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Investigating the Effects of Hydrogen on Wet Cement for Underground Hydrogen Storage Applications in Oil and Gas Wells

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***Abstract*—**Green hydrogen is quickly emerging as a new source of the renewable energy for the world. Hydrogen production using water electrolysis is deemed as an environmentally friendly and safe source of energy for transportation and other industries. However, storing high volumes of hydrogen seems to be a significant challenge. Abandoned hydrocarbon reservoirs are considered as viable hydrogen storage options because of the availability of the required infrastructure such as wells and surface facilities. However, long-term wellbore integrity in these wells could be a serious challenge. The aim of this research is to investigate the effect of stored hydrogen on the wellbore integrity such as casing cement. The methodology is to experimentally expose hydrogen to wet and dry cement and measure the impact on cement rheological and mechanical properties. Hydrogen reduces the compressive strength of a set cement if it gets in contact with the cement slurry. Also, mixing hydrogen with cement slurry slightly increases its density and rheological properties which need to be considered to have a successful primary cementing operation.

***Keywords*—**Green hydrogen, underground storage, wellbore integrity, cement, compressive strength.

I. Introduction

HYDROGEN is the future planed fuel for the world. Hydrogen is steadily growing as a new source of green and cleaner energy [1]. Hydrogen is an environment friendly and safe source of energy that could be used in several industries such as transportation. Many countries have already started conducting research studies to economically produce hydrogen as a source of energy [2]. Hydrogen can be produced from multiple sources including natural gas, coal, biomass, and electricity (water). Since water occupies two thirds of the surface of the earth, it is deemed as the future source to produce hydrogen using water electrolysis technique. Currently, water electrolysis represents only 2% of the hydrogen production and this is projected to increase rapidly in the near future [1]. Hydrogen, from the different sources mentioned, needs to be produced in huge quantities to accommodate the continuous need of energy, which is currently one of the challenges. Subsurface integrity is a paramount factor in having successful underground storage (UGS), which is the most common type of hydrogen storage [3]. Economically, abandoned oil and gas reservoirs are deemed to be the most viable option for hydrogen storage operations [4]-[7]. The essential part of UGS operation is wellbores.

The depleted gas reserves are simple to develop, manage, and operate due to existing infrastructure. Because of their well-identified geological features, the stability of their caprock, and the pre-existence of the requisite surface and subsurface equipment, they are ideally suited for underground hydrogen storage (UHS) operations. Due to the prior use of such areas for oil and gas production, storing hydrogen in exhausted oil and gas reserves significantly reduces geological exploration efforts.

Since hydrogen is the lightest molecule on the earth, it easily leaks from UGS facilities if proper safeguards are not followed. Drilled wells and other materials used in the oil and gas production are not always ideal for underground H_2 storage, and hydrogen leakage is a potential problem. Additionally, the geologic formation itself may possess channels through which H2 might flow.

Due to hydrogen's strong diffusivity, mixing it with natural gas has impacts on the well integrity and completion equipment. When hydrogen is mixed with methane, the viscosity of the mixture is lower than that of methane alone. The gas will flow through the flaws in a cement seal if the viscosity of the mixture decreases. If hydrogen concentration is low, the leakage is low; nevertheless, as hydrogen concentration rises, the leakage rises as well. Leakages in UGS wells that operate in the presence of hydrogen with natural gas can be reduced by using the finest completion cements with potentially low SiO_3 concentrations in cement binders. The use of low water cement ratios helps prevent the creation of drying voids and low fissures. Since the redox process (H_2/H_2O) is not highly active at low temperatures, the effects of hydrogen on cement barrier sealing characteristics and solubility are believed to be minor. The reverse occurs at high temperatures and depths. The above-mentioned processes are highly dependent on water saturation, formation rock salinity, temperature, pH value, and oxygen presence and concentration. Additionally, microbiological activity can enhance these processes; however, if siderite, dolomite, or calcite are present, these reactions can be hindered.

