

Bonding performance of cement sealants designed for CCS applications

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Keywords: CCS, cement plug, bond integrity, interface condition.

Summary:

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Abstract

Sealant/casing bond integrity is considered an important performance characteristic of sub-sea wells repurposed for Carbon Capture and Storage (CCS) operations, yet there is still lack of general agreement on how this should be assessed. This study presents an investigation into the shear bond performance of four cementitious sealants that are intended for use in the plugging of CCS wells. To this end, a series of samples designed as miniature versions of a plugged well-bore, comprising an outer cylindrical steel casing and an internal cement plug, were configured. These sealant/steel samples were then cured at elevated temperatures and pressure to simulate subterranean conditions, before undergoing a push-out shear-bond test. Bond strength was found to be directly influenced by the type of sealant and the condition of the sealant/casing interface observed after testing. Factors affecting bond integrity are discussed for each sealant in light of the results of shear-bond and compressive strength tests, as well as the condition of the sealant/casing interface.

Keywords: CCS, cement plug, bond integrity, interface condition.



1. Introduction

Carbon capture and storage (CCS) refers to a set of process technologies deployed to facilitate the capture, transport and sequestration of CO_2 underground, typically in deep man-made and natural geological formations (e.g., depleted oil wells and aquifers). According to the International Energy Agency (IEA), it is estimated that the current capacity of large-scale CCS facilities is 50 million tonnes of CO_2 per annum (Mtpa CO_2), with an additional 7 Mtpa of CO_2 facilities currently under construction [1]. **Figure 1** displays the recent and projected global capacity of large-scale CCS operations, along with the breakdown of current and projected capacities across early and advanced stages of developments. Once these planned facilities become operational, the global CCS capacity per annum is forecasted to exceed 100 Mtpa of CO_2 .

The development of major permanent storage infrastructure is clearly required to achieve the proposed CCS targets. Depleted oil and gas fields are a good candidate in this regard as they often have large storage capacity and extensive infrastructure in place that can be repurposed for CO_2 transport and storage. According to the recent OEUK estimate, within the UK Continental Shelf alone, almost half of 283 active oil fields and gas reservoirs will cease their oil productions by 2030 [2] and many of these fields have the potential to be repurposed for CO_2 storage. Similarly, within the Norwegian Continental Shelf of the North Sea, there are a vast number of oil and gas wells that are currently being considered for potential CO_2 storage, due to their proximity to the European mainland [3].

Given the projected timescales envisaged in the operation of CCS storage facilities (well in excess of the lifespan of traditional oil and gas wells), minimising the leakage rate of CO_2 is of utmost importance for effective performance [4]. As shown schematically in **Figure 2**, there are a number of potential leakage pathways that need to be addressed. Such pathways include leakage through the cement plug itself either via direct permeation or internal fractures, through the external/internal surface of the wellbore casing, through cement sheath around the well, or through the interface between cement sheath and surrounding rock formations.



It is clear that optimal adhesion of the well-plug material to both the external rock formation and the well-bore casing is essential. Historically, the adhesion of well-plug materials to the surrounding casing and/or formation has been assessed using a variety of bond tests (e.g., shear bond test, hydraulic bond test, gas bond test, or a combination thereof) [5-10]. Shear bond measures the shear strength of an interface and is generally obtained by measuring the force required to initiate movement of a cement plug encased in a metal cylinder, or a pipe/rod embedded in a cement cylinder [5-9]. Hydraulic bond evaluates the resistance between cement and surrounding casing and/or formation to fluid migration [5,6,9] and is typically assessed by measurement of the flow of hydraulic fluid through an interface over time. The principle of gas bond is similar in many respects to hydraulic bond but relies on the measurement of gas migration instead of fluid migration [10]. Due to the nature of the tests, hydraulic and gas bond can be expected offer a more direct representation of interface leakage performance. However, the tests are generally much more difficult to perform and more sensitive to experimental errors caused by gas leakage from the pressure containment cell and ballooning effects [10].

