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Using Geopolymers as a Sustainable Alternative Cementing Solution for Long-Term Zonal Isolation and Lost Circulation Challenges in Deep Geothermal Wells

Veerabhadra S Denduluri[1], Moneeb Genedy[1], Maria Juenger[1], Mike McDonald[2], and Eric van Oort[1]

[1]University of Texas at Austin [2]PQ Corporation

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ABSTRACT

Advancements in drilling technology currently allow geothermal wells to be constructed in very high-temperature conditions exceeding 600°F (315°C). Guaranteeing long-term zonal isolation and prevention of lost circulation events remain common challenges in these wells. Temperature-induced strength retrogression in Ordinary Portland Cement (OPC) slurries can be mitigated by the addition of silica, but little information is currently available regarding the very long-term stability of OPC under high-temperature conditions. Moreover, OPC is sensitive to acid gas attack (by H_2S and CO_2) and tends to de-bond from the casing, e.g., under induced cyclic pressure and temperature loads, due to low inherent casing-to-cement bond strength. Furthermore, the production of cement has a high carbon footprint, raising concerns about environmental sustainability.

These technical and environmental challenges could be solved by adopting alternative cementing materials known as geopolymers or alkali-activated materials (AAMs). Geopolymers have shown remarkable mechanical properties and thermal stability, with previous studies showing some materials stable up to a temperature of 800°C (1470°F). In addition, they exhibit very limited sensitivity to an acid gas attack and are made from waste materials with a low carbon footprint. The objective of the study reported here is to demonstrate the applicability of geopolymers as an alternative, sustainable cementing material in deep geothermal wells.

Geopolymer slurries made from fly ash or metakaolin and slag and activated with potassium-based alkaline solutions were designed for experimental investigation, which involved characterization of viscosity, pump time, and compressive/tensile/bond strength. These novel geopolymer slurries were found to be thermally stable at test temperature conditions up to 600°F. Furthermore, the geopolymer formulations exhibited exceptional casing-bond strength (up to an order of magnitude higher than OPC) with desirable compressive and tensile strength. They were also much less sensitive than OPC to contamination, making them superior candidates for use in cement squeezes to deal with severe lost circulation events. These advantages make geopolymers an effective cementing solution to overcome challenges such as long-term zonal isolation and lost circulation events in deep, high-temperature geothermal systems.

1. Introduction

With the increase in greenhouse gas level in the atmosphere, there has been a shift towards low carbon emission technologies, with geothermal energy receiving increased attention over the last decade (Lukawski et al. 2014). With advancements in drilling technologies, a large number of geothermal wells are being drilled in harsh temperature environments (i.e. up to – and exceeding - 600°F), in which drilling and well completion costs account for a major portion of the expenditure (Vivas et al. 2020). Geothermal well construction encounters challenges such as loss of casing & cement integrity, lost circulation events, failure to achieve long-term zonal isolation, etc., which together lead to an increase in nonproductive time (NPT) of a well (Finger and Blankenship 2012). Of all the challenges, lost circulation events are still the most problematic and largest cause of NPT, contributing to around 10 - 20% of geothermal well costs (Cole et al. 2017; Saleh et al. 2020). These challenges could be solved by designing a suitable cementing material that has high-temperature stability and helps in reducing geothermal well costs by mitigating lost circulation events and guaranteeing long-term zonal isolation.

Ordinary Portland Cement (OPC) is the conventional cementing material used in geothermal well construction. However, at elevated temperatures, OPC experiences several drawbacks such as strength retrogression, weak casing-cement bond, high shrinkage, contamination by non-aqueous drilling fluids, and degradation in CO_2-rich environments (Nasvi, Gamage, and Jay 2012; Nelson and Guillot 2006; Panchmatia et al. 2020). Also, the OPC manufacturing process generates high CO_2 emissions, which contribute up to 7% of all global, man-made CO_2 emissions. Alternative cementing materials e.g., calcium-aluminate-phosphate cement (CaP), which tend to be CO_2 resistant and survive hostile geothermal conditions, have been investigated for geothermal applications (Pyatina and Sugama 2018).