Injection wells are needed to inject the produced hydrogen into abandoned reservoir and production wells are needed to produce the stored hydrogen as required. To have successful injection and production operations, all the injection, producing and plugged wells must have strong integrity [8]. Otherwise, inefficient storage operation and costly environmental issues

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are inevitable. One of the critical elements of the wellbore integrity is cement sheath behind the casing. Fractures, vugs, cracks and weak points in the cement sheath will lead to hydrogen escape from the reservoir to uncontrolled formations or the surface [9].

There are ample research studies in the literature investigating the well integrity and cement strength in underground CO_2 sequestration in depleted oil and gas reservoirs. For example, salt can be added to cement to increase its strength in these operations [10]. On the contrary, few research have been conducted to study wellbore integrity in UHS sites. Cement shrinkage between the formation and the casing is a common problem occurring in cement leading to gas escape, which makes adding expanding agent necessary [11]. Well screening is a technique that can be utilized to select the right candidates for the storage purposes and repair the problematic ones.

The main objective of this research is to examine the effects of stored hydrogen in reservoir formations on cement rheological and mechanical properties behind the casing in both slurry and set states.

II. Experimental Setup

In this research, effects of hydrogen on cement rheological and mechanical properties were examined. Since cement class "H" is used by nearly 80% of the drilling companies worldwide, neat cement class "H" slurry with the density of 15.5 $\frac{lbm}{gal}$ was used for all experiments conducted in this research study. The API standards for Class "H" cement are presented on

TABLE I.

TABLE I
API Standards for Class H Cement

Typical Mill Run Analysis of Portland Cement		
Oxide	Class G, wt%	Class H, wt%
Silicon dioxide, S_iO_2	21.7	21.9
Calcium oxide, CaO	62.9	64.2
Aluminum oxide, Al_2O_3	3.2	4.2
Iron oxide, Fe_2O_3	3.7	5
Magnesium oxide, MgO	4.3	1.1
Sulfur trioxide, SO_3	2.2	2.4
Sodium oxide, Na_2O		0.09
Potassium oxide, K_2O		0.66
Total alkali as Na_2O	0.54	0.52
Loss on ignition	0.74	1.1
Insoluble residue	0.14	0.21
Phase Composition		
C_3S	58	52
C_2S	19	24
C_3A	2	3
C_4AF	11	15
Phase Properties		
% passing 325 mesh	87	70
Blaine fineness, cm^2/gm	3,470	2,610
Physical Requirements		
Water : cement ratio	1:40	1:38
Thickening time, min.	14	15
Compressive strength, 110 ºF (38 ºC)	928 psi (6.4 MPa)	650 psi (4.5 MPa)
Compressive strength, 140 ºF (60 ºC)	2,247 psi (15.5 MPa)	1,650 psi (11.4 MPa)
Free fluid, mL[(13)]	4.4	4.0

To conduct the compressive strength tests, cement samples were prepared cubes to follow the API procedure. Three cubes were prepared for each sample, shown in Fig. 1, to assure the accuracy of the measurements. TABLE II presents the volume of each component in the cement slurry sample. Each cement sample was prepared according to the procedure recommended in API RP 10B-2, using an OFITE Model 20 programmable constant speed blender. Hydrogen was added to the cement slurry samples during the mixing process. Different exposure times of hydrogen to the cement slurry were examined. Then, the slurry was purred into the three cubes and cured at 120 °F for 24 hours.

TABLE II
Cement Slurry Sample Volumes

Cement type	Class H
Slurry density	15.5 ppg
Slurry volume	500 ml
Cement volume	636.36 g
Water volume	292.16 g

Fig. 1 Cement samples cubes holder (3 cubes per sample)

Measurements of Rheological Properties and Cement Compressive Strength

To examine the effect of hydrogen on the strength of the set cement, three cement slurry samples were prepared with different exposure times with Hydrogen (see TABLE III). Before curing, the density and rheological properties (Plastic Viscosity (PV) and Yield Point (YP)) of each sample was measured. After curing the samples, unconfined compressive strength test was conducted on the samples using an OFITE compressive load frame.