Evans and Carter [5] were amongst the first to utilise the shear bond test to study the shear bond properties of different cement systems interfaced to metal casing and to two types of formation (sandstone and limestone). They found bond strengths to lie in the range 0.07–2.76 MPa. In a follow up study, Carter and Evans [6] noted that shear and hydraulic bond strengths are influenced by a range of factors. Both bond parameters were found to increase with increasing surface roughness, to decrease with surface contamination (such as mud, oil and drilling fluids), to increase with increasing internal pressure and temperature, and to decrease with casing expansion. In addition, they found that bond strength increases with the viscosity of the injecting fluid, resulting in a slower bond failure progression. Apart from surface roughness, Kakumoto and co-workers [7] found that effective confining pressure has an important role to play. They found that the mean bond strength of a steel rod with a surface roughness of 2.8 microns (embedded in cement cylinder) increased linearly from ~0.9 MPa at zero effective confining stress to almost 2.5 MPa at 3 MPa effective confining stress. The same trend was observed from steel rods with a surface roughness of 17 microns, giving the corresponding bond strengths of ~2.5 MPa and ~4.8 MPa, respectively. A similar trend was reported by Yoneda *et al.* [8].



More recently, Kamali and co-workers [9] performed both shear and hydraulic bond tests on three different sealant materials. The specimens for the shear bond test had both inner and outer steel casing, whereas those for the hydraulic bond test had only external steel casing. Clean machined and corroded steel were used in both series of the experiment to represent possible varying conditions on site. They found that regardless of the type of sealant used, the mean bond strengths obtained from samples with embedded pipe were in the range 1.4-3.1 MPa and higher than those obtained from samples encased in metal casing, which produced mean bond strengths in the range 0.6-1.5 MPa only. They attributed the difference to the effect of cement shrinkage. They also found that corroded steel surface resulted in higher mechanical and hydraulic bond properties. No correlation was however found between the shear bond and hydraulic bond strengths obtained from the different sealant materials tested, up to a differential pressure of 34.5 bar (i.e., the maximum pressure considered in their hydraulic bond tests due to safety concerns). It is paradoxical that although a variety of investigative techniques have been developed to study the bond properties of various sealant materials, these have often relied on the use of specialist equipment, thereby hindering widespread adoption. Moreover, the evaluation of key properties of materials are often required to be done locally. A simple test procedure that is technically less specialised would be useful, particularly in laboratories where access to sophisticated equipment is limited.

In this article, we present a laboratory-based test procedure designed to evaluate the bond strength of cementitious sealants encased in rigid (in this case, metal) cylinders. Major advantages of the procedure are its reliability, conceptual simplicity, and ease of implementation within the range of standard testing facilities available in a general cement/concrete testing laboratory, thus making it a more widely available tool to researchers. Here, details of the test methodology are described along with its application to the assessment of the bond performance of four cementitious sealants, including a newly developed proprietary blend designed specifically for CCS well-plug applications. Factors influencing the shear bond strength obtained from the procedure are discussed.



2. Experimental Programme

2.1 Materials

The cementitious sealants used in this experimental programme are presented in **Table 1**, together with their respective specific gravity and technology readiness level (TRL). Sealant S1 is a representative of material used for old oil and gas wells and contains a standard Portland cement Class G [11] (supplied by Dyckerhoff) and silica flour (35% by weight). This sealant is included in this work as a baseline reference. The other sealants are proprietary materials whose details cannot be fully disclosed. Sealant S2 is also a representative of a wellbore plugging material, containing an expansive agent and mineral addition, designed particularly to produce very low permeability [12]. Sealant S3 is a modified version of Sealant S2 and contains additional CO_2 sequestering mineral additives, namely RePlug[®] [13]. S4 is a proprietary calcium aluminate system currently deployed in high temperature wells and considered highly acid resistant [12]. Both Sealants S3 and S4 are designated proprietary blends for new CCS wells. A typical oxide composition of the Portland cement is presented in **Table 2**.

