Application of a Novel and Sustainable Silicate Solution as an Alternative to Sodium Silicate for Clay Swelling Inhibition

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ABSTRACT: Shale swelling during drilling operations causes many problems mainly related to wellbore instability. The oil-based muds (OBMs) are very effective in controlling the swelling potential of clay-rich shale formation, but their environmental concerns and the economic aspects curtail their usage. In the application of water-based mud (WBM), it is mixed with various swelling inhibitors such as inorganic salts (KCl and NaCl), sodium silicate, polymers, and amines of various types. The above-mentioned materials are however afflicted by some limitations in terms of their toxicity, their effect on drilling mud rheology, and their limited tolerance toward temperature and oil contamination. In this study, we investigated a novel hybrid aqueous alkali alumino silicate (AAAS) as a shale swelling inhibitor in WBM. The AAAS is a mixture of sodium, aluminum, and silicon oxides. Experimental investigations were carried out using a linear swell meter, hot rolling and capillary suction timer, ζ-potential test, filtration test, and rheology test. The application of hybrid silicate as a swelling inhibitor was studied in two phases. In the first phase, only silicate solutions were prepared in deionized water at various ratios (1, 2, and 5%) and tested on sodium bentonite and shale samples containing high contents of kaolinite clay. Further testing on commonly used inhibitors such as KCl and sodium silicate solutions was conducted for comparative purposes. In the second phase, different drilling mud formulations consisting of various percentages of AAAS were mixed and tested on original shale samples. It was observed that the novel silicate-based mix proved to be a strong shale swelling inhibitor. Its inhibition performance was better as compared to the sodium silicate solution and KCl solution. It not only inhibits shale swelling but also acts as a shale stabilizer due to its high adsorption on the shale surface, which prevents the shale/water reactivity, makes the shale formation stronger, and prevents caving.

1. INTRODUCTION

During drilling operations, different types of formations are encountered, and various of them is shale, which is difficult to handle operationally. Shale swelling leads to many issues such as caving, wellbore instability, high torque, stuck pipe, and sometimes complete loss of a well if not handled properly.1 Shale swelling also affects the wellbore cleaning and drilling efficiency due to the accumulation of shale cuttings at the bottom of the well. Such well complications seriously affect the drilling and increase its nonproductive time, which contributes to the increased exploration and production costs. The drilling of sensitive shale rock with water-based mud (WBM) is extremely vulnerable to wellbore stability. Sensitive shale formations are more susceptible to wellbore instability problems due to hydration and swelling of clay in the wellbore formations. During the drilling operations, around 75% of drilling problems and 90% of wellbore instability issues are associated with shale rock.2 Therefore, the application of WBM without swelling inhibitors stipulates wellbore problems during the drilling operation. Oil-based mud (OBM) has superiority over WBM in terms of shale inhibition, lubricity, and stability at high temperatures.3 However, the application of OBMs has been restricted due to environmental concerns and extreme costs.4,5 Therefore, it is need of the hour to develop green shale swelling inhibitors for effective drilling mud properties so that the cost issues related to wellbore instability can be minimized.

Mostly electrolytes such as NaCl and KCl are applied as shale swelling inhibitors to curb shale swelling up to some extent.6−9 However, these electrolytes could adversely affect the properties of drilling mud, which leads to high fluid loss and flocculation of bentonite.10,11 High contents of KCl in drilling mud could be toxic to the marine environment, drilling environment, and disposal area.12 Further, KCl-based drilling mud causes bit balling by coagulating the cuttings around the...
bit. There are several organic and inorganic additives that can minimize the clay–fluid interactions and effectively reduce the hydration of the clay minerals. The main organic additives used as clay swelling inhibitors are cationic molecules containing quaternary ammonium groups in their structures. Organic polymers such as quaternary polyamines and quaternary polyacrylamides are also used as additives for stabilization and swelling inhibition of clay. In addition to cationic polymers, high-performance shale inhibitors have been developed such as surfactants, copolymers, ionic liquids, and modified nanoparticles. Recently, gemini cationic surfactants have been tested for potential use as shale swelling inhibitors. There are numerous limitations of the abovementioned swelling inhibitors, which include but not limited to temperature limitation, low swelling inhibition, and salt resistance.