Geopolymers, also known as alkali-activated materials, have been evaluated for utilization in concrete and oil & gas industries and found to be a suitable cementing material alternative to OPC (van Oort et al. 2019; M. Juenger et al. 2011). Geopolymers are made from industrial by-products (e.g., fly ash or slag) or natural alumina-silicate rich materials (e.g., metakaolin or natural pozzolan) activated with different alkali activators such as potassium/sodium silicates and hydroxides). The raw materials used in making geopolymers reduce carbon emissions by 80% fewer compared to OPC (Nawaz, Heitor, and Sivakumar 2020). Additionally, geopolymers have high resistance to contamination by non-aqueous drilling fluids (Liu et al. 2019), exhibit self-healing abilities when damaged (Panchmatia et al. 2020), and show lower shrinkage than OPC (Liu 2017; Olvera et al. 2019). Class F fly ash-based geopolymers have been used for oil well cementing and were found to have considerable advantages over OPC (Salehi et al. 2019). However, with coal-fired energy plants phasing out, there will be a deficit in the future supply of fly ash (M. Juenger, Snellings, and Bernal 2019) and exploring alternative binders (metakaolin, slag, natural pozzolan, etc.) for oil & gas or geothermal cementing operations is necessary. Metakaolin and slag-based geopolymers have been studied by various researchers and have been used in mud cake solidification methods (Bu et al. 2020) and deepwater oil well-cementing purposes (Jiapei et al. 2018). Moreover, metakaolin and slag-based geopolymers have shown excellent thermal stability at temperatures as high as 800°C (Bernal et al. 2011).

This paper demonstrates the applicability of fly ash and metakaolin/slag based geopolymers for deep geothermal well cementing. The mechanical properties (compressive, tensile, and bond strength), viscosity, and thickening time tests were evaluated in the laboratory using these geopolymer formulations.

2. Materials and Experimental Methods

2.1 Cementitious Materials

Three types of cementitious materials/aluminosilicate sources were used in this study to formulate geopolymer slurries: Class F Fly Ash (FA), Metakaolin (MK), and Ground Granulated Blast Furnace Slag (GGBFS). Table 1 shows the oxide compositions (by the percentage of mass) of the cementitious materials.

Table 1: Chemical composition of geopolymer raw materials used in this study.

Material	**Content (%)**							
	Al_2O_3	**SiO_2**	**Fe_2O_3**	**CaO**	**MgO**	**SO_3**	**Na_2O**	**K_2O**
FA	23.2	44.7	24.2	3.2	0.8	0.6	1.0	0.0
MK	36.0	54.6	2.9	0.4	0.3	0.0	0.0	1.4
GGBFS	10.6	35.2	1.5	39.0	10.7	2.6	0.3	0.5

2.2 Activators and geopolymer preparation

Three different commercially available potassium-based activators were used in this study: Liquid Potassium Hydroxide (LPH), Liquid Potassium Silicate (LPS), and Solid Potassium Silicate (SPS). Activating solutions were prepared by maintaining the concentration of alkali (K) at 8 M for both FA and MK/GGBFS based geopolymers. Table 2 shows FA based geopolymer mix parameters where W/S indicates water to solid ratio and SiO_2/K_2O is silicon dioxide to potassium oxide weight ratio. MK/GGBFS mix parameters are presented in Table 3, where SiO_2/Al_2O_3 is the molar ratio of silicon dioxide to aluminum oxide and K_2O/Al_2O_3 indicates the molar ratio of potassium oxide to aluminum oxide.

Table 2: FA based geopolymer mix parameters.

Activator	W/S	SiO_2/K_2O	K_2O by weight of FA
LPS	0.33	0.12	0.20
SPS	0.33	0.12	0.20
LPH	0.33	----	----

Table 3: MK/GGBFS based geopolymer mix parameters.

MK/GGBFS	Activator	W/S	SiO_2/Al_2O_3	K_2O/Al_2O_3
90% MK, 10% GGBFS	LPS	0.45	3.40	0.85
90% MK, 10% GGBFS	SPS	0.45	3.40	0.85
90% MK, 10% GGBFS	LPH	0.45	----	----

Potassium hydroxide pellets of 98% purity were added to deionized water (resistivity of 18.2 million ohm-cm) and the resulting alkali solution was allowed to cool to room temperature before mixing with the silicate activator and cementitious material. The final geopolymer mixing was carried out using the paddle stirrer operated at 600 rpm for 45 seconds, to obtain a uniform blend. In the case of geopolymer mix with SPS, SPS was dry blended with solid aluminosilicate precursor (FA or MK/GGBFS) before adding to the liquid alkali solution. On the other hand, LPS is added to the liquid alkali solution.

