# Grouting of well leakage and migration pathways using colloidal silica: a preliminary experimental investigation

Arianna Gea Pagano, Grainne El Mountassir, Rebecca Jane Lunn

Department of Civil and Environmental Engineering, University of Strathclyde, Glasgow G1 1XJ, UK

## Abstract

Hydrocarbon well decommissioning and abandonment require the long-term sealing of potential fluid migration pathways. Current grouting technologies based on the use of cementitious grouts may not be able to achieve proper penetration and sealing of migration pathways, this resulting in an environmental risk due to the migration of hydrocarbons or other well fluids out of the well and into the environment.

Colloidal silica (CS) based grouts differ from traditional cement grouts due to their low viscosity (similar to water), and their excellent penetrability governed by the size of the silica nanoparticles (~15nm). As a result, CS grouts have the potential to seal migration pathways (i) at cement/casing or casing/cement interfaces where the cement bond is compromised, and (ii) due to micro-channels and micro-annuli within the cement annulus. This study presents a preliminary laboratory investigation to demonstrate the suitability of CS grout for the purpose of creating multiple barriers within hydrocarbon wells. To this end, CS grout was injected into fractured cement cores using a high pressure/high temperature core-holder apparatus to simulate target downhole scenarios. Microstructural analysis (micro-CT scanning) prior and post treatment were conducted to image the spatial distribution of the grout within the fracture network. The effectiveness of the treatment was measured by evaluating i) the core permeability before and after treatment with CS, and ii) the unconfined compressive strength of treated cement cores.

## Introduction

Hydrocarbon well decommissioning and abandonment requires the long-term sealing of potential fluid migration pathways. Current grouting technologies based on the use of cementitious grouts are not able to penetrate fine fissures and cracks and therefore may not seal all potential migration pathways, resulting in the possible long-term migration of hydrocarbons or other well fluids out of the well and into the environment.

Colloidal silica (CS) based grouts are an alternative to traditional cementitious grouts [1] - [4]. CS is a non-toxic, low viscosity aqueous suspension of silica particles which forms a gel upon destabilisation, typically triggered by the addition of an accelerator (electrolyte solution). The gelation process is controlled by several factors, including particle size, particle concentration, pH, electrolyte concentration and valency, and temperature [5]. Analytical models able to predict gelling depending on the grout composition are available in the literature [6].

CS grouts differ from traditional cement grouts due to their low viscosity (similar to water), and their excellent penetrability governed by the size of the silica nanoparticles (~15nm). As a result, CS grouts have the potential to seal migration pathways (i) at cement/casing or casing/cement interfaces where the cement bond is compromised, and (ii) due to micro-channels and micro-annuli within the cement annulus. Moreover, the well-established pozzolanic activity of colloidal silica particles may offer further benefit to treated cement by promoting the generation of additional cementitious compounds, making use of available calcium sources naturally occurring in hydrated cement pastes [7] – [9].

This study presents a preliminary laboratory investigation to demonstrate the suitability of CS grout for sealing cracks within the cement annulus. To this end, CS grout was injected into a fractured cement core with constant fracture aperture and cured at reservoir conditions.

Experiments were carried out within a high pressure/high temperature core-holder apparatus to simulate target downhole scenarios. The effectiveness of the treatment was measured by evaluating the core hydraulic conductivity before and after treatment with CS.

# Materials and methods

Cylindrical cement cores (75mm height x 38mm diameter) with constant fracture aperture of 0.2 mm were prepared using Ordinary Portland Cement. Cement paste was prepared by mixing cement powder with de-ionised water by means of a rotary mixer at a water-to-cement ratio w/c = 0.375. The two halves of each cement core were cast in silicon rubber moulds, specially designed to obtain a constant fracture aperture of 0.2 mm. The cement mix was left to cure in the moulds for 24 hours under controlled humidity and temperature. Then, the hardened cement pastes were de-moulded and placed in deionised water to cure for an additional 27 days. The specimen preparation procedure carried out in this study is showed in Figure 1.