The test sample for the bond test took the form of miniature versions of a plugged wellbore, comprising an outer cylindrical steel casing and an internal cement plug (see the schematic diagram in **Figure 3**). The steel casing had a length of 50 mm, an outside diameter of 50.8 mm, and a wall thickness of 3.2 mm. The use of mild steel casing (Grade CFS 3BK) instead of super duplex steel (used typically for CCS well) was sought by the project consortium at the onset of the project to allow for possible occurrence of corrosion to take place (thereby simulating long-term operation), in addition to facilitate the machining of the two ends of the casing.

The internal sealant cement plug had a diameter of approximately 44.4 mm and a 10 mm protruding end on either side, which had a diameter of 41 mm. These protruding ends were provided to minimise the boundary effects during mechanical bond testing: the top part was provided to remove the undesirable effects from the presence of relatively poor quality of material and uneven surface at the top during sample preparation, whereas the bottom part was to minimise the effect of high concentration of stresses under the loading plate during testing. The manufacture of this protruding shape was



facilitated by two PTFE end caps which encapsulated the sealant during sample fabrication and curing. The top PTFE cap had a 20 mm central hole for casting purpose, while the bottom cap was fitted without any alteration to provide a flat surface for bond testing.

2.2 Sample fabrication and curing

Prior to the fabrication of the main test samples, each steel tube was cut to length (i.e., 50 mm) and then immersed in a water-based degreasing solution (Ambersil degreaser concentrate, diluted at 1:6 ratio with tap water) for 24 hours. The inner and outer faces of the tube were then thoroughly wiped with paper towels. This process was repeated one time to ensure that any oils that were present on the inner face of the tubes (required for cutting and milling) were fully removed. After thoroughly cleaned, the steel tubes were then transported to Norway for sample fabrication purpose.

A total of eight samples were produced for each sealant material, six samples for bond testing and the remaining two samples for electrical testing (discussed in a parallel publication). All test samples were prepared by the technical team at Halliburton in Norway, in accordance with API Recommended Practice 10B-2 [14]. Sealants S1 and S2 were manufactured on the same day, whereas Sealants S3 and S4 were manufactured a few months after. The fabrication of all sealants followed the same sample preparation procedure to allow for direct comparisons.

For each sealant, casting was done by syringing fresh slurry into a 20 mm hole on the top cap, which was then sealed using a 3D-printed plastic cap. All samples were subsequently stacked inside a stainless-steel autoclave chamber. As illustrated in **Figure 4**, the following curing regime was adopted:

- (i) The internal pressure was first increased to 300 bars
- (ii) The internal temperature was then raised from ~20 to 80°C over a period of 4 hours
- (iii) The internal temperature was held at 80°C for 3 days before being raised again to 150°C over 7 days before being held constant at 150°C for a further 21 days



(iv) The internal temperature was lowered from 150°C to 20°C over a period of 7 days. During this stage, the internal pressure regulator was released when the temperature reached 100°C to allow for the chamber to reach equilibrium depending on the temperature level.

Stage (ii) was considered to simulate general cementing, where cement slurry is typically injected into a wellbore typically at ambient (low) temperatures. The temperature of the slurry can be expected to increase at greater depths, eventually reaching equilibrium with the temperature of the surrounding rock formations, which, for a 2 to 3 km deep offshore reservoir, could range from 80°C to 120°C [15,16]. The 80°C temperature was selected in this work based on the minimum CCS operational temperature and practicality during bond testing.

The high-temperature curing employed in Stage (iii) (i.e., 150°C) was intended to enable the sealants to achieve near or full hydration within a reasonable timeframe, thereby avoiding further changes in properties post curing. Subsequent electrical monitoring confirmed that maintaining 150°C over the 3-week duration was adequate. It should be noted that the internal autoclave temperature was gradually increased to 150°C and then steadily lowered back to 20°C, to minimise thermal effects on the interfacial bond during the heating and cooling stages. At the end of the curing regime, all samples were wrapped in wet clothes and airlifted back to the UK for subsequent bond testing. These samples were kept in a water bucket upon delivery and stored in a temperature-controlled laboratory (20±1°C) until required for testing.