2.3 Mechanical Properties

2.3.1 Curing conditions

All the geopolymer formulations were cured in a High-Pressure High-Temperature (HPHT) curing chamber simulating the geothermal well conditions (bottom hole static temperature of 600°F and bottom hole pressure of 3000 psi were selected for this study) following the API recommended practice (described in API RP 10B-2, 2010). The formulations were cured for 3 days, and then the samples were tested to estimate the mechanical properties. The ramping profiles of pressure and temperature were as follows:

a) Pressure was increased from atmospheric pressure (14.96 psi) to bottom hole pressure (3000 psi) in 30 seconds and was maintained constant throughout the curing period of three days.
b) Temperature was increased from surface temperature (73.4°F) to bottom hole circulating temperature (450°F) in three hours and then to bottom hole static temperature (600°F) in 10 hours, which was maintained constant throughout the curing period of three days.

c) At the end of the curing period, pressure and temperature were ramped down in 30 minutes to reach atmospheric pressure and surface temperature.

2.3.2 Compressive Strength

Three cylinders (4 in length and 2 in diameter) of geopolymer slurries were cured for three days at 3000 psi and 600°F as described in section 2.3.1. A universal testing machine was used with a ramp rate in compression of 48 psi/sec to failure. The recorded peak load was used to calculate the compressive strength of the samples. Compressive strength tests were conducted following ASTM recommended practice (ASTM C39/C39M 2018).

2.3.3 Tensile Strength

Three cylinders (4 in length and 2 in diameter) were filled with geopolymer slurries and cured for three days as described in section 2.3.1. The tensile strength test (Brazilian split tension test) was conducted with a ramp rate of 2.75 psi/sec, adapted from ASTM recommended practice (ASTM C496/C496M 2011).

2.3.4 Bond Strength

The bond strength of geopolymer slurries was measured using a push-out test. A stainless-steel rod (0.5 in diameter) was placed in the center of a 2 in diameter cylindrical mold. Three geopolymer slurries of each mix were prepared, transferred to the molds, and then cured for three days at 3000 psi and 600°F. Then, a compressive load was applied with a ramp rate of 2.5 psi/sec to the steel rod until it was pushed out of the mold, with the peak load used to determine the bond strength. Also, to emulate the effect of surface contamination on the bond strength of geopolymer with steel casing, the steel rods were coated with a thin layer of synthetic-based mud (SBM). Geopolymer slurries were then cast around the SBM coated steel rods and clean steel rods before placing them into the curing chamber.

2.4 Viscosity

Viscosity tests were conducted using a Couette coaxial cylinder rotational oilfield viscometer at surface temperature (73.4°F). The rheological constants such as yield stress, consistency index, and fluid behavior index for each geopolymer formulation were calculated using the Herschel-Bulkley (H-B) model (also referred to as the yield power-law model). The model relates shear stress (τ) and shear rate ($\dot{\gamma}$) as follows:

$$\tau = \tau_y + K\dot{\gamma}^n \quad (1)$$

where τ_y is yield stress, K is consistency index, n is fluid behavior index

- when n = 1, H-B model becomes the Bingham plastic model
- when τ_y = 0, H-B model becomes the Power law model

2.5 Thickening / Set Time

An HPHT consistometer was used to measure the thickening time of geopolymer formulations following API RP 10A (2019). The test pressure was increased to 1000 psi immediately at the start of the test and to 3000 psi in three hours, which was then kept constant until the end of the test. The test temperature was increased from the surface temperature (73.4°F) to the bottom hole circulating temperature (450°F) in three hours and was then kept constant until the end of the test. Each slurry was tested three times.

Thickening / Set time is the duration where geopolymer slurry is in a liquid state and can be pumpable. It is determined as the time required by slurry to reach a consistency of 70 Bearden consistency (Bc).

3. Experimental Results and Discussion

3.1 Mechanical Properties

Geothermal well cement is typically designed to have a compressive strength of at least 1000 psi, according to the API task group on cement for Geothermal Wells (Johnson et al. 2018). The compressive strengths of geopolymer slurries formulated in this study are shown in Figure 1. All the formulations have exceeded the minimum required compressive strength for geothermal wells. Moreover, FA-based geopolymer formulations activated with LPH and SPS had higher compressive strengths than OPC (1670 ± 160 psi as reported in the literature by Genedy et al., 2021). FA and MK/GGBFS geopolymer formulations activated with LPS had the lowest compressive strengths (1050 psi) among all the formulations, but still exceeded the minimum strength requirement.