TABLE III
Cement Slurry Samples Prepared (1st Group)

Cement sample	Exposure time with H2
A0	No exposure (as a reference)
A1	20 seconds
A2	2 minutes
A3	4 minutes

X-Ray Fluorescence (XRF) and CT Scan

To better understand the effect of Hydrogen on the cement strength, two more samples were prepared using the same procedure mentioned in the experimental setup. The exposure time of each sample is presented in

TABLE IV. The cube cement samples were taken to a CT scanner (Fig. 2) to make x-ray slices. Also, the crushed samples, resulted from the compressive strength tests, were deployed to conduct XRF analysis to determine percentage of each phase component. pH level of the wet cement slurry was also tested using a pH paper.

TABLE IV
CEMENT SLURRY SAMPLES PREPARED (2ND GROUP)

Cement sample	Exposure time with H2
B0	No exposure (as a reference)
B1	4 minutes
B2	8 minutes

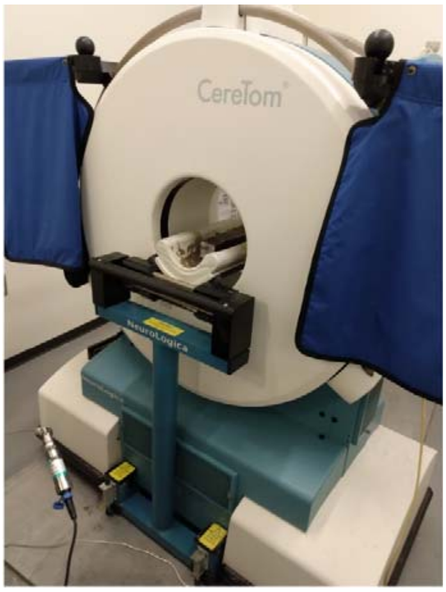


Fig. 2 CT scan equipment

Stability Test

Stability tests were conducted on the samples at the room temperature (71°F) and pressure (14.7 psi) (Fig. 3).

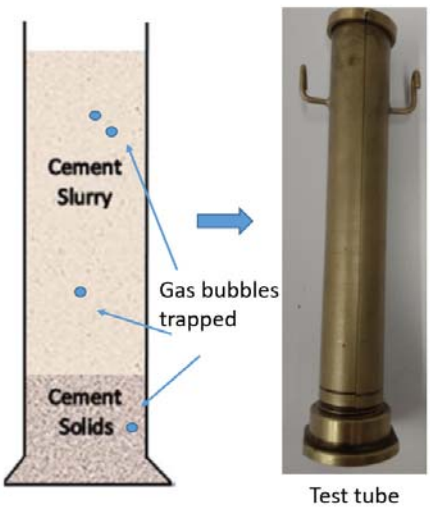


Fig. 3 Cement Stability test tub

The sedimentation test requires freshly mixed cement slurry inside a tube length of 200 mm with an inner diameter of 25 mm. The tube is filled with the slurry prepared samples. A top cover is placed on the test tube to prevent evaporation and then the sample is left in the tube for a 24-hr curing period. After curing, the sample is then divided into four segments (Fig. 4). The samples are then wet cut at each of the marks. Using Archimedes principle, the relative density of each of the four segments are calculated. The first top segment is used as a reference density and then the percentage differences are calculated for the rest of other segments. Two cement slurry samples, one Class "H" neat sample without being exposed to hydrogen and the other slurry being exposed to hydrogen for 8 minutes, were prepared for this test.

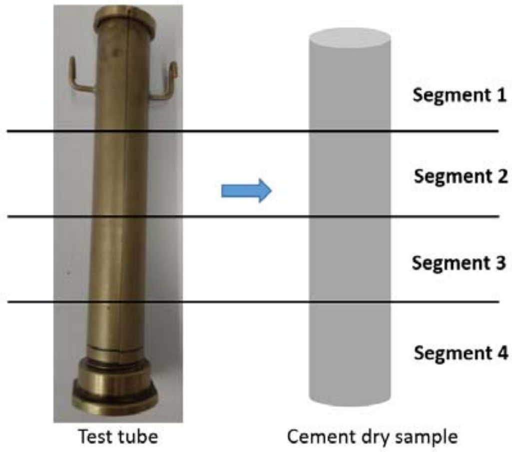


Fig. 4 Cement sample is broken into four segments for density measurements

III. RESULTS

TABLE vV shows the compressive strength results of the three samples; A_1, A_2, and A_3, exposed to H_2 while A_0 was not. As shown in Fig. 5, there is an inverse relationship between the compressive strength of the cement samples and exposure time to hydrogen. Also, the measured density and the rheological properties of the samples are presented in Figs. 6-8. The results demonstrate that there are direct relationships between the density and the rheological properties of the cement samples and the exposure time indicating that there is a chemical reaction between hydrogen and the cement slurry. The pH level (level = 12-13) of the wet sample was also measured after being exposed to hydrogen (Fig. 9).