2.3 Shear bond testing

One day prior to bond testing, all samples were pre-conditioned in an environmental chamber to increase the sample temperature back to 80°C over 24 hours. During this pre-conditioning stage, all samples were kept in water to buffer and stabilise the temperature. Bond testing was then performed using a 100kN Instron 5892 testing machine. The test apparatus utilised in the experiment is shown schematically in **Figure 5**, with test procedures essentially following those detailed in Patent No. NO20191422 (US11054353B2) [17]. Prior to the commencement of each test, each sample was removed from the chamber and then immediately (tightly) wrapped with a 30 mm-thick pipe insulation



to minimise temperature loss during bond testing. The sample was then placed on a central-hollow steel bracket (also pre-conditioned at 80°C) in an upside-down position to present the flat face of the sample to the loading plate on top. The load was then applied through a hemispherical steel plate placed on the top of the sealant in a force-controlled manner, at a rate of 5kN/min. The machine automatically stopped when a sudden drop of resistance (i.e., by more than 20%) occurred, due to either sliding or crushing of the inner cement plug.

The apparent mean bond stress, τ (MPa), during testing can be calculated as

$$\tau = \frac{P}{A} \tag{1}$$

where *P* is the applied load (N) and *A* is the contact area between the sealant and steel casing ($\approx 44.4 \times 50 \text{ mm}^2$).

4. Results and discussion

4.1 Mean Bond Strength and Stiffness

Figures 6(a)-(c) display the mean bond stress vs top displacement relationships for the four sealants, with each Figure comparing the results of three notionally identical samples for each sealant with those of Sealant S1. In general terms, all sealants displayed a similar increase in the mean bond stress with increasing displacement until failure, which occurred suddenly due to sliding of the sealant in the vertical direction. The individual sealants exhibited varying bond strengths and overall stiffnesses. For reasons of clarity, the mean apparent bond strength and stiffness, and their respective standard deviation (SD) and coefficient of variation (CoV), are presented in **Table 3** for the four sealant types.

With reference to **Figures 6(a)-(c)**, a general reduction in the overall stiffness and bond strength is evident compared to those of Sealant S1, with both parameters decreasing in the order: S1 < S3 < S2 < S4. For instance, in case of Sealant S2, the mean bond strength and stiffness were 1.51 MPa and 8.95 N/mm³, which are only 32.5% and 80.6% of those of Sealant S1, respectively. Sealant S3 exhibited higher mean bond strength and stiffness, with mean values of 3.54 MPa and 9.20 N/mm³, corresponding



to 75.9% and 82.9% of those of the reference value, respectively. Sealant S4 demonstrated the lowest mean bond strength (= 0.54 MPa), which is only 11.5% of that of the reference sealant, and the lowest overall stiffness (= 2.88 N/mm³, or only 25.9% of that of Sealant S1). This might be attributed to sliding shear movement between the sealant and the steel casing during testing.