Sodium and potassium silicate-based drilling muds are one of the WBMs that can match the shale inhibition properties of OBMs. The effectiveness of silicate-based systems for shale inhibition has been known since 1930. The shale inhibition properties of silicate-based muds have been well documented in the literature. The rise of silicate-based muds was driven by a combination of their cost-effectiveness, demand, and better environmental benefits. They have gained commercial acceptance and are widely used particularly for shale stabilization around the globe for onshore and offshore drilling operations. Over the decades, most of the drilling muds have been formulated with sodium silicate with different ratios of silica oxide to sodium oxide (SiO₂/Na₂O) in the silicates. During the drilling operation in unconventional shale formations containing high contents of organic compounds, the drilling muds with silicate solution should show robust endurance toward oil contamination. Further, silicates are applied in oil well cementing operations as extenders and accelerators. The cement of oil-bearing formation can impact the setting time as compared to sodium silicate. The AAAS acts as a blocking agent as well by crystallizing at low pH conditions. The AAAS crystallization reaction is not a simple precipitation reaction. The AAAS crystallization reaction is not affected by the presence of NaCl due to its tolerance toward salts. In addition, it has shown great tolerance for organic contaminants. The AAAS-based slurries have higher mechanical strength, greater resistance toward water, and high controllable setting time as compared to sodium silicate. The AAAS acts as a blocking agent as well by crystallizing at low pH conditions. To the best of the authors’ knowledge, the application of AAAS as a shale swelling inhibitor and shale stabilizer has not been reported in the literature. The swelling inhibition performance of AAAS was evaluated using linear swelling tests, hot rolling, capillary suction timer (CST), and ζ-potential. The effect of AAAS on rheological and filtration properties of drilling muds modified with concentrations of AAAS was investigated. Further, the inhibition performance of the AAAS was compared with commercially used clay stabilizers such as KCl and sodium silicate.

2. RESULTS AND DISCUSSION

2.1. Linear Swelling of Bentonite Clay. In this study, the swelling of sodium montmorillonite-based clay was tested using a linear swell tester. The clay wafers were exposed to different drilling muds prepared by mixing of various concentrations of AAAS. Their swelling percentages were measured and compared with those of KCl and sodium silicate.

2.1.1. Effect of AAAS on Bentonite Clay. In this study, the swelling inhibition performances of 2% solutions of Na-silicate and AAAS were tested and compared with that of the 3% KCl solution. All of the tested solution performed better than deionized water (DW). It was observed that the AAAS solution at 2% provided a strong inhibition performance compared to the Na-silicate solution mixed at the same concentration. The clay swellings of AAAS solution and Na-silicate solution were 61 and 91% after 24 h, respectively (Figure 1).

![Swelling Plot of Na-silicate- and AAAS-based Solutions](https://dx.doi.org/10.1021/acsomega.0c01777)

**Figure 1.** Swelling plot of Na-silicate- and AAAS-based solutions prepared at 2% concentration.

swelling of 2% AAAS was quite similar to that of the 3% KCl solution as both solutions provided similar inhibition performance. At the end of the swelling period of 24 h, a little variation in swelling was gained. It showed that AAAS provided strong inhibition capacity at lower concentrations as compared to the KCl solution. The inhibition mechanism of silicate is physiochemical. Silicates not only attach with negative ions of clays but also adsorb in the layers of clays by forming a thin layer upon reduction in pH. This thin layer prevents water invasion. Silicates show a double inhibition mechanism. On the other hand, K ions replace the cations in the clay layers and attach to the negative ions in the layers. As AAAS showed promising results compared with KCl, it can be tested as an alternate solution of KCl in the field application.

2.1.2. Effect of KCl and AAAS Mixtures. The inhibition performance of AAAS was evaluated in a mixture with the commonly used inhibitor, KCl. The mixtures of 3% KCl and
two different percentages (1 and 2%) of AAAS solutions were tested for clay swelling.

Figure 2 shows the trend of clay swelling for 3% KCl and mixtures of KCl and AAAS solutions. Initially, the evolution of swelling was similar for all solutions for 8 h. Later, it was observed that the mixture of KCl and AAAS reduced the clay swelling as compared to the 3% KCl solution. After 24 h of exposure, the 3% KCl solution resulted in 59.76% swelling of clay. On the other hand, a mixture of 3% KCl and 1% AAAS swelled the clay by 52%. Further addition of AAAS by 2% brought down the swelling to 50%, which showed that a mixture of salt and high-concentration AAAS outperformed further inhibition capacity of the mixture.