Tensile strengths of the geopolymer slurries are shown in Figure 2, and these follow the same trend as compressive strength results. Tensile to Compressive strength ratio (T/C) of OPC is typically around 0.1. All the formulations used in this study had similar or high T/C than OPC as shown in Figure 3, which indicates ductile behavior and also helps in extending the life of cement sheath (De Paula et al. 2018). Due to thermal-induced loads and the presence of natural fractures in geothermal wells, crack propagation is a common challenge experienced during operations (Kamali and Ghassemi 2018). The geopolymer which exhibits ductility can prevent the occurrence of crack propagation and can enhance the cement barrier integrity.

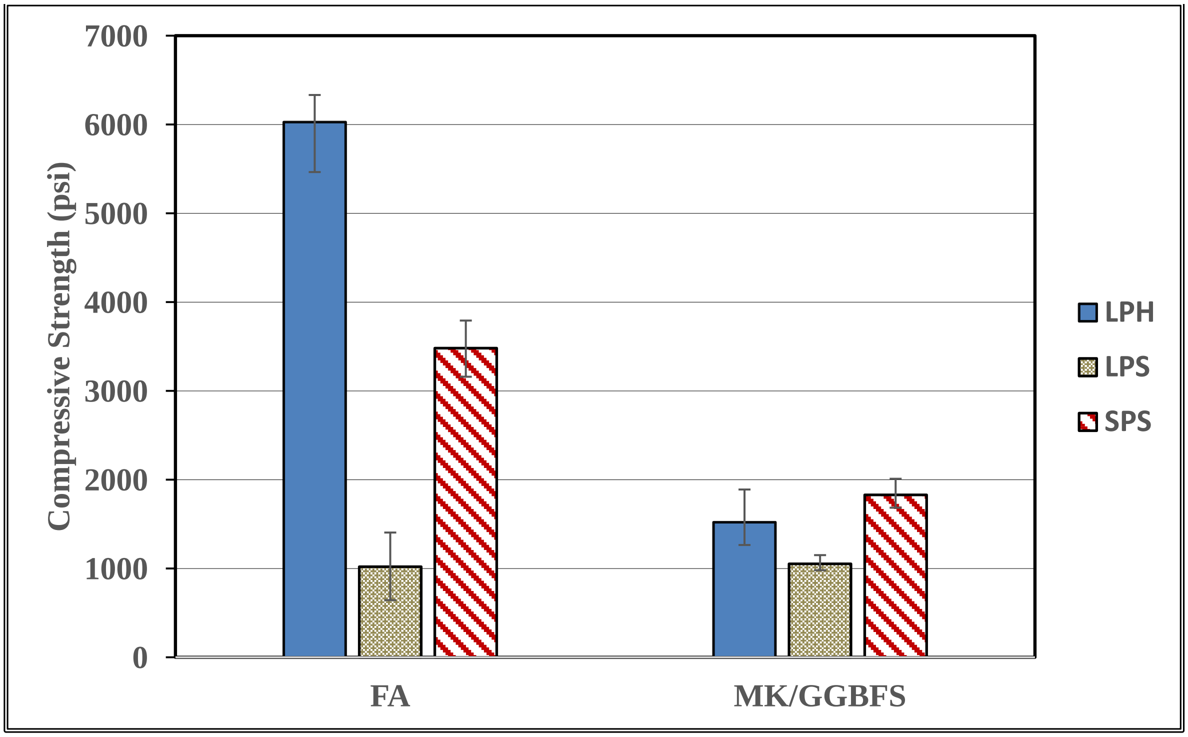

Figure 1: Compressive strength results of FA and MK/GGBFS based geopolymer slurries subjected to geothermal well curing conditions for 3 days. Error bars shows the range of compressive strength values for three replicates.

