



Cement/rock interaction in geothermal wells. The effect of silica addition to the cement and the impact of CO₂ enriched brine

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ABSTRACT

The limited durability of the cement in acidic fluids is one of the main issues controlling the lifetime of a geothermal well. The most critical region is the outermost layer of the cement adjacent to the formation.

This paper provides insight into the way in which well cementing formulations interact with a common rock formation in geothermal environments, at 150 °C in brine, with and without CO₂ exposure.

The results show that chemical reactions occur in the ITZ, which are driven by Ca²⁺ and OH⁻ migration from the cement into the rock, which are dependent on the type and amount of silica added to the cement.

The presence of CO₂ influences the extent of cement/rock interaction which is dependent on the cement formulation. The current work indicates that the carbonation in these systems is a very complex set of reactions, where poorly crystalline and amorphous calcium carbonates may form.

1. Introduction

One of the main issues associated with ongoing exploitation of the geothermal resources in New Zealand has been concerns of the durability of the cement sheath formed during downhole cementing and the impact it has on the lifetime of wells, many of which do not last as long as desirable. Research is being conducted to improve the understanding of how the current cement formulations perform in the high temperature, high CO₂ concentration and low pH fluids found within New Zealand's geothermal systems where CO₂ induced corrosion has been shown (Silva and Milestone, 2016).

The cements used in geothermal well cements are essentially those devised for oil and gas wells and are Portland based cements prepared to American Petroleum Institute (API) specifications. At elevated temperatures and pressures, different hydration reactions occur in Portland cement systems, compared to those found in civil construction (Silva and Milestone, 2016). With neat cement, continuous exposure to hydrothermal conditions induces a gradual loss in compressive strength in hardened Portland cement. This phenomenon known as 'strength retrogression', was investigated by Kalousec and Adams (1951) and Kalousec (1954, 1955) who showed it to be caused by the formation and slow growth of new, crystalline high Ca/Si ratio calcium silicate hydrate phases, which are associated with increased porosity and permeability as the crystals enlarge and densify in the alkaline medium,

leaving a rather porous matrix that has little strength. At hydrothermal temperatures above 120 °C, crystalline α -di-calcium silicate hydrate (α -C₂SH) forms along with Ca(OH)₂, causing a high permeability and low compressive strengths (Milestone et al., 2012a) with phases such as jaffeite and reinhardbraunsite being formed at higher temperatures.

Pure quartz flour is normally used as a strength retrogression inhibitor. Kalousec (1955) found that by adding > 30 wt% silica flour (very fine quartz) by weight of blend (BWOB), the overall Ca/Si ratio in the binder was decreased and strength retrogression could be avoided through the formation of a new, crystalline, low Ca/Si ratio phase, tobermorite (C₅S₆H₅), which has low permeability and good compressive strength. Below 100 °C quartz will not react readily (Milestone et al., 2012a; Bensted, 2018; Ramachandran and Beaudoin, 1999). Recent work (Iverson et al., 2010) suggests that even > 30 wt% silica flour may be insufficient to completely eliminate strength retrogression. The reactions with silica depend critically on the form of silica present. For instance, the use of a coarse silica sand gives rise to the formation of poor cementitious products, whereas the use of silica flour enhances the formation of tobermorite which is a favourable mineral for cement performance (Bensted, 2018). Amorphous forms of silica do not crystallise to tobermorite and so have been neglected (Luke, 2004; Grabowski and Gillott, 1989a,b).

Most of the thermal waters tapped for power generation lie in pervious, acid silicate rocks such as rhyolite, andesite or ignimbrite.

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These rocks are known to cause problems due to alkali silicate reactions (ASR) if used in conventional concrete. Although there are considerable variations in conditions, even within the same geothermal field, several factors emerge which can influence any cement/rock interactions. The alkaline cement will react with acidic rocks in a similar way to ASR in conventional concrete, with the high temperature (up to 320 °C), hydrothermal fluid composition (geobrines) and stratigraphy (rock formation) playing important roles. This study has focused on interactions of several cement slurries with fresh ignimbrite, one of the most common rocks found in NZ geothermal fields.

Little has been published on the cement/rock interaction that occurs in hydrothermal conditions. However, from several references (Poole, 1992; Criaud et al., 1994; Criaud and Defossé, 1995; Andrei and Criaud, 1996), it is reasonable to suppose there is some similarity with the reaction products in ASR at ambient, even though the high temperature curing can produce modified products, irrespective of the cement hydrates and the microstructure of the cement-rock system.

From their extensive literature review, Hodgkinson and Hughes (Hodgkinson and Hughes, 1999) concluded that the Portland cement main reaction products below 110 °C were the formation of calcium silicate hydrate (CSH) gels, while above 110 °C, the main products were zeolites and feldspars, especially in the presence of NaOH or KOH. Andrei and Criaud (1996) showed that the products of reaction at 150 °C were similar in texture (amorphous and polymorphous gels) to those at ambient conditions with a broad range for the chemical composition. At temperatures above 150 °C, in experiments conducted with alkali hydroxides (NaOH and KOH), zeolites and feldspars rapidly formed with the high pH, causing the alteration of smectite to illite or illite/smectite interlayers (Chermak, 1992; Chermak, 1993; Inoue, 1983; Komarneni and White, 1981; Komarneni and White, 1983; Johnston and Miller, 1984).