Further, bentonite wafers were exposed to AAAS-based drilling mud formulations to study the inhibition capacity of AAAS in the presence of other additives. It was observed that AAAS showed a strong inhibition capacity, as shown in Figure 3. Upon increasing the concentration of AAAS, the inhibition performance of drilling mud improved. For example, 2% AAAS-based and 5% AAAS-based drilling muds resulted in 46.17% and 37.38% swellings after 24 h, respectively.

The least swelling was obtained in 5% AAAS-based drilling mud as it resulted in an almost 60% reduction in swelling from base drilling mud. In addition to strong inhibition, it was noticed that clay wafers became harder and dense and were difficult to break by hand force, as shown in Figure 4. The silicate layers coated the whole wafer and enhanced the strength of samples. Such kind of additional benefit assists in preventing the dispersion of shale cutting and reduces the caving problem during drilling operations. This thin layer formation happens due to a reduction in the pH of AAAS-based mud upon interaction with water contents during clay formation. The AAAS-based drilling muds had high pH. On the other hand, the water in the shale had low pH. Upon interaction with the clay water content, AAAS reacted with ions present in the layers of clay and formed a thin film on the surface of the clay. As a result, the swelling of clay reduced.

**2.2. Free Swelling of Bentonite.** The free swelling test is conducted on clay or shale wafers in which clay wafer is submerged in a swelling inhibitor fluid using a glass plate under ambient conditions. This test provides the swelling pattern of clay or shale sample. The free swelling was observed by taking photographs at different time intervals (5 min, 5 h, and 24 h). In this experiment, bentonite wafer was exposed to DW, 3% KCl, and AAAS-based solutions prepared in different concentrations (1, 2, and 5%) and digital photographs were taken at different time intervals (5 min, 5 h, and 24 h) as shown in Figure 5. Bentonite swelled upon interaction with...
water. After some time, when water penetrated the layers, it swelled them multiple times. Upon exposing bentonite wafers to the 3% KCl solution, the wafer was disintegrated and dispersed immediately. A similar trend was noticed in AAAS solutions. However, this dispersion reduced with an increase in the concentration of AAAS as the 5% solution showed strong integration as compared to other concentrations and the 3% KCl solution. It was concluded that the addition of AAAS reduced disintegration.

2.3. ζ-Potential Measurement. Figure 6 provides the ζ-potentials (ZPs) of base and AAAS-based drilling muds. Two different concentrations of AAAS (1 and 2%) were tested for ζ-potential measurement. The performance was compared with that of the Na-silicate-based drilling mud. ZP values are affected by the solids in the inhibition solution. The base drilling mud without silicates resulted in a ZP of −36.5 mV. Upon the addition of silicate to the base drilling mud, variations in ZP were noticed as it imbalanced the electric charge on the surface of bentonite. For 1% AAAS and 2% AAAS, the ZP reduced to −26.9 and −22.3 mV, respectively. The reduction in ZP showed that there was a reduction in repulsive forces between bentonite particles in the drilling mud that brought them closer and increased their particle size. This phenomenon showed the inhibition performance of AAAS. Similarly, Na-silicate resulted in a less negative ZP value, which could be attributed to its inhibition capacity.

2.4. Capillary Suction Time. The inhibition performance of AAAS silicate was evaluated by conducting a capillary suction time test, and the results are shown in Figure 7. The test was conducted on AAAS with two different percentages (1 and 2%). The performance was compared with that of the 2% Na-silicate drilling mud. It was noticed that the addition of AAAS in the base drilling mud reduced the capillary suction time. This reduction in time showed the inhibition performance of AAAS silicate as bentonite released the water instead of absorbing it. When 1% AAAS was admixed into the base mix, the capillary suction time reduced from 654.6 to 116.65 s. Further loading of AAAS leads to a slight increase in the capillary suction time. This increase in time could have happened due to the polymerization of AAAS in the drilling mud, which prevented water release. On the other hand, the addition of Na-silicate reduced the capillary suction time to 360.5 s, which was higher than that for AAAS-based drilling mud systems. From capillary suction time results, it can be depicted that AAAS silicate has a strong inhibition performance than Na-silicate.

