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Faisal Ur Rahman Awan
Edith Cowan University, f.awan@ecu.edu.au

Alireza Keshavarz
Edith Cowan University, a.keshavarz@ecu.edu.au

Hamed Akhondzadeh
Edith Cowan University, h.akhondzadeh@ecu.edu.au

Sarmad Al-Anssari
Edith Cowan University, s.alanssari@ecu.edu.au

Stefan Iglauer
Edith Cowan University, s.iglauer@ecu.edu.au

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A Novel Approach for Using Silica Nanoparticles in a Proppant Pack to Fixate Coal Fines

Faisal Ur Rahman Awan[A, B, D]
Alireza Keshavarz[A]
Hamed Akhonzadeh[A]
Sarmad Al-Anssari[A, C]
Stefan Iglauer[A]

[A] School of Engineering, Edith Cowan University, Joondalup, 6027, Western Australia, Australia
[B] Department of Petroleum and Gas Engineering, Dawood University of Engineering and Technology, Karachi, 74800, Sindh, Pakistan
[C] Department of Chemical Engineering, University of Baghdad, Baghdad 10071, Iraq
[D] Corresponding author. e-Mail: f.awan@ecu.edu.au

Abstract

Hydraulic fracturing operations in coal seam gas (CSG) reservoirs are highly prone to release coal fines. Coal fines inevitably cause mechanical pump failure and permeability damage as a result of their hydrophobicity, aggregation in the system, and pore-throat blockage. Thus, one approach to affix these coal fines at their source, and to retard generation, is to introduce a nanoparticle-treated-proppant pack. Thus, this research explores coal fines retention (known as adsorption) in a proppant pack using nanoparticles. In the study, the electrolytic environment, pH, flow rate, temperature, and pressure were kept constant, while the variables were concentration of silica nanoparticles (0 – 0.1 wt%) and coal fines concentration (0.1 – 1 wt%). The objective was to identify silica nano-formulations that fixate coal fine dispersions effectively. Subsequently, the coal suspensions flowed through a glass bead proppant pack treated with and without nanoparticles, and then analyzed via a particle counter. The quantitative results from particle counter analysis showed that the proppant pack with nanoparticle treatment strongly affects the fixation ability of coal fines. The proppant pack without nanoparticle treatment showed up to 30% adsorption and flowed through the proppant untreated, whilst proppant pack treated with nanoparticles showed up to 74% adsorption; hence, more exceptional affixation ability to the coal fines. Further, the results indicated that the zeta potential of silica nanoparticles at higher salinity becomes unstable, i.e., ~ -20 mV; this low value helps the proppant pack treated with nanoparticles to attach coal fines to it. The ability of nanoparticles to adsorb coal fines is due to its highly active surface, and high specific surface area.

Keywords
Coal fines, nanoparticles, nanofluid, fines fixation, CBM, Proppant pack

1. Introduction

The application of nanoparticles has proven to be beneficial in the upstream oil and gas industry (Yang *et al.* 2015). Several applications like water flood displacement (Yuan and Moghanloo 2018; Bahraminejad *et al.* 2019; Najimi *et al.* 2019), fines fixation (Huang *et al.* 2010b; Assef *et al.* 2014; Yuan 2017; Zheng *et al.* 2018), and enhanced oil recovery (Yousefvand and Jafari 2015; Bera and Belhaj 2016; Yuan and Wood 2018; Asl *et al.* 2020) have been studied and experimented upon in this capacity. The use of nanoparticles to control fines release and fix them at or near the source of their origin has been studied comprehensively in both sandstones (Kia *et al.* 1987; Mohan and Fogler 1997; Rozo *et al.* 2007; Zeinijahromi *et al.* 2012; Bin *et al.* 2016; Hasannejada *et al.* 2017) and carbonates (Qajar *et al.* 2013; Al-Anssari *et al.* 2016; Al-Anssari *et al.* 2017b; Al-Anssari *et al.* 2018). The behavior of Coal Bed Methane (CBM) reservoir fines with nanoparticles, which are considerably different from sandstone fines, has yet to be comprehensively explored.