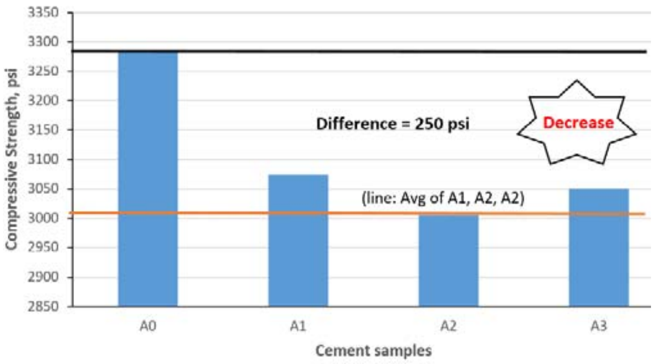


Fig. 5 A plot of compressive strength for the cement samples

TABLE V
COMPRESSIVE STRENGTH (PSI) FOR THE CEMENT CUBES SAMPLES (AVERAGE FOR THE THREE CUBES PER EACH SAMPLE)

Cement sample	Compressive strength, psi
A0	3286
A1	3074
A2	3005
A3	3050

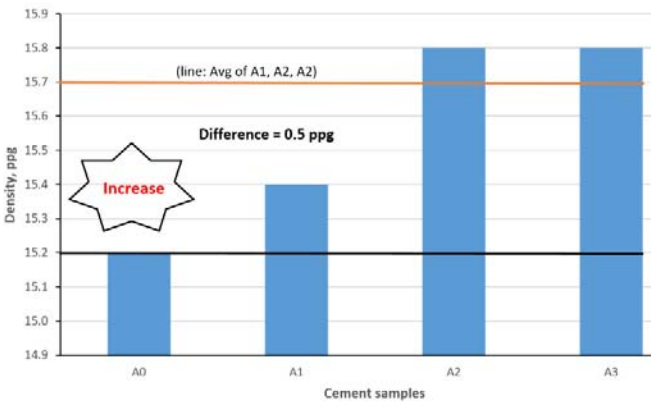


Fig. 6 A plot of density for the cement samples

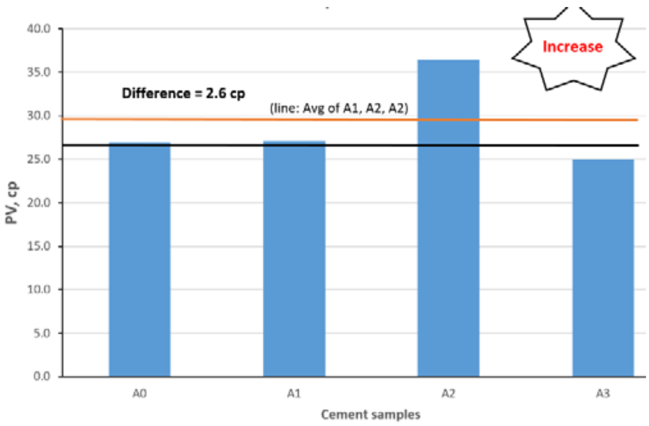


Fig. 7 A plot of Rheology (PV) for the cement samples

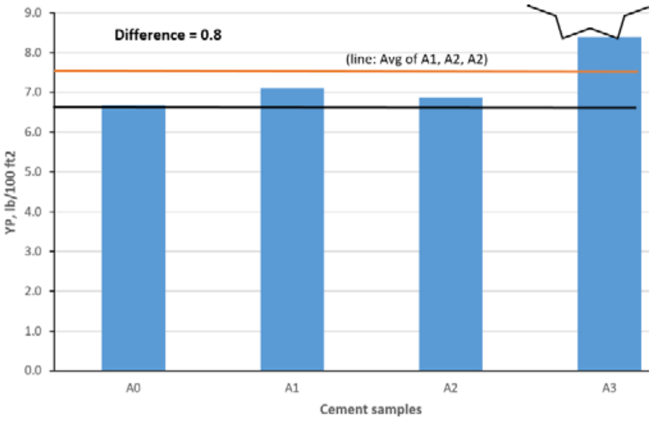


Fig. 8 A plot of Rheology (YP) for the cement samples

Fig. 9 PH level of the cement slurry

Fig. 10 shows the XRF results. The XRF analysis, as shown in Fig. 10, describes the elemental composition of materials of cement powder from which the results show similar composition as listed in

TABLE I. The results in Fig. 11 demonstrate that there is no difference in the pattern except for the portlandite peaks.

The set (dry) cement cubes of the slurries that are exposed to hydrogen and were scanned using the CT scanner (Fig. 2). Hydrogen bubbles, black dots in the images (Fig. 12), are easily noticeable in the CT scan images. The results demonstrate increasing the exposure time resulted in increasing the number of the bubbles, see TABLE VI and Fig. 13. Hydrogen bubbles are trapped in cement results in reducing the strength of the cement.

TABLE VI
PERCENTAGE OF HYDROGEN BUBBLES IN CEMENT CUBES (SELECTED IMAGES)

Cement sample	Cube area, mm x mm	Selected area, mm x mm	Area of Bubbles, %
B2	50.8 x 50.8	41.60 x 46.53	2.00
B1	50.8 x 50.8	44.81 x 41.74	1.06
B0	50.8 x 50.8	43.21 x 38.93	0.20

TABLE VII represents the results of the stability tests measured for two cement slurry samples (with and without being exposed to hydrogen). Calculating the density of all the four segments of each sample, it was observed that there is a difference in the change in density between the neat cement and the cement slurry which was exposed to hydrogen. The percentage difference in density of the segments is shown in Figs. 14 and 15. It is clear that exposing cement slurry to hydrogen makes the set (dry) sample unstable.