2.5. Linear Swelling of Shales. In this part of the study, shale samples were exposed to AAAS-based drilling muds and shale swelling was measured for 24 h.

2.5.1. Effect of AAAS-Based Drilling Muds. The AAAS-based drilling muds were prepared and tested for inhibition performance. Three different concentrations of AAAS were investigated in this part of the study, as shown in Figure 8. The swelling test was conducted for 24 h. It was noticed that the addition of novel silicate reduced the swelling of shale. The minimum shale swelling was obtained on application of 5% AAAS-based drilling mud followed by 2 and 1% silicate-based drilling muds.

Further, it was noticed that a high concentration of silicate is adsorbed on the surface of the shale and reduced its interaction with water, as shown in Figure 9, where a thin layer of silicate accumulated on the surface of the shale pellet. This thin layer not only prevented water penetration into the layers of shale.
but also provided consolidation and integration to shale and prevented further dispersion of shale. This formation of a thin layer occurred due to a reduction in the pH of the silicate solution upon interaction with the shale water content. Further, it was observed that with low AAAS contents, a crystallization phenomenon occurs that also resulted in reducing the swelling of shale.

2.5.2. Effects of Salt and AAAS Solution on Shale. In this part, the impact of 5% AAAS solution was investigated as a swelling inhibitor and its performance was compared with that of 3% KCl, as shown in Figure 10. It was observed that AAAS at 5% concentration outperformed KCl. The shale swelled by 14% in the AAAS solution and 16.34% in KCl. It showed that AAAS showed superior inhibition capacity against commercially used clay stabilizers. Mostly, shale responds to the drilling fluid either by swelling or disintegration. It depends on the type of clay contents in the shale. The shale with high smectite contents swells, and if the kaolinite contents are higher, shale disintegrates and caves in the wellbore. It has been reported in the literature that the inhibition mechanism of silicate is physiochemical.\textsuperscript{36,37} Silicate plays an important role by inhibiting the shale expansion and preventing the disintegration of shale. When silicate interacts with shale or clay, a reduction in the pH of the silicate solution takes place, and as a result, silicate reacts with ions present in the layers of shale and forms a thin film on the surface of the shale. This thin film works as an osmotic layer and prevents water penetration into the layers of shale. In addition to that, silicate polymerizes and provides another barrier for further prevention. This polymerization phenomenon provides integration to the shale. It fills the fractures of shales and prevents their disintegration. Further, the alumina in novel silicate also plays an important role in shale stabilization. Usually, it also precipitates and gel inside the pores of shale and stops the hydraulic flow.\textsuperscript{25,38}

2.6. Hot Rolling Dispersion Test. The hot rolling test was conducted to study the disintegration of shale cuttings. Table 1 provides the results of a hot rolling dispersion test conducted on shale samples using water, AAAS-based solutions, and 3% KCl. In this test, it was investigated that the retention weight of shale cuttings increased with an increase in the concentration of AAAS silicate. The highest retention was obtained at 2% AAAS with a value of 98.48%. The reason for this high retention could be due to the coating of shale cuttings by the AAAS solution that inhibited the penetration of water and avoided the disintegration of cuttings. The least retention obtained with water was by the KCl solution. As previously observed in bentonite swelling, the addition of KCl disintegrated the clay. So, it could be the reason that shale retention reduced in this case as well. Further, it has been reported in the literature that the KCl solution has a prominent effect on kaolinite clay and disintegrates it upon reaction.\textsuperscript{25}

2.7. Rheological and Fluid Loss Properties. In this part of the study, drilling muds were formulated using various percentages of AAAS. The rheological properties such as plastic viscosity (PV), yield point (YP), and apparent viscosity (AV) were calculated using the Bingham plastic model and are shown in Figures 1–3. It has been observed that the addition of AAAS changed the rheological properties of drilling mud. The drilling mud plastic viscosity increased due to the addition of AAAS. For 1% AAAS, there was no change in PV as compared to the base drilling mud. As the percentage was increased to 2%, PV started gaining change with a 12.9% increment as compared to the base drilling mud. Upon further increase in the concentration to 5%, a drastic change that happened in PV as it increased from 6.08 to 15.46 cP, as shown in Figure 11.