Direct comparisons of the mean bond strengths obtained from previous studies are difficult due to the scarcity of published values, with most samples being cured at ambient or limited pressure. However, some comparisons are provided in Figure 7(a). Note that in this Figure, the x and y error bars represent one standard deviation, and in cases where the error bar appear to be missing, they are smaller than the markers themselves. Additionally, a few data obtained from metal pipes encased in sealant, which tend to produce higher bond strengths [9], are included for comparative purposes. It is interesting to note from Figure 7(a) that the mean bond strengths of Sealants S2 and S4 lie within the range of reported values, whereas those of Sealants S1 and S3 are generally higher. Interestingly, the mean bond strength of Sealant S2 (= 1.51 MPa), which contains expansive agent, is comparable to the 1.5 MPa average reported by Kamali et al (2022) [9], which was obtained from cement test samples containing magnesium oxide expansive agent. By contrast, the mean bond strength of Sealant S1 (=4.66 MPa) is much higher than the 0.6 MPa average obtained from the same study, measured on Portland cement (type G) cement samples encased in metal casing; these samples were cured in 90°C chamber at 34 bars for only 7 days. The mean bond strength of Sealant S1 is also higher than the 1.4–3.1 MPa range obtained from metal pipe encased in sealant [9], and the range 0.12-0.26MPa reported by Mabeyo et al. (2020) [18] for cement samples encased in cement mortar and cured in 80°C water bath at ambient pressure over a 28-day period. Therefore, it is possible that the lower bond strength of Sealants S2 and S4 observed earlier in Figures 6(a) and (c) is not due to their reduced strengths, but rather because of the elevated bond strengths of Sealant S1 and S3.

The overall stiffness of the curves presented in **Figures 6(a)-(c)** can primarily be attributed to the deformation of the top protruding part of each sample under loading and the initial slip between the sealant and the metal casing during bond testing, in addition to the deformation of the entire test setup,



which can be expected to be similar for all samples. Therefore, comparing the overall stiffness obtained from each sealant with their modulus of elasticity (MOE) could provide an indication of the extent of initial slip. **Figure 7(b)** compares these parameters, displaying the overall stiffness obtained from each sample plotted against their respective MOE value. Note that the MOE of each sealant was measured in a separate experiment on notionally identical materials cured at 150°C and 300 bar for 28 days, producing compressive strengths of 107, 76, 32, 21 MPa for Sealants S1 to S4, respectively. It is evident from **Figure 7(b)** that the overall stiffness obtained from bond testing exhibits an almost linear relationship with MOE. The only exception is Sealant S3, which displayed higher stiffness than the linear relationship (indicated by an arrow). This indicates that this sealant must have experienced less sliding during bond testing than the other sealants. Sealant S4 exhibited slightly lower stiffness than the average (represented by the trendline), suggesting the occurrence of comparatively greater initial slip during bond testing. To understand the contributing factors, the internal sealant plug was extracted from the metal casing post-testing and is discussed below.

4.2 Inner Surface Condition and Failure Mechanism

On completion of the bond testing, each sample was machined to extract the internal cement plug from the steel casing to allow the condition at the sealant/steel casing interface to be visually inspected. This was done by milling into the outer surface of the steel casing to create a longitudinal recess steadily until it cut through the entire thickness of the steel. Upon completion of this process, the cement sealant was extracted and then photographed. This was done by placing a sealant sample at a fixed distance from a DSLR camera and taking multiple photos while rotating the sample. Twelve images were acquired from each sample to ensure sufficient overlap between images for stitching. These images were then corrected for distortion in Adobe Photoshop [19] and stitched together using the Photomerge feature in the software to create a seamless panoramic image.



Figure 8 displays the acquired images from each sealant. Sealant S1 displayed a significant presence of corrosion products on the entire surface of the sealant, primarily in dark brown and black colours, which indicates the result of oxidation in a low moisture and oxygen environment [20]. Few patches of the original surface of the sealant were apparent as the rust layer came off during the extraction, or due to the milling process as shown in the right-hand side. Sealant S2 displayed an interesting surface pattern, comprising small white/light-grey patches dispersed across a darker grey background. On examination, the white/light-grey zones were found to be the material that had been directly adhered to the steel surface but had separated under the application of shear load. The darker background is the exposed sub-surface of the sealant material immediately beneath the sealant/steel interface, an effect resulting from the separation of the still adhering sealant surface from the sample body (see **Figure 8**). Thus, it is inferred that apparent bond failure of the S2 samples results from a combination of material failure beneath the steel/material interface, and *partial* failure of the sealant/steel bond.