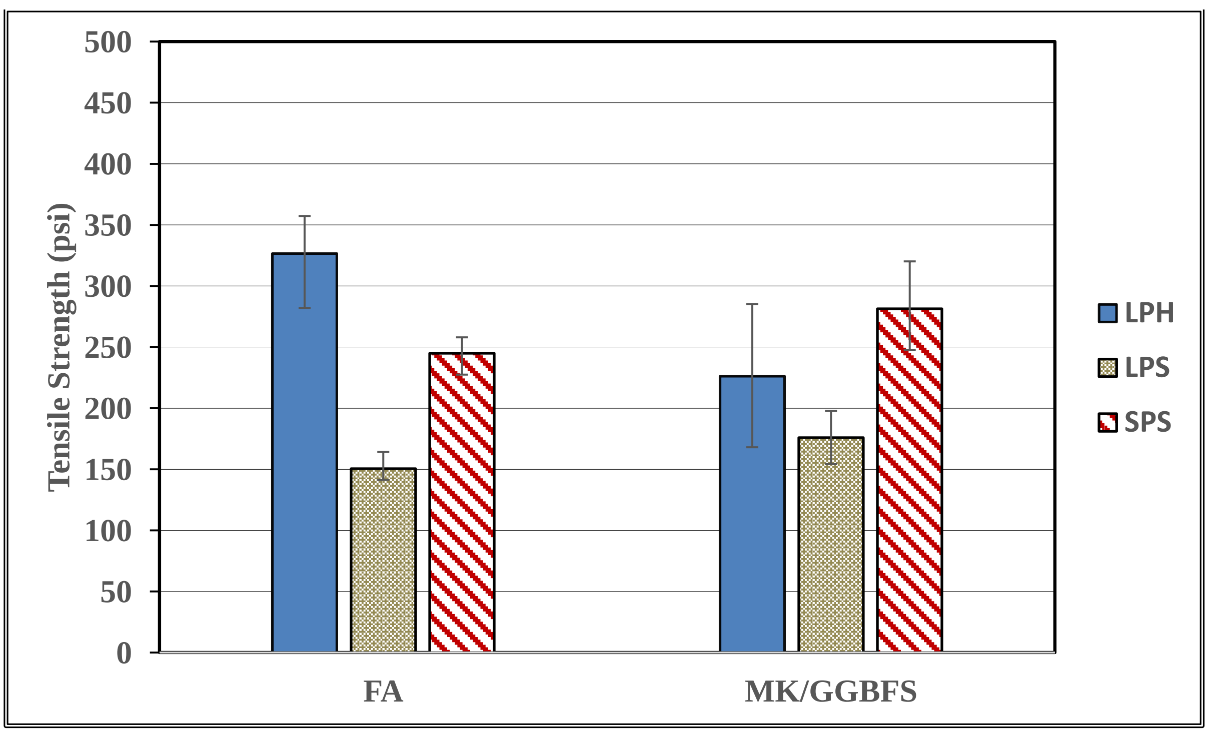

Figure 2: Tensile strength results of FA and MK/GGBFS based geopolymer slurries subjected to geothermal well curing conditions for 3 days. Error bars shows the range of tensile strength for three replicates.