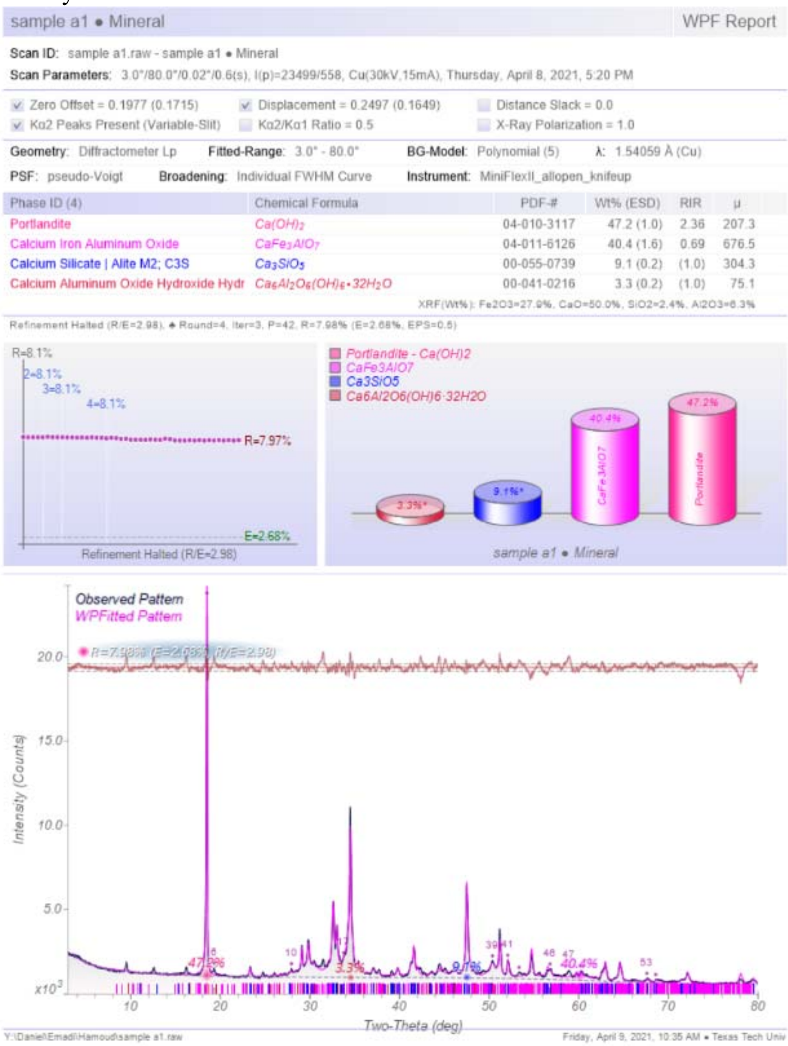


Fig. 10 XRF analysis of the cement powder

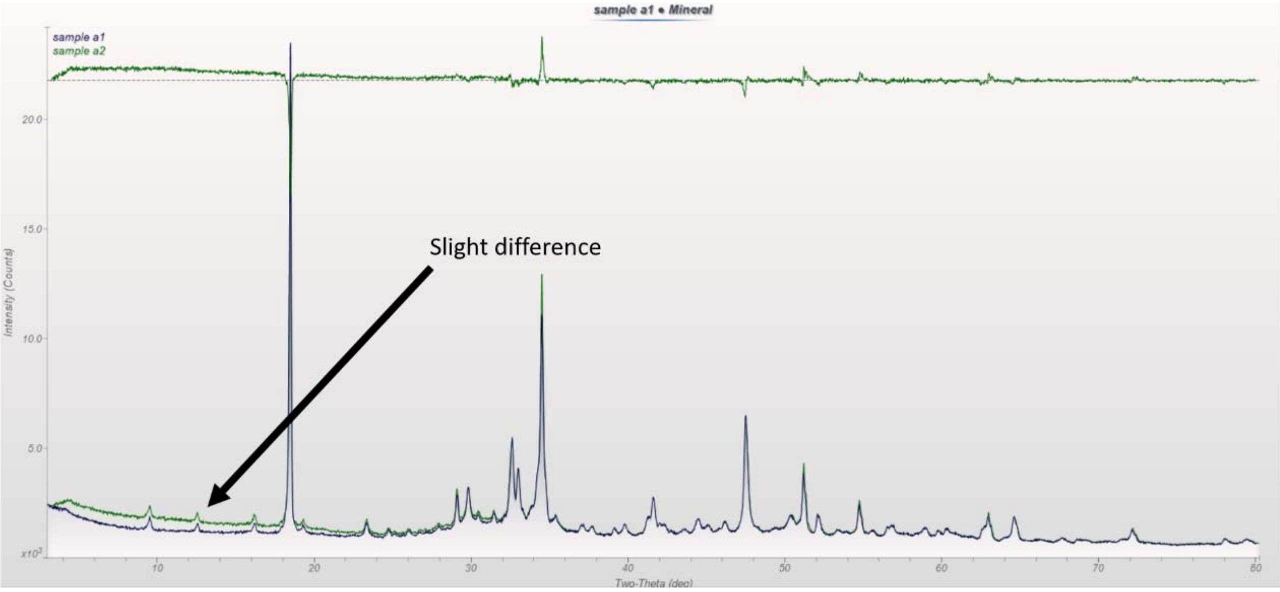