Sealant S3, like Sealant S2, displayed only a minor extent of corrosion, concentrated primarily at both ends of the sample. In contrast, however, Sealant S3 exhibited a clean surface representing the appearance and texture of hardened cement paste in general (see **Figure 8**). As shown in **Figure 9**, the inner surface face of the tube was also relatively clean, with a minor extent of corrosion present. This would indicate that failure of Sealant S3 samples was governed by bond failure at the interface between the sealant and metal casing. Sealant S4 displayed a moderate level of corrosion (brighter in colour) and the presence of a failure surface where a layer of the sealant material near the interface spalled off. Other than these regions, the remaining surface of Sealant S4 appeared to be clean.

4.3 Effect of Corrosion

Due to the expansive nature of corrosion [21], the formation of corrosion products in a confined space, such as at the sealant/casing interface, can be expected to lead to the development of internal confining stress which would, in turn, increase bond strength. To understand the effect of confining stress on bond strength, two additional series of tests were undertaken. The first series involved the fabrication of test



samples at laboratory temperature of 20°C and normal atmospheric pressure, whereas the second series involved the fabrication of test samples at enhanced temperature and pressure as before.

In the first series, seven test samples were cast using the same mild-steel tube casing. Each sample contained 2.4mm diameter stainless-steel rod embedded along the longitudinal axis of the sample at the time of casting. These samples were stored under water for 25 days. As illustrated in the schematic diagram in **Figure 10(b)**, the lower part of the samples was then immersed in water in a small plastic container. On the same day (Day 26), four of the samples were connected to a DC power supply to artificially induce corrosion. This was done by connecting the stainless-steel central rod to the positive terminal of the power supply (acting as a cathode) and the metal casing to the negative terminal (acting as an anode). Two of the samples were exposed to a constant current of 15mA and the other two to 30mA, corresponding to an average current density of 0.21 and 0.42 mA/cm², respectively. They were connected to the power supply for 6 days, followed by 1 day rest to allow for various measurements to be undertaken (discussed in another article). This continued on a weekly basis until required for bond testing at 90 days. The remaining three samples were left unconnected and used as a benchmark.

Figure 10(a) displays the 90-day mean bond stress vs top displacement relationships for the three groups of sealants, with the tests performed at 20°C and under a crosshead speed of 0.2 mm/min. It is evident that there is a noticeable difference in bond performance between the three groups of samples in terms of bond strength and overall stiffness. In general, an increase in the 90-day mean bond strength with increasing current levels is evident, from 0.91 MPa at 0mA to 1.19 MPa at 15mA (or 30.8% increase) and 3.18 MPa at 30mA (349% increase). This confirms the above postulate that the enhancement in bond strength is affected by the extent of corrosion at the sealant/casing interface.

Further evidence of the effect corrosion could be obtained from the second series of samples. Three samples were cast: two using the same mild-steel tube casing and the other using stainless-steel mould. These samples were cured at enhanced temperature and pressure, following the curing regime shown previously in **Figure 4**. They were also tested using the same test procedures, involving sample pre-



conditioning and testing temperature of 80°C. **Figure 11** displays the ratio of the mean bond strength of samples encased in corroded mild-steel casing to that encased in stainless steel casing. It is evident that corrosion at the interface caused an artificial increase in bond strength (in the range 22-39%), thereby further reaffirming the above hypothesis on the secondary effect of corrosion on bond strength. It is of interest to note that even tough corrosion is shown to result in an elevated bond strength, due to the presence of confining pressure [7,8], this effect is considered non-permanent and is expected to gradually disappear in the long term due to the creep and shrinkage of the sealant over time [22]. Hence the elevated bond strengths observed from test samples with corroded metal casing can be considered to be *artificial*. Due to the expansive nature of corrosion, coupled with creep and shrinkage of the sealant over time, the build-up of corrosive layers on the inner casing surface may increase in porosity when the corrosion progresses at more advanced stages [21]. This layer may therefore eventually turn into leakage paths. It is therefore recommended to avoid corrosion in the first instance i.e., through the use of non-corrosive casing and sealants that do promote corrosion over long-term.