Compared to conventional reservoirs, CBM reservoirs are relatively weaker and are likely to fail in all operating stages of well development, especially while drilled in horizontal or high-angle directed trajectories (Palmer *et al.* 2005). CBM reservoirs have critical conditions to release fine. A critical salt concentration (CSC) exists in porous media (Khilar and Fogler 1984; Blume *et al.* 2005) below which fines can detach from the rock matrix. However, no presence exists of the validated agreement as to CSC in fractured media. The theoretical explanation of fines detachment / attachment has been explained using the Derjaguin−Landau−Verwey−Overbeek (DLVO) theory and extended DLVO theory. The basic premise is that if the attaching force/torque is higher than the detaching (repulsive) force/torque, then the particle will dislodge from the bulk rock matrix/structure and vice versa if the detaching (repulsive) force/torque is higher than the attractive force/torque. (Lever and Dawe 1984; Sharma *et al.* 1992; Schembre and Kovscek 2005; Rosenbrand *et al.* 2015). The size of coal fines varies according to the production stage, i.e., the usual size of coal fines found during the water production stage is usually lower than the size of coal fines generated during the gas production stage. When salinity is decreased, mobile particles also decrease in the order of size. This means that larger particles are released when ionic strength is more, and when ionic strength is decreased the smaller particles are mobilized (Keshavarz *et al.* 2014). The grain size and concentration of coal fines differ during various production stages (Zhao *et al.* 2016). Generally, two size categories can be distinguished, as follows:

1. Small-sized coal fines are produced during post-completion hydraulic fracture back-flow and in the production stage.
2. Large-sized coal fines are produced during drilling and completion (Han *et al.* 2015).

Coal fines generation and migration cause a reduction in conductivity and fracture length, thereby damaging the dewatering process (Zou *et al.* 2014). The factors that affect the production of coal fines output are:

1. Engineering aspects (well type, drilling operations/technology, hydraulic fracturing, completion technology, and the CBM production system) (Zhao *et al.* 2016; Geng *et al.* 2017).
2. Geological factors (tectonic stress, rock characteristics, structural characteristics of the coal, coal strength) (Zhao *et al.* 2016).

Bituminous coal fines have been shown to cause a 35% decline in permeability when subjected to water flow (Guo *et al.* 2016) and a 24.4% decline in conductivity when subjected to only 2% of coal fines flow into the proppant pack (Zou *et al.* 2014). Thus, coal fines can block the proppant pack and cause a decline in fracture conductivity (Bai *et al.* 2017), resulting in reduced production and failure of production equipment (Marcinew and Hinkel 1990; Badalyan *et al.* 2016). Several authors have studied coal fines suspension thus far, but have largely performed experiments in DI or distilled water (Zou *et al.* 2014), while hydraulic fracturing fluid is usually saline (nearly 0.6 M) to be geo-chemically compatible with reservoir formations fluid (Guo *et al.* 2015; Patel *et al.* 2016; Shi *et al.* 2018). The zeta-potential of coal suspension in DI water has been reported as -43.34 mV for bituminous (Shi *et al.* 2018) and anthracite as -20.5 mV (Zou *et al.* 2014); however, pH has not been mentioned as a crucial factor in determining the dispersion stability via zeta-potential measurements and has not been studied accordingly.

Fixating coal fines at their source of origin may be done by:

a) using nanoparticles and modified nanoparticles that alter the surface chemistry of fines by interacting with nanoparticles (Huang *et al.* 2010a),

b) employing micro-proppants before introducing larger proppants (also known as graded proppant injection) so that the fine particles are inhibited from moving into the proppant pack at or near its place of origin (Kumar *et al.* 2012; Keshavarz *et al.* 2014; Keshavarz *et al.* 2015; Keshavarz *et al.* 2016),

c) adding chemicals to the fracturing fluid for agglomerating fragments of coal fines and fixing them at the source (Shi *et al.* 2018). Consequently, dispersing coal fines can be completed via two modes: physical and chemical.

i. Physically, researchers have developed models for straining particles in the pore throat, where the critical value of consensus among them is of one to six ratio, also known as a one-sixth rule. This rule notes that if the reservoirs formation fines diameter (d) is six times less than its gravel pack diameter (D) i.e. $\frac{d}{D} < \frac{1}{6}$, the fines will not strain (Elena Rodríguez 2007; Zou *et al.* 2014).

ii. Polymeric surfactant-based chemicals injected along with fracturing fluid to disperse and move up to the surface along with well fluids (Magill *et al.* 2010); (Pan *et al.* 2015).