The yield point was impacted by the addition of AAAS. It quite changed with various concentrations of AAAS, as shown in Figure 12. The 1% AAAS changed the YP from 6.1 to 6.3 lbf/100 ft\textsuperscript{2} but a further increase in the concentration to 2 and 5% enhanced the YP to 29.5 and 39.7 lbf/100 ft\textsuperscript{2}, respectively. At 5% AAAS, the drilling mud behaved as a gel.

The apparent viscosity was impacted by the incorporation of AAAS, as shown in Figure 13. It was noticed that AV increased with the concentration of AAAS. The highest AV was obtained for the drilling mud mixed with 5% AAAS. At high AV, it requires a high pump pressure to circulate the drilling mud out of the wellbore.

The carrying capacity property (YP/PV) was evaluated to define the effect of AAAS-based drilling muds on cutting suspension and removal capacity. The carrying capacity

![Figure 10. Shale swelling in the presence of 3% KCl and 5% AAAS solutions.](https://example.com/figure10)

Table 1. Hot Rolling Dispersion Test Results

| fluids | volume (vol %) | rolling period (h) | temperature (°C) | initial weight (g) | final weight (g) | recovery (%) |
|--------|---------------|--------------------|------------------|-------------------|-----------------|-------------|
| DW     | 100           | 16                 | 65               | 20.16             | 18.2            | 90.27       |
| AAAS   | 1             | 16                 | 65               | 20.10             | 19.2            | 95.52       |
| AAAS   | 2             | 16                 | 65               | 20.36             | 20.05           | 98.48       |
| KCl    | 3 wt          | 16                 | 65               | 20.12             | 19.1            | 94.93       |

![Figure 11. Plastic viscosity variation for AAAS-based drilling muds.](https://example.com/figure11)
property (YP/PV) is shown in Figure 14. Commonly, any value of YP/PV ≥ 0.75 displays good carrying capacity behavior of the drilling mud,39,40 consequently delivering an improved wellbore cleaning performance. All AAAS-based drilling muds including base drilling mud showed good wellbore cleaning and cutting suspension ability at all concentrations. Nevertheless, a high YP/PV will surge the annular frictional pressure loss, consequently increasing the equivalent circulating density (ECD) in the wellbore, which may break the formation rock if exceeded beyond the fracture point of a rock. It has been established in the literature that to ensure the utmost wellbore cleaning while circumventing undue ECD, YP/PV values should be in the range of 0.75–1.00 (lbf/(100 ft² cP)).39,40 These results further strengthen our previous observations that the base drilling mud and 1% AAAS showed better performance, whereas 2% AAAS and 5% AAAS showed good cutting carrying capacity but could result in high ECD problems. The YP/PV ratio can be customized by adding any friction reducer or a dispersant in highly concentrated AAAS-based drilling.

In the filtration experiment, the fluid loss controlling and wall building properties of AAAS-based drilling muds were measured. In this test, only two different concentrations (1 and 2%) of AAAS-based drilling muds were tested. Figure 15 provides the results of fluid loss with time. It was observed that fluid loss was impacted by the addition of AAAS. It was increased from the base drilling mud without AAAS. The base drilling mud provided 12 mL of fluid loss at the end of 30 min. On the other hand, 1% AAAS increased the fluid loss to 22.8 mL at the end of 30 min. The increase in fluid loss could have happened due to the inhibition of bentonite. The AAAS adsorbed on the surface of bentonite and prevented the penetration of water into its layers. So, water was released by the layers instead of absorbing it. Once the concentration was increased to 2%, the fluid loss was reduced. The reduction in the fluid loss at 2% happened due to gelling and polymerization that leads to a reduction in the fluid loss. The YP/PV ratio supported this argument of polymerization and improved the carrying capacity at 2% of AAAS. The increase in fluid loss happened in even KCl-based drilling muds as reported by Murtaza et al.13 The high fluid loss demands a fluid loss controller to be added in the drilling mud prepared with AAAS silicate to prevent the fluid loss.

### 3. CONCLUSIONS

In this study, a novel silicate solution was introduced as a clay swelling inhibitor and shale stabilizer. The performance of the AAAS solution was evaluated using rheology, fluid loss, linear swelling test, free swelling, hot rolling, capillary suction time, and ζ-potential tests. Further, novel silicate performance was compared with those of the commonly used KCl and sodium silicate.