4.4 Effect of Setting Time

During the initial few hours after gauging, cement slurry undergoes setting (increasing in rigidity and transitioning from a liquid to a solid state) [23]. Although no direct measurements of setting time were undertaken, it was observed that the setting time of the four sealants at ambient laboratory temperature varied significantly, increasing in the order: S4 (less than 30 mins) < S2 (around 45 mins) < S3 and S1 (more than 1 hour). Coincidentally, this aligns with the order of the mean bond strengths obtained from the four different sealants. Given that cement slurry with a quick setting time can be expected to solidify at a faster rate, particularly when exposed to a rapid temperature increase (simulated here by the rapid initial temperature rise from 20°C to 80°C in 4 hours), it is postulated that a rapid state change may compromise bonding with the metal casing. Moreover, in this work, despite using high pressure (300 bar) curing, the situation is likely further exacerbated by the expansion of the metal casing during the initial heating stage and the absence of lateral confinement provided by surrounding rock formations.



In view of above, it was considered necessary to study the effect of setting time. The testing for Sealant S4 was repeated to check if the low bond strength observed was attributed to its rapid setting characteristic. Hence, in the repeat test, a retarding agent of undisclosed type and quantity was added to the original mix to slow down the initial reaction kinetics of the cement. The same test procedures, with elevated curing temperature and pressure, were implemented as previously. However, prior to bond testing, the material protrusion above the casing rim, through which the bond test load is applied (see **Figure 3**), was found to be cracked radially. This required milling of the sealant material flush with the steel casing rim, to enable load application during the bond test. The mechanism underlying the radial cracking is not understood but might have resulted from tensile stress developed in the protrusion above the steel rim due to the restraint on expansion of the sample body during curing imposed by the steel casing.

Figure 12 compares the mean bond strength obtained from this repeat test (denoted S4R) with the original test (denoted S4). It is immediately apparent that the mean bond strength of the repeat samples was significantly higher than that of the original S4 mix, averaging at 5.45MPa, clearly highlighting the benefits of slowing down the initial reactions of cement on bonding. This corresponds to an approximately five-fold net increase once the difference in the mean bond strengths of the reference samples in the two batches has been considered. Interestingly, it was found that failure of the repeat samples was governed by crushing of the sealant face under the loading applicator instead of shear sliding at the sealant/steel interface. Following bond testing, the metal casing was split into two to get access to the sealant and inner casing surface. Photographs of this are presented in **Figure 12**. Interestingly, there is a large proportion of the sealant material still strongly adhered on the steel surface, confirming the high bond strength observed. Despite this, a moderate level of corrosion, comparable to that observed in the S4 mix (see **Figure 8**), was evident, suggesting that corrosion is governed by the sealant characteristic itself.



4.5 Effect of Curing Conditions

Cement hydration (and the resulting formation of crystalline and amorphous material formation within the hardening matrix) involves complex chemical processes that are thermally activated. The in-situ elevated temperature and pressure environment of a deep re-purposed oil or gas well should therefore be replicated in the curing and testing of CCS sealant materials as closely as possible. Figure 13 compares the mean bond strengths obtained from test samples cured under elevated temperature and pressure (up to 150°C and 300 bar over 38 days, denoted S1 and S1R) and those cured under laboratory condition (20°C and atmospheric pressure over 28 days, denoted S1A), plotted against their respective mean compressive strengths. Note that S1R was obtained from sealant samples encased in stainlesssteel, whereas S1 and S1A are from a similar sealant encased in mild-steel. The beneficial effect of enhanced curing is strongly indicated by the bond and compressive strengths of samples cured under elevated conditions (S1 and S1R) compared to those cured under ambient temperature and atmospheric pressure (S1A). It is interesting to note that while enhanced curing resulted in an approximately twofold increase in compressive strength (~35 MPa to 60-75MPa range), it resulted in four to fivefold increase in bond strength (~0.9 MPa to 3.6-4.6 MPa range). The high pressure (300 bars) used in the main curing regime could be expected to produce sealant pore densification (hence increasing compressive strength) and shrinkage reduction (hence increasing bond strength). The results shown in Figure 13 suggest that the effect of enhanced pressure plays a greater role in shrinkage reduction than in pore densification.