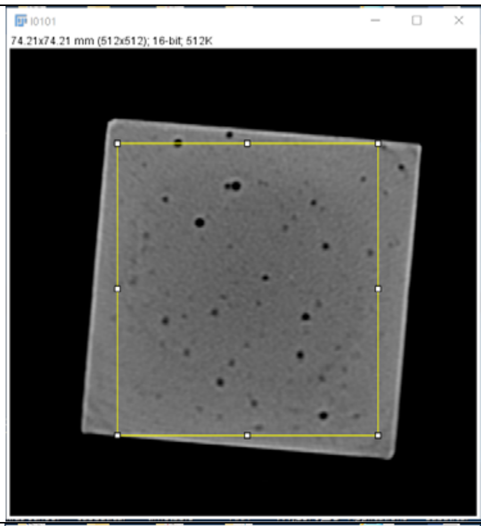
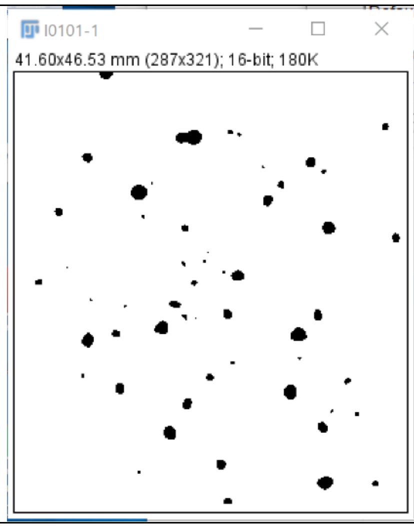
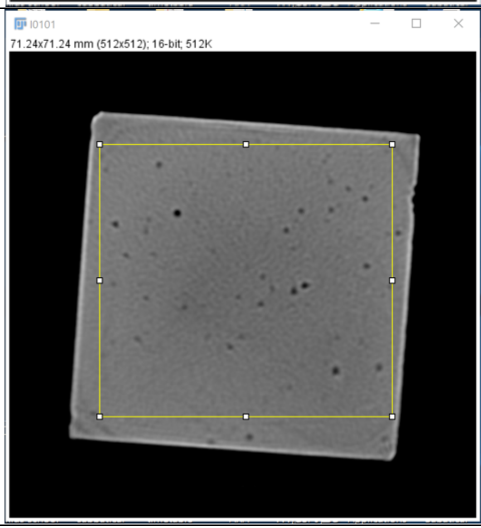
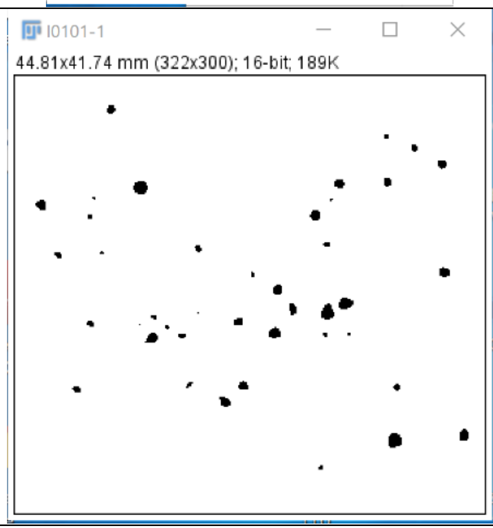