Following conclusions can be drawn:

- The clay and shale swellings were inhibited with an increase in the concentration of AAAS.
- All AAAS-based solutions showed much better inhibition performance as compared to DW and sodium silicate solutions. The equivalent inhibition performance of AAAS with KCl was achieved at low concentrations.
- Free swelling results indicated that novel silicate inhibited the swelling and reduced the disintegration and dispersion of bentonite as compared to KCl.
Table 2. Composition of Na-Bentonite

| quartz (%) | cristobalite (%) | K-feldspar (%) | montmorillonite (%) | muscovite (%) | illite (%) | gypsum (%) |
|------------|------------------|----------------|---------------------|---------------|------------|------------|
| 1          | 15               | 9              | 54                  | 7             | 12         | 2          |

**Table 3. Composition of Shale**

| quartz (%) | kaolinite (%) | feldspar (%) | muscovite (%) |
|------------|---------------|--------------|---------------|
| 34.9       | 21.2          | 21.2         | 22.8          |

- AAAS silicate reduced the ζ-potential and made it less negative, which depicted its high inhibition performance.
- The capillary suction time was reduced upon the addition of AAAS, which showed the inhibition capacity of AAAS.
- The AAAS solution acted as a shale stabilizer due to its high adsorption on the shale surface, which prevented the shale/water reactivity. The AAAS enhanced the shale stability as the shale wafer became strong and hard enough that could not be broken with hand force.
- Rheological properties were impacted in different proportions by the different concentrations of AAAS with the major impact that occurred at 5% AAAS concentration.
- The carrying capacity improved with the addition of AAAS.
- The fluid loss increased with the addition of AAAS. AAAS-based drilling mud at 1% concentration resulted in high fluid loss than the 2% solution.

4. MATERIALS AND EXPERIMENTAL PROGRAMS

4.1. Materials. The swelling inhibition capacity of AAAS silicate was evaluated by performing different swelling tests on bentonite and shale samples. Tables 2 and 3 provide the compositions of bentonite and shale samples, respectively. Bentonite is mainly composed of high content of montmorillonite, which has high swelling characteristics. The montmorillonite clay mineral consists of a layered structure with a negatively charged tetrahedral sheet of silica and octahedral sheets of alumina. The interlayer spacing of clay minerals usually contains cations such as Na+, K+, Ca+, and Mg+, which are present due to the isomorphic substitution of metal cations. The composition analysis of bentonite was conducted using an X-ray diffraction test (XRD). It was found that bentonite mostly composed of montmorillonite (54%) with other minerals such as cristobalite (15%), feldspar (9%), illite (12%), and gypsum (2%). The surface area and cation exchange capacity (CEC) of bentonite are 710 m²/g and 81 mequiv/100 g, respectively.

The shale sample was mainly composed of kaolinite clay (21.2%), quartz (34.9%), muscovite (22.8%), and feldspar (21.2%). The appearance of shale was from brown to red and composed of kaolinite clay. The surface area of kaolinite was the highest among other minerals present in the shale sample, as shown in Table 4. The kaolinite shale surface area and CEC are 35 m²/g and 17 mequiv/100 g, respectively. The shale was brittle as it mostly consisted of kaolinite, quartz, and feldspar. It disintegrates upon interaction with water rather than swelling. The kaolinite mineral is 1:1 layered consisting of one layer of the tetrahedral sheet and one layer of the octahedral sheet. Kaolinite has little to nonswelling capacity upon hydration, but it disperses and causes fine migration upon interaction with water or other drilling muds without inhibitors.

Sodium silicate is mainly composed of oxides of sodium and silica. AAAS silicate is composed of oxides of sodium, silica, and alumina and is supplied by PQ corporation. The sodium silicate was acquired from Sigma-Aldrich. The compositions of sodium silicate and novel green silicate (AAAS) are listed in Table 5.

The thermogravimetric analysis (TGA) of AAAS was conducted using HP3-DSC from Linseis. Figure 16 provides the thermal scan of AAAS from 30 to 300 °C. It was observed that AAAS lost 27.5% of its mass upon reaching 300 °C, excluding water loss of 12.5% upon reaching 100 °C as the AAAS solution had water content. The major mass loss happened between 30 and 150 °C temperatures. Further, it showed that liquid silicate was transformed into solid state after 150 °C. It can be deduced from TGA analysis that AAAS can be used in reservoirs with a temperature up to 150 °C, which can vary depending on pressure.