In a geothermal environment where the fluids are enriched with high levels of dissolved CO₂, carbonation is a common issue where the dissolved CO₂ interacts with the calcium silicate or aluminosilicate hydrates present in the hydrated well cements. Carbonation results in the formation of CaCO₃, the exact form depending on the compounds present and the exposure conditions. Generally, at 150 °C, any Ca(OH)₂ present carbonates to calcite, while carbonation of C-S-H is slower and with metastable aragonite being the initial crystalline product, which slowly converts to calcite (Milestone et al., 1986a). Carbonation of Ca(OH)₂ is an expansive reaction so the carbonated product occupies more volume than the hydroxide (Milestone et al., 2012b). Contrastingly, carbonation of a low Ca/Si ratio crystalline calcium silicate hydrate such as tobermorite, C₅S₆H₅, results in a solid volume contraction of 33% with a consequent increase in porosity (Milodowski et al., 2011). Milestone et al. (1986b) concluded that the rate of carbonation of a hydrothermally treated cementitious binder varied considerably, and depended on the amount of addition of silica flour (SF), normally added to counter the strength retrogression at high temperatures (over 100 °C), as well as the phases present. In their experiments, they showed that the crystalline, low Ca/Si ratio phases of tobermorite and xonotlite that form with addition of ca. 40 wt% SF to give binders with low permeability and high strength, carbonated rapidly and became porous when exposed to CO₂. This contrasts with the samples made with neat Portland cement, where the high Ca/Si ratio phases, α-C₂SH at 150 °C and reinhardbraunite above 200 °C, behave better when exposed to CO₂. Despite being poor binders, carbonation of these high Ca/Si ratio phases was slow and a dense protective carbonation sheath with low permeability and high strength formed around the substituted. The carbonation rate is very dependent on the volume of available Ca in any unit volume, particularly if any Ca(OH)₂ was present. Thus the impact of CO₂ on the durability of cement is extremely dependent on the overall Ca/Si ratio (Milestone and Aldridge, 1990).

Hodgkinson and Hughes (1999) were among the first to recognise the significance of the effect that the CO₂ on the cement/rock system at ambient temperatures: CSH gels and portlandite have a tendency to

become carbonated and calcite readily precipitates whenever the hydroxide fluid comes into contact with a carbonate source. In his experimental model consisting of sandstone cylinders filled with class H neat paste exposed to CO₂, Duguid (2009) observed a general degradation pattern where a single visible reaction zone formed, which progressed over time. In his experiments carried out at 50 °C, the cement invaded the stone to a depth of 0.8–1 mm.

The goal of this paper is to enhance the understanding of the role that the type and amount of silica addition and CO₂ exposure play on the cement/rock interaction within typical New Zealand geothermal environments.

2. Materials and methods

2.1. Raw materials

The rock used was Ongatiti ignimbrite (abbreviated by IGN), obtained from the Hinuera quarry. This material is mostly an amorphous glassy deposit with ca. 15% plagioclase feldspars and 1.5% quartz. API Class G cement (Holcim), quartz silica flour (SSA-1) and Microsilica 600 (mostly amorphous silica) were used to prepare the cement formulations. These are respectively abbreviated by G, SF and MS. The XRF analyses of these materials are shown in Table 1.

The particle size distribution analyses obtained for the cement and admixtures are given in Fig. 1. G and SF have a very similar particle size distribution with most particles lying within the 0.5–90 μm diameter range. MS on the other hand, shows a different particle size distribution, with a significantly lower average particle size compared to G and SF.

2.2. Methodology

The four different cementing formulations used to examine rock/cement interactions were API Class G cement (G), API Class G + 20%wt silica flour (G20SF), Class G cement + 40%wt silica flour (G40SF), and Class G cement + 20% Microsilica 600 (G20MS).

Twenty-five mm diameter cavities were cored in small blocks of rocks, which were immersed in a geobrines solution¹ at 90 °C for several days to equilibrate the pore water. Cement slurries were prepared according to API RP10 at a water solids (w/s) ratio of 0.45 and poured into different holes. The assemblages were then set/cured for 2 days in geobrines at 90 °C before autoclaving for 82 days at 150 °C at steam pressure of 11 bar, giving a total of 84 days of curing pressure.

In the autoclave, the specimens were fully immersed in geobrines. After closed and sealed, the autoclave was heated gradually until the desired inner temperature was reached, which took a couple of hours. The experiment was made without any gas exchange with the exterior, except for the experiments with CO₂ injection, where a CO₂ overpressure of 6 bar (0.042 mol/kg dissolved CO₂) was maintained.

After curing, each cement-rock system was cut in half, photographed and samples prepared for X-ray diffraction (XRD), Transmitted Light Optical Microscopy (OM), Scanning Electron Microscopy with Electron Dispersive Spectroscopy (SEM/EDS) and Thermogravimetry/Mass spectrometry (TG/Mass Spec).

The instrument used for XRD was a Philips PW1700 series diffractometer running Co K alpha radiation Long Fine Focus tube with tube power settings of 40 kV and 35 mA. SEM images were taken on a Quanta 450 SEM equipped with a tungsten filament. The elemental

¹ The hydrothermal solution (geobrines) utilized is that used in the experimental work undertaken by the Callaghan Innovation (previous IRL) cement research team (Milestone et al., 2012a). The recipe is based on a geothermal fluid assay supplied by Mighty River Power in 2009, and prescribed by GNS Science in February 2010: 0.19 g sodium sulfate (Na₂SO₄), 0.05 g calcium chloride dehydrate (CaCl₂·2H₂O), 15.6 g liquid precipitated silica (SiO₂), 4.1 g potassium chloride (KCl), 15.8 g sodium chloride (NaCl) are well mixed and filled with water to make up 20 L.

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