Fig. 11 XRF pattern comparison analysis of the cement powder

<table>
<tr><td>Cement sample B2</td><td>
</td><td>
</td></tr>
<tr><td>Cement sample B1</td><td>
</td><td>
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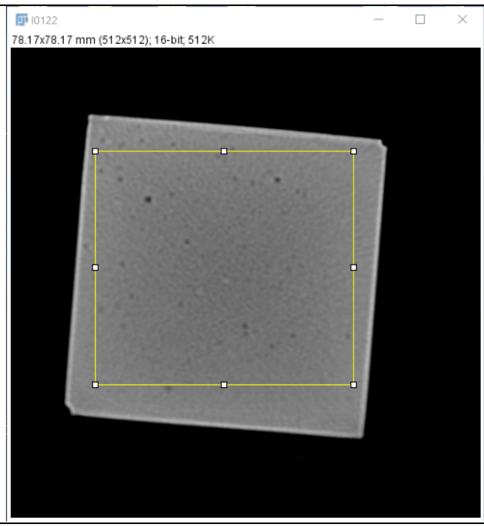
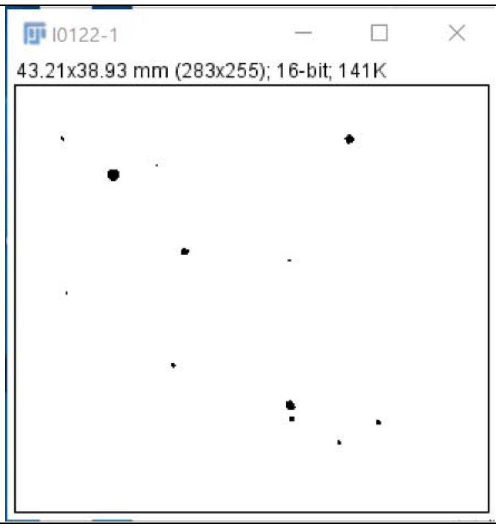
Cement sample B0		