Further, a detailed investigation of the AAAS performance was investigated previously. AAAS can be crystallized by diluting it with deionized water. Table 6 indicates the time for crystallization at 45 °C. It is obvious from the results that the addition of water changes the crystallization time. At high water contents, AAAS is made to crystallize in a short time due to a reduction in the pH. Further, the solution becomes highly unstable as the shale wafer becomes strong and hard enough that could not be broken with hand force.

![Figure 16. Mass loss (%) conducted using HP3-DSC at a rate of 10°C/min from 30 to 300 °C for AAAS silicate.](https://dx.doi.org/10.1021/acsomega.0c01777)
viscous in the presence of high water contents, as shown in Figure 17, and the solution color is milky. As the water content reduces, the crystallization time increases. At the highest content of AAAS, no crystallization happens in the solution. The color is light milky at high AAAS contents. The crystallization helps in blocking the pores of the formation and prevents the further penetration of water into the shale layers.

Further, an investigation on viscosity change with water dilution was conducted at room conditions for two different solutions. The AAAS was diluted with water in two different ratios (2:3) and (1:2). The test was conducted using an atmospheric viscometer (model 900). The solution was stirred at 6 rotation per minute (RPM), and the viscosity change with time was observed. As the AAAS was diluted with high water contents, a steep increase in the viscosity reached in a short time as compared to the solution with less water contents (Figure 18). For solution mixed with 1:2 ratio reached the crystallization time at 15 min. On the other hand, a solution with 2:3 ratio reached the crystallization time at around 50 min. It shows that water dilution leads to more polymerization reaction and enhances the viscosity. This crystallization time changes with temperature as well. As the temperature increases, the time reduces, and as a result, crystallization occurs in a shorter time.

### 4.2. Experimental Program

Several experiments were conducted to study the inhibition performance of AAAS silicate. Table 7 describes the experimental program used to conduct these experiments. Deionized water (DW) was used for the preparation of all AAAS-based solutions and drilling muds. The AAAS solution was tested at different concentrations, and the optimum concentration was reported considering its impact on rheology, fluid loss, and swelling. The performance of AAAS was compared with those of 3% KCl and 2% Na-silicate.

Similarly, AAAS-based mud was prepared and tested for shale swelling. In AAAS-based muds, AAAS was added in various concentrations (1, 2, and 5%), and the compositions of drilling muds are listed in Table 8. In all tested drilling muds, the total volume of the mud was 350 mL.

The pH value of AAAS-based drilling muds was measured and are reported in Figure 19. The addition of AAAS increased the pH of the mud, and the maximum pH was obtained at 5% concentration. Caustic soda was not added in AAAS-based drilling muds as AAAS provided high pH to the muds.

#### 4.2.1. Linear Swell Test

The linear swell test was conducted to investigate the clay or shale swelling with time. The
bentonite and shale wafers were prepared using the compactor. The cell assembly was filled with 12 g of bentonite powder and loaded in the compactor. The bentonite powder was compressed for 30 min at 6000 psi pressure using a hand pump. At the end of 30 min, the pressure was released, and the wafer was removed from the cell. Later, the wafer was loaded in the cup assembly of a linear swell tester and 150 mL of testing fluid was poured into the cup assembly. The test was conducted for 24 h, and the fluid was stirred at 100 RPM throughout the testing period. The test was conducted at room temperature using the OFITE dynamic swell meter (model 150-80-1).

4.2.2. Hot Rolling Dispersion Test. The hot rolling dispersion test is applied to investigate the inhibition performance of the AAAS solution to prevent hydration and disintegration of shale cuttings. In this test, shale cuttings were ground and sieved on 6-mesh size screen. The weighed shale cuttings were immersed in an aging cell that contains the inhibition fluid and the lid was sealed tightly. The aging cell was placed in the hot rolling oven (model M1750) at 65 °C for 16 h. At the end of the aging period, the shale cuttings were removed and washed over with deionized water and sieved on a 12-mesh size screen. The retained cuttings on the 12-mesh screen were collected and dried in the oven at 105 °C for 3 h. Once the cuttings were dried, the final weight was measured, and the retention percentage was calculated from the initial weight. The sample with high retention percentage proves to be a good inhibitor. In this test, two different concentrations (1 and 2%) of AAAS silicate were tested. The water dispersion was taken as a reference. Further, the inhibition performance of AAAS was compared with that of the 3% KCl solution.