7. Concluding Remarks

Shear bond tests were undertaken to assess the bond performance of four distinct cementitious sealants contained within cylindrical steel casing. Samples were cured at elevated temperature (up to 150°C) and pressure (300 bar) to provide conditions representative of deep offshore CCS wells. From the results presented, the following remarks can be made:

- The sealant/steel samples exhibited varying bond strengths, ranging from 0.54 MPa for Sealant S4 to 4.66 MPa for Sealant S1.
- 2. The shear bond strengths of cementitious sealants are influenced by a range of factors. In this work, curing condition, sealant setting time, and corrosion at the interface between the sealant and steel



casing emerged as the dominant factors. Although compressive strength was found to influence bond strength, it did not guarantee good bonding performance, as evidenced by the result for Sealant S2 samples.

- 3. The high bond strength of Sealant S1 samples was found to be related to the presence of internal confining pressure caused by the formation of corrosion products at the inner surface of the metal casing. This artificial enhancement of bond strength is expected to gradually disappear in the long term due to creep and shrinkage of the sealant matrix and cannot, therefore, be regarded as a performance indicator.
- 4. Sealant S3 was found to have comparatively better sealant/steel bond strength despite its moderate compressive strength (lower than Sealants S1 and S2). Combined with its known CO₂ sequestering capability, the elevated bond strength of Sealant S3 suggests that this sealant may perform better as a CCS wellbore plug.
- 5. The extent of corrosion was observed to be sealant specific and most prominent in Sealant S1 samples. Sealant S3 exhibited the lowest level of corrosion and the smoothest de-bonded interface, while Sealant S4 displayed a moderate level of corrosion both in the original experiment and in a repeat test with higher bond strength. The use of stainless-steel casing in future experimental work is recommended to isolate corrosion from other factors.
- 6. The elevated curing temperature and pressure employed in this work was found to significantly increase both the shear-bond and compressive strengths of the sealants. The high curing pressure was particularly effective in reducing volumetric changes (e.g., due to cement shrinkage), thereby enhancing the measured bond strength.
- 7. For Sealant S4, the use of a retarding agent in a repeat experiment was found to significantly enhance the bond with the steel casing (a relative fivefold increase in strength) but did not alter the level of corrosion. While the dosage level of retarding agent used in the repeat S4 experiment was not disclosed, it is possible that its addition to the other sealant materials could enhance their bond performance.



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Table 3. Summary of bond test results for the four sealants.

Sealant ID	Description	SG	TRL
S 1	Standard Portland cement (PC)-silica blend	1.90	7
S2	Reduced permeability PC-silica blend for field use	1.90	7
S3	PC-silica blend with reduced permeability, containing CO ₂ sequestering mineral additive RePlug [®]	1.90	3
S4	Calcium aluminate cement-based blend	1.80	7

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CaO	SiO ₂	Al_2O_3	Fe_2O_3	MgO	MnO	TiO ₂	P_2O_5	Sr	SO_3	K ₂ O	Na ₂ O
62.0	19.63	4.08	5.94	1.19	0.12	0.25	0.08	0.17	4.08	0.81	0.17

Table 3. Summary of bond test results for the four sealants.

Sealant ID	Mea	n bond stre	ngth	Overall Stiffness			
	Mean	SD	CoV	Mean	SD	CoV	
	(MPa)	(MPa)	(%)	(N/mm ³)	(N/mm^3)	(%)	
S1	4.66	0.48	10.3	11.10	0.23	2.04	
S2	1.51	0.09	6.0	8.95	0.20	2.28	
S3	3.54	0.62	17.6	9.20	0.24	2.63	
S4	0.54	0.34	63.5	2.88	0.84	29.4	