A commercially available colloidal silica (CS) suspension, Meyco MP320, was used in this work. Meyco MP320 is a solvent-free, low viscosity suspension of nanometric colloidal silica particles (viscosity ~10 mPa.s, particle size ~15 nm). CS grout was prepared by mixing the as-delivered CS suspension with de-ionised water (5:1 CS-to-water ratio by volume). No accelerator (i.e. electrolyte solution) was added to the grout.

Hydraulic conductivity measurements and flow-through experiments at relevant downhole conditions were performed in a bespoke high pressure/high temperature core holder (Figure 2). A pre-treatment hydraulic conductivity measurement was carried out on the fully cured core by injecting tap water at varying flow rates. The resulting inlet pressure was continuously monitored in order to derive hydraulic conductivity using Darcy's law:

$$k = \gamma_w \cdot \frac{Q}{A} \cdot \frac{L}{\Delta P_{IN-OUT}}$$

where k is the hydraulic conductivity [m/s],  $\gamma_w$  is the unit weight of water [kN/m<sup>3</sup>], Q is the flow rate [m<sup>3</sup>/s], A and L are the whole cross sectional area [m<sup>2</sup>] and length [m] of the core respectively, and  $\Delta P_{IN-OUT}$  is the pressure differential across the core [Pa].





Proceedings of the RM4L2020 International Conference



Figure 2. Schematic view of the bespoke high pressure/high temperature core holder.

Hydraulic conductivity was derived by considering the average value of Q and  $\Delta P_{IN-OUT}$  once stable values had been reached during the test. Once the initial hydraulic conductivity had been determined, the cement core was then treated by injecting CS grout. The treated core was cured in the core holder at a selected high temperature and high pressure combination (P = 15 MPa, T = 80 °C, simulating 1.6 km depth) for 10 hours, then left to equalise to ambient conditions for a further 62 hours (total curing time: 72 hours). Hydraulic conductivity measurements were carried out after treatment by injecting tap water through the core at increasing inlet pressure, from 0 to 1.4 MPa. Inlet pressure was ramped up in steps. During each step, the resulting flow rate through the core was continuously monitored to derive hydraulic conductivity. The selected values of confining pressure, back pressure, injection pressure and flow rate adopted for each stage of the experiment are shown in Table 1.

			-
	Before treatment	Curing	After treatment
Confining pressure [MPa]	3	15	3
Flow rate [mL/min]	4 to 10	-	measured
Inlet pressure [MPa]	measured	15	0 to 1.4
Outlet pressure [MPa]	0 (atmospheric)	15	0 (atmospheric)
Temperature [°C]	20	80	20

Table 1. Core-holder experimental parameters during flow-through core tests and curing

# **Results and discussion**

The results of the hydraulic conductivity measurements obtained before and after treatment are shown in Figure 3a and 3b respectively. Before treatment, the fractured core exhibited a hydraulic conductivity of ~10<sup>-6</sup> m/s. After treatment, a hydraulic conductivity reduction of 4 orders of magnitude (down to ~10<sup>-10</sup> m/s) was observed. The treated cement core was able to withstand injection pressures up to 1.4 MPa for 12 hours, with no sign of water breakthrough. Furthermore, the CS grout treatment successfully bonded together the two halves of the core. The observed decrease in hydraulic conductivity and fracture sealing may be ascribed to two separate processes occurring within the fracture when colloidal silica comes into contact with cement: i) gel formation, and ii) calcium silicate hydrate (C-S-H) production. Gel formation is the consequence of the destabilisation of the colloidal suspension, usually induced by the addition of an accelerator (electrolyte solution). Since no accelerator was added during the preparation of the grout, the formation of siloxane bonds between colloids and consequent gelling is to be attributed only to the presence of cations (predominantly calcium ions) released by the cement into the fluid present in the fracture. In addition, the exposure to high temperature (80°C) to simulate reservoir conditions is expected to increase the collision rate of particles in suspension, thus facilitating the gelation process.