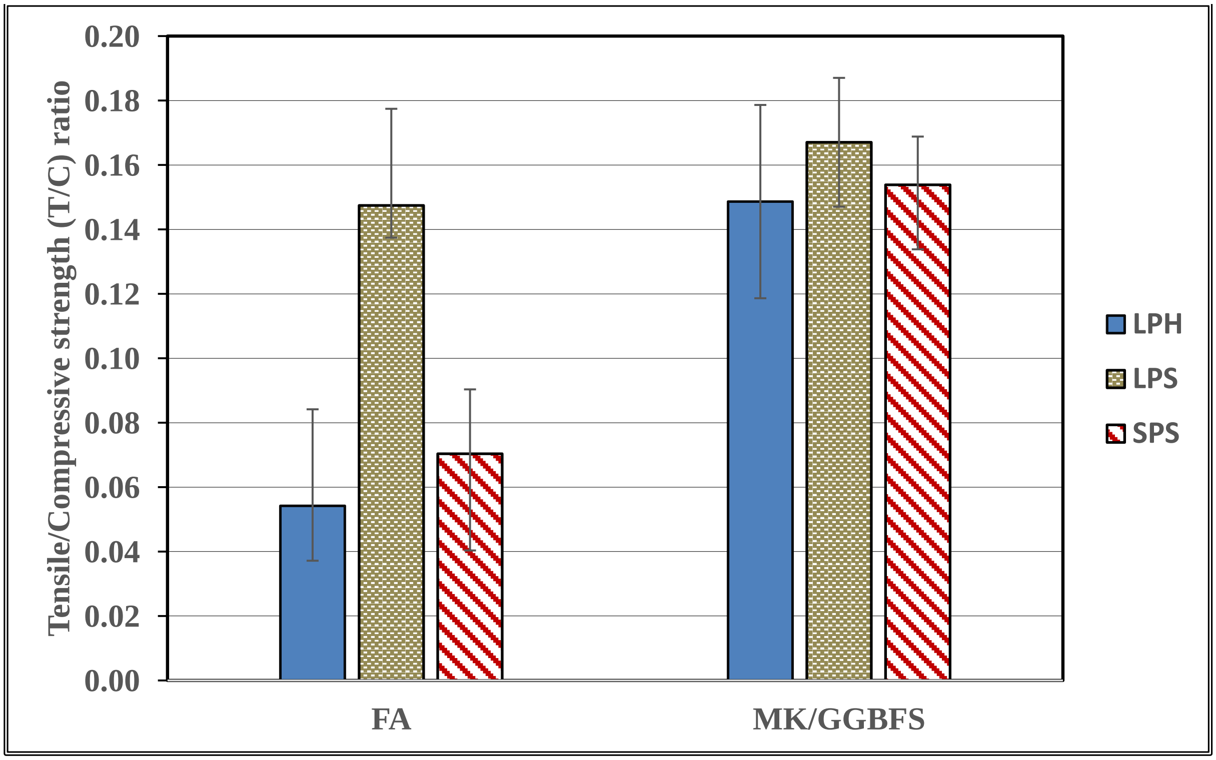

Figure 3: Tensile to compressive strength ratios of geopolymer slurries.

The bond strength results for FA and MK/GBFS geopolymers are presented in Figures 4 and 5, respectively. Bond strength tests were carried out with clean steel rods and SBM contaminated steel rods, respectively. Bond strength values (with clean surfaces) for all the geopolymer formulations were found to be a factor 3.0 – 10.0 higher than OPC (bond strength for OPC is 70 ± 20 psi as reported in the literature by Genedy et al. 2021). Furthermore, OPC had almost zero bond strength in the situation where the casing is contaminated with drilling mud (Genedy et al. 2021). On the other hand, geopolymer formulations in this study were found to have adequate bond strength when the surface was contaminated with SBM (50 to 200 psi). This remarkable difference suggests that the geopolymers are more suitable than OPC for cementing operations in deep geothermal wells, to provide long-term zonal isolation.

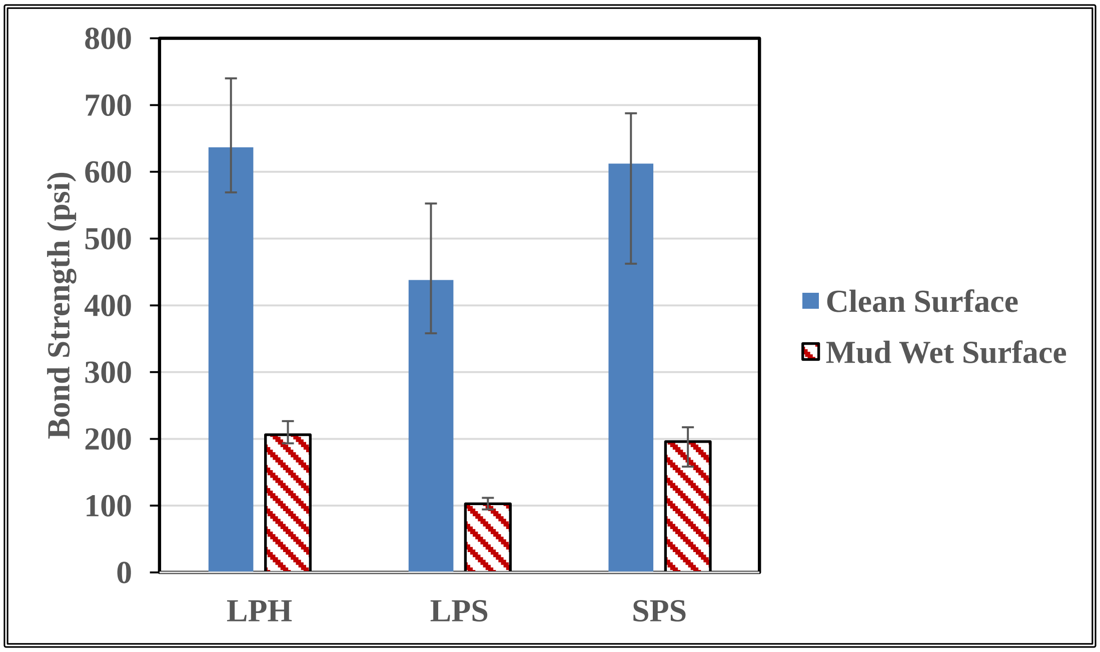

Figure 4: Bond strength results (with clean and mud wet surfaces) of FA based geopolymer slurries subjected to geothermal well curing conditions for 3 days. Error bars shows the range of bond strengths for three replicates.