4.2.3. Capillary Suction Timer. A capillary suction timer (CST) is used to study the colloidal shale properties and to determine the concentration of electrolyte in fluids for borehole stabilization. In this study, a OFITE CST (model 294-50) was used to examine the inhibition characteristics of AAAS-based drilling muds. The CST calculates the filtrate time that travels among radially separated electrodes interacting with special filter paper. The aqueous sample is loaded in the cylinder, and the pressure of suction of filter paper under the tested sample takes out the filtrate. When filtrate approaches first pair of electrodes, the timer will start. As the liquid approaches the third electrode, the time stops and the alarm sounds. A liquid-crystal display (LCD) counter displays the CST reading up to tenths of a second. The procedure is fast and straightforward. The procedure involves placing a special filter paper under the suction pressure unit and pouring 5 mL of mud solution in the sample cylinder. The filtrate advanced outwardly in a principally elliptical pattern with the timer starting when the filtrate reached the first pair of electrodes. When the filtrate reached the third electrode, the timing ceased and a noticeable sound beeped. The reading showed on the CST meter indicates the time of tenths of a second.

4.2.4. ζ-Potential Test. ζ-potential is a way of measuring the surface charges of the particles. It is the best way to find out the stability of fluids as the stability is related to interactions between particles. In this test, ζ-potentials of inhibition drilling muds were measured using an instrument supplied by Dispersion Technology Instruments of model DT-1202. The inhibition drilling mud was prepared by mixing 4% bentonite in DW and hydrated it for 24 h over the stirrer. After 24 h, the AAAS was added and dispersed for another 24 h. Later, the ζ-potential test was conducted with enough hydration and dispersion of bentonite in inhibitor. The three sample readings were taken for each mud, and an average reading was reported in this study. Two different concentrations of AAAS (1 and 2%) were tested and compared with those of the base and Na-silicate-based muds.

4.2.5. Rheology Test. During drilling, the drilling mud is circulated from the bottom of the well to the surface to carry the drill cutting and perform other functions. The drilling mud circulation is mostly controlled by its rheology as it impacts the performance of the fluid both in dynamic and static conditions. It defines the workability and pumpability of a fluid. In this study, an atmospheric viscometer manufactured by GRACE (model M3600) was used in measuring the rheology of the drilling mud. Different rheological parameters such as the yield point (YP), plastic viscosity (PV), and apparent viscosity (AV) were measured by following procedures of the American Petroleum Institute Standard. The drilling mud sample was agitated at various shear rates starting from 3 to 600 RPM, and the shear stress was measured at each shear rate. The plastic viscosity, yield point, and apparent viscosity were calculated using the Bingham plastic model by applying eqs 1–3.

\[
P V (cP) = \phi_{3000cp} - \phi_{500cp}
\]  
\[
YP \left( \frac{lbf}{100 \ ft^2} \right) = \phi_{500cp} - PV (cP)
\]
where $\phi_{600RPM}$ is the dial reading at 600 RPM and $\phi_{300RPM}$ is the dial reading at 300 RPM.

4.2.6. Filtration Test. The filtration test was conducted to investigate the fluid loss controlling and wall building properties of drilling muds using an API filter press by FANN Series 300. In this test, the filtration cell was filled with 350 mL of drilling mud and loaded in the filtration instrument. A pressure of 100 psi was applied under room temperature conditions. The filtrate was collected for 30 min at different time intervals. At the end of 30 min, the pressure was released, and the drilling mud cake was collected.

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**ABBREVIATIONS USED**

| Abbreviation | Description |
|--------------|-------------|
| AAAS | aqueous alkali alumino silicate |
| WBM | water-based mud |
| OBM | oil-based mud |
| KCl | potassium chloride |
| NaCl | sodium chloride |
| Na | sodium |
| DW | deionized water |
| API | American Petroleum Institute |
| CST | capillary suction timer |
| TGA | thermogravimetric analysis |
| RPM | rotation per minute |
| PV | plastic viscosity |
| YP | yield point |
| AV | apparent viscosity |
| cP | centipoise |
| lbf | pound force |
| wt % | weight percent |
| vol % | volume percent |
| ECD | equivalent circulating density |
| ZP | $\zeta$-potential |

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