, resulting from the phase change of liquid pore water surrounding the heated element (and, hence, the formation and release of water vapor). Upon extrusion from the acrylic container, the hydrogel failed along the largest crack located directly above the heated element, as shown in Figure 3b.

On the other hand, lowering the silica concentration from 34%wt to 17%wt (Figure 2c) appeared to significantly improve the performance of the hydrogel against temperature-induced cracking compared to Experiment 2. Despite the presence of isolated voids and mm-sized cracks, the X-ray tomographies did not detect a network of connected cracks within the hydrogel. Similar to Experiment 1, the gel remained intact upon extrusion (Figure 3c), but exhibited a softer consistency compared to both hydrogels created in Experiments 1 and 2.

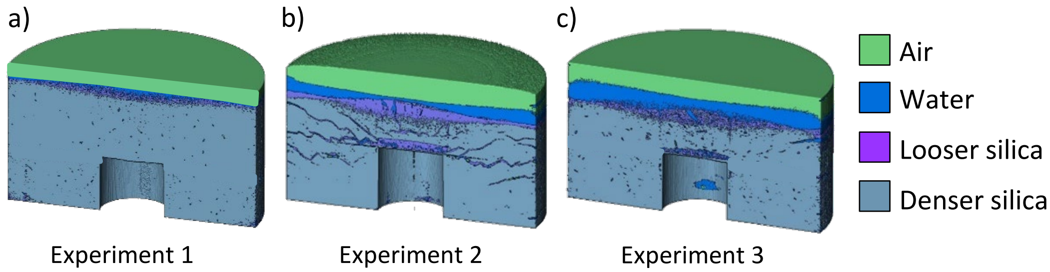

Fig. 2. X-ray tomographies of the mid-sections of silica gel grouted around a cylindrical object at T = 60°C (a), and T = 120°C (b and c). At 60°C (a), no cracks are observed upon 7-hour exposure to temperature. At 120°C (b and c), cracks are formed. Lowering the silica concentration in the grout from 34%wt (b) to 17%wt (c) significantly improves the performance of the hydrogel against temperature-induced cracking.

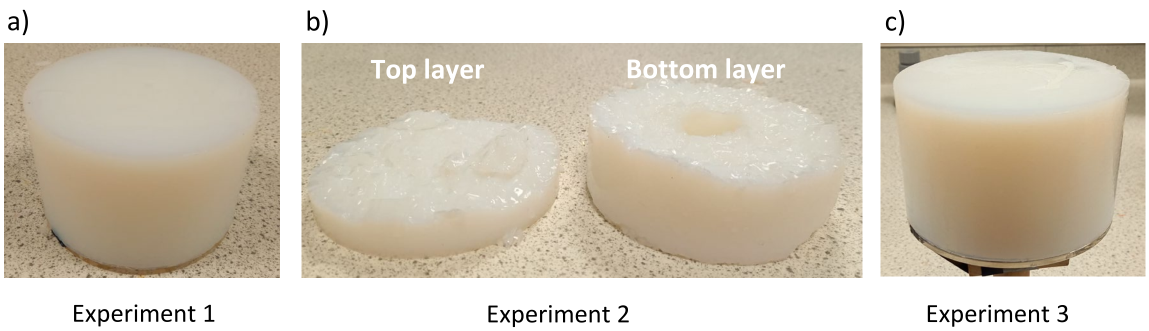

Fig. 3. Gel volumes after extrusion from the acrylic container, at the end of Experiment 1 (a), Experiment 2 (b) and Experiment 3 (c).

The cracking behavior observed at different silica concentrations can be explained by considering the results of the mechanical tensile and shear tests performed in this study (Figure 4 and 5 respectively). Upon tensile loading, which is representative of the loading condition triggering crack initiation upon water vapor release, the hydrogel at higher silica concentration (34%wt) exhibited the highest tensile strength, as shown in Figure 4 (in blue). This is due to the larger quantity of silica nanoparticles in the grout compared to the case of lower silica concentration, thus forming a stronger gel structure (i.e. more silica nanoparticles leads to more siloxane bonds formed). However, the higher tensile strength is accompanied by a stiffer stress-strain response, i.e., by a limited ability to accommodate deformation as the tensile stress increases. The stiffer stress-strain response of the hydrogel at 34%wt silica concentration is likely to be responsible for the severe cracking observed in grouting Experiment 2. On the other hand, the lower silica concentration of 17%wt allowed the hydrogel to accommodate larger vertical displacements upon tensile loading (Figure 4, red line), thus justifying the absence of extensive cracks in grouting Experiment 3.

Similar trends were observed upon UU triaxial testing (Figure 5). At 34%wt silica concentration, the hydrogel exhibited a higher shear strength and a brittle-like failure, with the formation of a clear shear band. At 17%wt silica concentration, the hydrogel exhibited a lower shear strength and a ductile stress-strain behavior, followed by barrelling. While a higher gel deformability is beneficial against crack initiation and propagation, it is also responsible for a softer consistency, and a reduction in shear strength. This may pose a limitation on the ability to handle the hydrogel (and, hence, any object grouted with colloidal silica). Therefore, the silica concentration of the grout needs to be carefully selected to allow appropriate deformability as to prevent/limit temperature-induced cracking, while ensuring gel integrity upon handling.