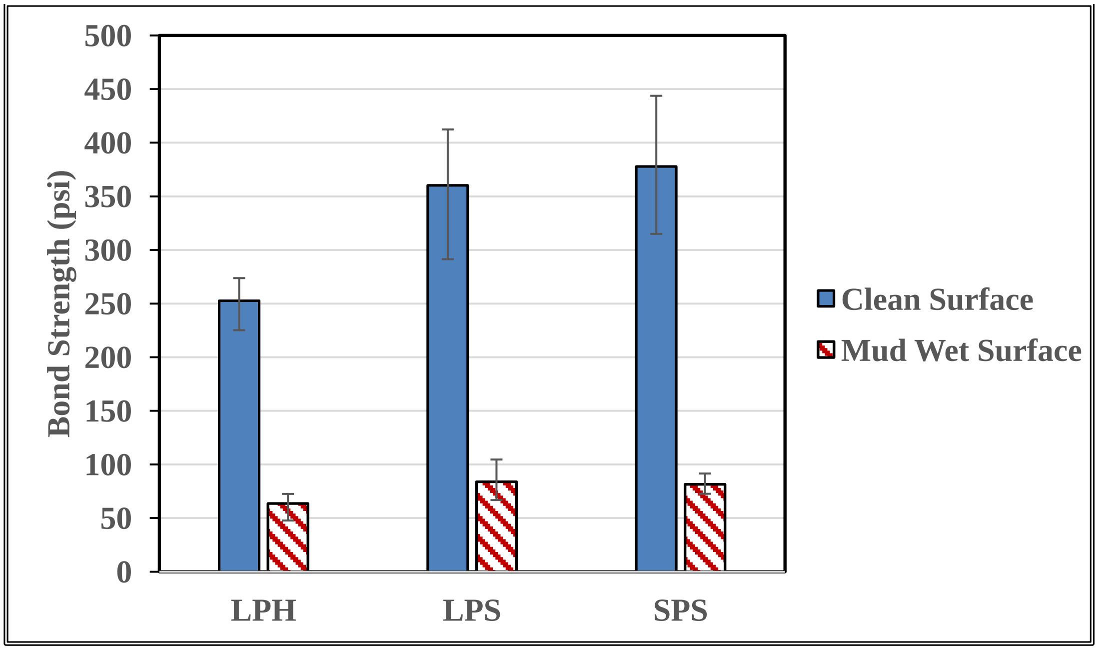

Figure 5: Bond strength results (with clean and mud wet surfaces) of MK/GGBFS based geopolymer slurries subjected to geothermal well curing conditions for 3 days. Error bars shows the range of bond strength for three replicates.

3.2 Viscosity

The rheological model constants obtained from testing each geopolymer formulation are shown in Table 4, with the shear rate vs shear stress plots presented in Figure 6. All geopolymer formulations have a flow index (n) close to 1 and hence can be considered as Bingham plastic fluids (0.95 < n < 1.05). MK/GGBFS-based geopolymer activated with SPS was found to be very viscous, as the apparent viscosity could not be measured by the viscometer (> 0.32 cp). Other geopolymer slurries have more desirable rheological properties and are considered pumpable.

Table 4: Rheological model constants for geopolymer formulations.

Cementitious material		FA			MK/GGBFS		
Activator		LPH	LPS	SPS	LPH	LPS	SPS
Apparent viscosity @ 300 rpm (Pa-s)		0.20 ± 0.07	0.21 ± 0.08	0.30 ± 0.03	0.16 ± 0.05	0.26 ± 0.04	> 0.32
Herschel-Bulkley rheological model constants	***Yield stress (Pa)***	2.62 ± 0.98	0.57 ± 0.93	3.20 ± 1.05	3.48 ± 1.11	2.79 ± 0.96	4.95 ± 0.94
	Fluid behavior index (n)	1.02 ± 0.08	1.01 ± 0.10	1.04 ± 0.04	1.02 ± 0.04	1.03 ± 0.06	0.95 ± 0.12
	Consistency index (K)	0.18 ± 0.09	0.19 ± 0.05	0.22 ± 0.04	0.14 ± 0.06	0.17 ± 0.03	3.58 ± 0.05