On the other hand, silica nanoparticles, interacting with calcium sources present on the fracture surfaces (e.g. portlandite, Ca(OH)<sub>2</sub>), may undergo pozzolanic reactions to form additional cementitious compounds, such as calcium silicate hydrate (C-S-H). The formation of C-S-H may reduce or even bridge the gap between the two fracture surfaces, resulting in a hydraulic conductivity reduction and/or bonding of the two halves of the core.

The formation of siloxane bonds (gelling) and C-S-H products are competing phenomena: the higher the gelation rate, the lower the amount of available reactive sites for C-S-H formation and vice versa. Further investigations quantifying the amount of silica gel versus C-S-H produced upon treatment and their dependence on silica concentration and pore water composition are ongoing.



Figure 3. Flow-through experiment results: a) before treatment (k =  $\sim 10^{-6}$  m/s), and b) after treatment (K =  $\sim 10^{-10}$  m/s).

#### Conclusion

This preliminary experimental investigation shows the promising potential of CS grout treatment for the long-term sealing of fluid migration pathways in oil and gas wells for decommissioning and abandonment. The injection of CS grout cured at reservoir conditions was observed to successfully reduce hydraulic conductivity by 4 orders of magnitude, even in the absence of an accelerator. Colloidal silica makes use of calcium sources naturally occurring within the pore space and fracture network of cementitious materials to i) induce grout gelling, and ii) encourage the formation of additional cementitious compounds, thus activating a self-healing process within degraded cements. Further understanding of the mechanisms of gel formation and C-S-H production will be investigated via microstructural and mineralogical analyses, taking into account the composition of the water within the pore space and for a range of fracture geometries.

# Acknowledgment

The authors gratefully acknowledge the financial support of the OGTC (Aberdeen, Scotland) under project WC-P-018, "Grouting of well leakage and migration pathways: Biogrouting and colloidal silica".

# References

[1] Moridis G, Persoff P, Apps J, Myer L, Pruess K, Yen P. A field test of permeation grouting in heterogeneous soils using a new generation of barrier liquids. Presented at the ER '95 Conference Committed To Results: Barriers for Long-Term Isolation 1995.

[2] Moridis GJ, Finsterle S, Heiser J. Evaluation of alternative designs for an injectable barrier at the Brookhaven National Laboratory Site, Long Island, New York. 1999 Water Resources Research Vol 35, pp. 2937–2953.

[3] Hakem N, Al Mahami, I, Apps J, Moridis G. Sorption of cesium and strontium on Savannah River soils impregnated with colloidal silica. 1997 International Containment Technology Conference, Petersburg pp. 652–657.

[4] Manchester K, Zaluski M, North-Abbott M, Trudnowski J, Bickford J, Wraith J, Grout selection and characterization in support of the colloidal silica barrier deployment at Brookhaven National Laboratory. 2001 Proceedings International Containment and Remediation Technology Conference and Exhibition, Orlando, FL.

[5] Iler RK. The Chemistry of Silica: Solubility, Polymerization, Colloid and Surface Properties, and Biochemistry. (John Wiley & Sons 1979).

[6] Pedrotti M, Wong C, El Mountassir G, Lunn RJ. An analytical model for the control of silica grout penetration in natural groundwater systems. Tunnelling and Underground Space Technology. 2017 Vol 70 pp. 105-113.

[7] Li H, Xiao HG, Yuan J, Ou J, Microstructure of cement mortar with nanoparticles. Composites Part B: Engineering. 2004 Vol 35 pp. 185–189,

[8] Cardenas H, Struble LJ. Electrokinetic nanoparticle treatment of hardened cement paste for reduction of permeability. Journal of Materials in Civil Engineering. 2006 Vol 18 pp. 554-560.

[9] Maddalena R, Hamilton A. Low-pressure silica injection for porosity reduction in cementitious materials. Construction and Building Materials. 2017 Vol 134 pp. 610-616.