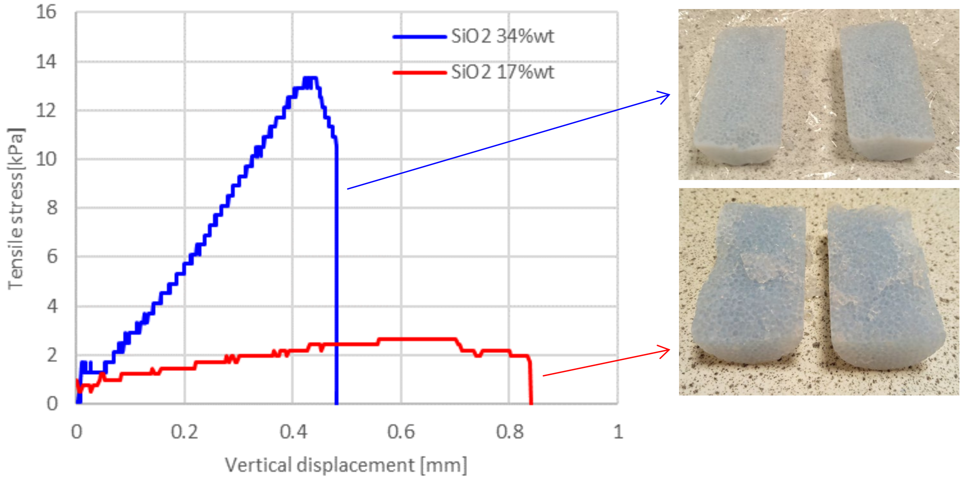

Fig. 4. Results of splitting tensile tests on colloidal silica grouted glass beads at different silica concentrations (34%wt and 17%wt), cured for 11 days at 20°C. Gels at higher silica concentration (34%wt) exhibited a higher tensile strength and a stiffer stress-strain response, as opposed to gels at lower silica concentration (17%wt). The ductile behavior of gels at lower silica concentration is responsible for the enhanced ability to accommodate deformation upon crack initiation.

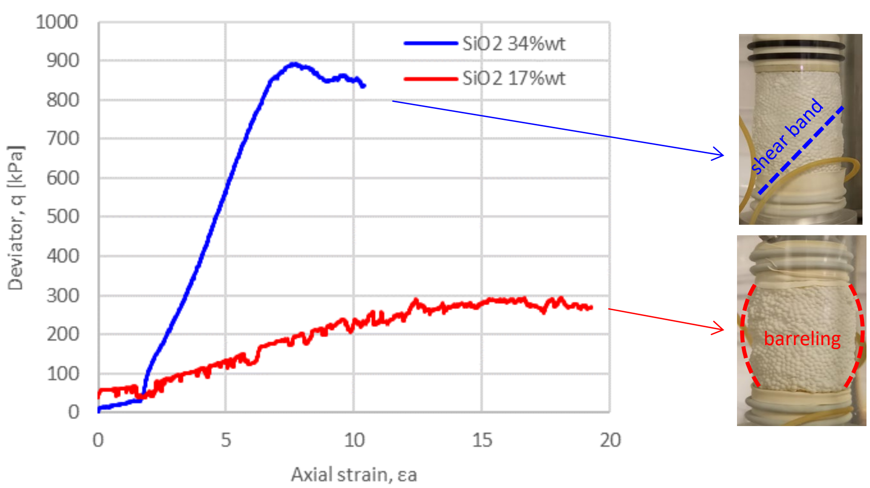

Fig. 5. Results of triaxial tests on colloidal silica grouted glass beads at different silica concentrations (34%wt and 17%wt), cured for 11 days at 20°C. Gels at higher silica concentration (34%wt) exhibited a higher strength and a brittle-like behavior, as opposed to gels at lower silica concentration (17%wt). The ductile behavior at lower silica concentration is responsible for the softer consistency of the gel.

CONCLUSIONS

The experimental campaign carried out in this study confirmed the potential of colloidal silica grouting for spent fuel removal applications. The results of the grouting experiments and mechanical tests showed an excellent performance of colloidal silica hydrogel around objects at 60°C, with no temperature-induced cracking observed over 7 hours exposure time. At higher temperature (120°C), the phase change of the hydrogel's pore water was showed to trigger the formation of cracks. The combined analysis of grouting experiments and mechanical testing upon different loading conditions showed that a) lowering the silica concentration in the grout has a beneficial effect on the ability of the hydrogel to undergo deformation, thus inhibiting/reducing crack initiation and propagation; b) at the same time, lowering the silica concentration can be detrimental with respect to the gel's strength and, hence, its ability to be retrieved and handled. Therefore, the properties of the grout need to be carefully designed and optimized prior to starting a grouting campaign around radioactive waste, in order to minimize temperature-induced cracking while ensuring the integrity of the grouted waste upon removal.

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ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support of the UK Research and Innovation-EPSRC under grant EP/S01019X/1, 'TRANSCEND: Transformative Science and Engineering for Nuclear Decommissioning'.