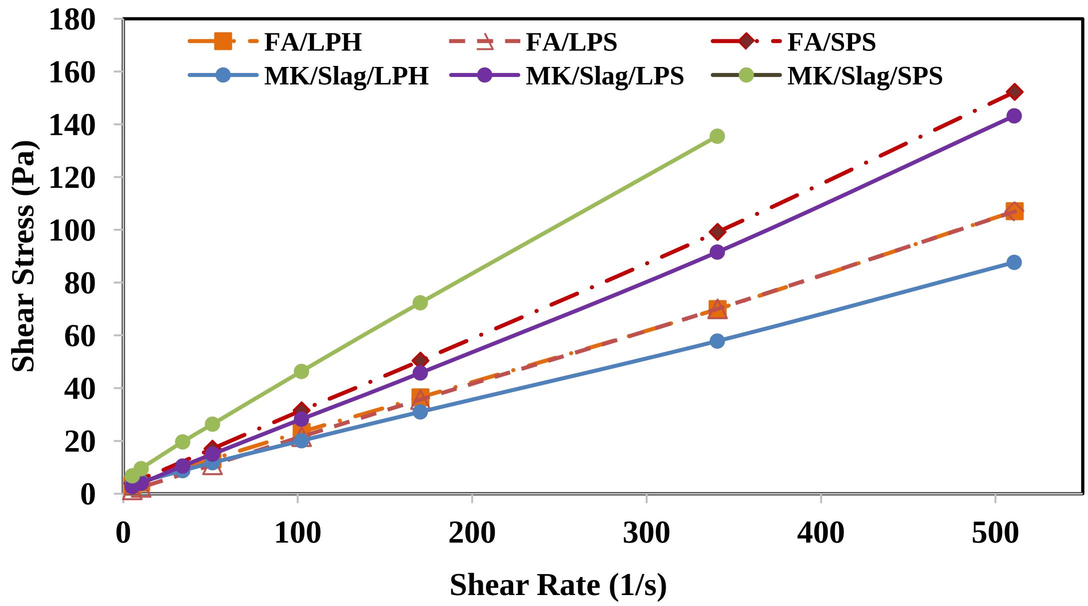

Figure 6: Rheological behavior of geopolymer formulations.

3.3 Thickening / Set Time

Thickening time results of FA and MK/GGBFS based geopolymers under geothermal well conditions are summarized and shown in Table 5. These tests were conducted using an HPHT consistometer and thickening time was interpreted as the time required by the geopolymer slurry to reach a consistency value of 70 Bc. It was found that the results obtained with all the activators can still be improved. The conventional retarders (sodium & calcium lignosulfonates, borax, zinc oxide, etc.) were used in this study to extend thickening time and were found to be an unsuitable option in the geothermal well conditions, due to the high alkalinity of geopolymers. Additional work is ongoing to improve the geopolymer mix design and obtain a higher thickening time.

Table 5: Thickening time test results.

Geopolymer Mix	Activator	Thickening Time (Minutes)
FA	LPH	90 ± 14
	LPS	70 ± 18
	SPS	85 ± 11
MK/GGBFS	LPH	92 ± 10
	LPS	96 ± 21
	SPS	87 ± 08

4. Summary and Conclusions

This paper reviews the feasibility of using geopolymers made from fly ash and metakaolin/slag as a sustainable and alternative cementing material in deep geothermal wells for primary cementation and remedial cementation (such as squeeze cementing and lost circulation mitigation). It is shown that the geopolymers exhibit excellent thermal stability at 600°F confirming the findings by other sources in the literature. Hence geopolymers can be used as a potential cementing material for high-temperature geothermal wells.

This paper also illustrates that the geopolymer formulations have desirable rheological properties with apparent viscosities ranging from 0.2 to 0.3 Pa-s (except MK/GGFS geopolymer activated with SPS, which has apparent viscosity of > 0.32 Pa-s) and are pumpable. It is also shown that all the geopolymer formulations exhibit adequate compressive and tensile strength exceeding the minimum requirement. Moreover, the T/C ratio of geopolymers includes ductile behavior, which benefits geothermal wells in preventing crack propagation and thus enhancing the efficiency of the geothermal systems. The geopolymers were also found to have exceptional casing bond strength and good contamination resistance to synthetic-based mud, unlike OPC. With these excellent mechanical properties, high thermal stability, and desirable rheological properties, geopolymers can be a perfect choice for applications in geothermal well cementing to prevent severe lost circulation events and guarantee long-term zonal isolation.

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