Fig. 12 CT scan of selected images of cement cubes

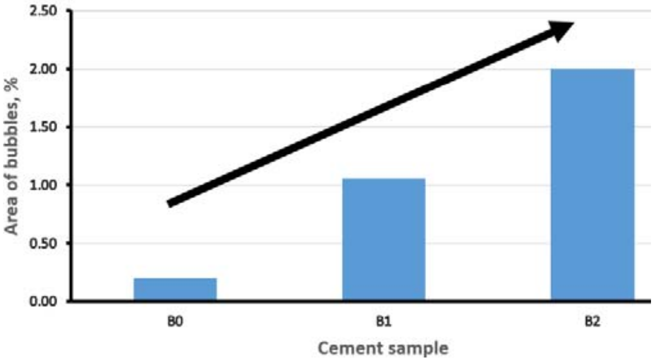


Fig. 13 Plot of area of Hydrogen bubbles occurred in cement cubes

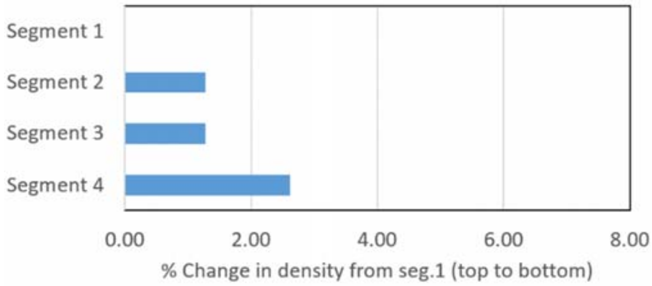


Fig. 14 The percentage density difference per segment for the normal cement sample

TABLE VII
RESULTS OF THE STABILITY TESTS

Cement Sample		Dry weight, g	Wet weight, g	SP gravity	Density, ppg	% difference from seg. 1
Neat cement sample	Segment 1	50.79	25.7	1.97	16.46	0.00
	Segment 2	45.23	22.6	2	16.67	1.28
	Segment 3	48.41	24.18	2	16.67	1.28
	Segment 4	41.41	20.42	2.02	16.89	2.61
Sample exposed to hydrogen	Segment 1	40.63	20.91	1.94	16.18	0.00
	Segment 2	49.94	25.2	1.98	16.5	1.98
	Segment 3	45.77	23.36	1.95	16.32	0.87
	Segment 4	52.98	25.61	2.06	17.23	6.49

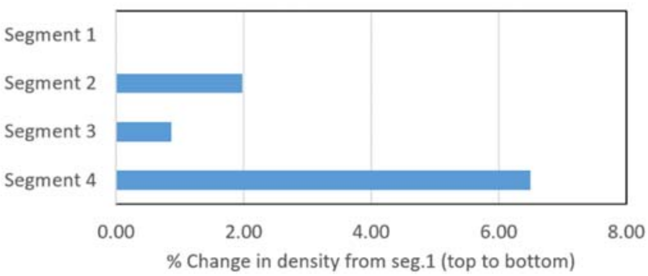


Fig. 15 The percentage density difference per segment for the cement sample exposed to hydrogen

IV. CONCLUSIONS AND RECOMMENDATIONS

The following conclusions were drawn from this research:

1. Exposing the cement slurry to hydrogen causes reduction in the compressive strength of the set cement sample.
2. Mixing hydrogen with cement slurry slightly increases its density and rheological properties and therefore having higher chance for cement losses.
3. During cementing a new well in a hydrogen storage reservoir, hydrogen bubbles will be trapped in the cement during setting period leading to reduction in cement strength and consequently, poor zonal isolation.
4. Exposing cement slurry to hydrogen makes the set (dry) cement slightly unstable.
5. Investigations of effects of different cement classes,

different densities of class “H”, curing temperature, and cement additives are recommended for future studies.

6. Further investigation into the effect and exposure of hydrogen on set cement is also advised.

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