Thus, fines migration is one of the most crucial phenomena for formation damage in CBM, where this challenge has not been addressed comprehensively. Limited studies are available to suggest that coal fines generation could be controlled at or near the source using metal oxides nanoparticles (Huang *et al.* 2010a; Patel *et al.* 2016). This paper will provide further insights into coal fines behavior when treated with silica nanoparticles in a proppant pack. The results show that coal fines can be adsorbed effectively using silica nanoparticles (NPs). An effective concentration of 0.1 wt% of silica NPs is recommended for achieving better adsorption of fines.

2. Methods and Materials

2.1. Materials

Coal lumps from coal mines in Morgantown, West Virginia, USA, were retrieved with a vitrinite reflectance of 0.91, indicative of highly volatile bituminous coal (Moore 2012). Furthermore, the composition of macerals and minerals was 97.4% and 2.6%, respectively. The properties of the coal sample are displayed in Table 1. The coal lump was crushed into smaller sizes by mortar and pestle method, where the coal fines used in this study were sieved using an electric sieve shaker. Subsequently, the coal fines sieved in 0.038 mm – 0.020 mm were used for the series of the experiment of adsorption and coal fines fixation in the proppant pack column. The coal fines size range studied in this work (0.038 mm – 0.020 mm) is consistent with previously reported studies (Huang *et al.* 2010a; Bai *et al.* 2015; Zhao *et al.* 2016; Bai *et al.* 2017).

The glass bead proppant was kindly provided by Potters beads of Metal Finishing Glass Beads Potters, with a nominal diameter (80%) Ballotini ® Metal Finishing Beads of Potter Designation B, US Sieve 30-40 (600-475 microns), and minimum roundness of 65%, where the composition of glass beads can be seen in Table 2. Using the electrical sieve shaker, proppants were sieved to US sieve size of 35 (~475 microns) that were used in the adsorption proppant pack experiments.

The silica nanoparticles are insoluble, hydrophilic, and non-polar in water, where those used in this study were procured from Sigma-Aldrich. The properties of the silica nanoparticles are presented in Table 3. An electrolyte of 0.6 M NaCl (as compared to standard saline) was used in the adsorption experiments. A constant pH of 9.0 ± 0.2 was set in all of the experiments as similar to coalbed methane reservoirs. Various concentrations of silica nanoparticles were used to formulate nanofluids. DI-water (Ultrapure Type 1 Water) was used as a base fluid for the nanofluid and to formulate 0.01 M and 0.6 M brine after mixing with NaCl (58.44 g/mol, purity ≥ 99.5 mol%, from Rowe Scientific).

2.2. Methodology

Various SiO_2 NPs concentrations (0.01 – 0.1 wt%), based in 0.01 M and 0.6 M brine-based solutions, were investigated in various coal concentration (0.1-1 wt%) to examine their stability, adhesion to the glass beads proppant, and coal fines adsorption efficiency. The formulation of nanofluids was carried out with two constituents, the dispersed phase and the dispersion medium. The dispersed phase was nano-sized silicon dioxide (SiO_2) - also known as SNPs or Silica NPs, with weight percentages ranging from 0.01 to 0.10, while the dispersion medium was DI water, 0.01 M and 0.6 M NaCl. The dispersion stability was investigated using Malvern Z3600 Nano zeta sizer. The nanofluids were formulated by sonicating nanoparticles in the base fluid (Mahdi Jafari *et al.* 2006) using an ultrasonic processor (VCX 750, a 750-watt ultrasonic processor, frequency - 20 kHz from Sonics & Materials, Inc.). Whilst the time and power of the sonication process depends mainly on a load of dispersed nanoparticles (Shen and Resasco 2009), in this work all the formulated dispersions were sonicated with the same sonication time, energy, amplitude and power of 300 seconds, 4 MJ, 30%, and 240 V respectively to assure a duplicated conditions for all formulations. The prepared nanofluids were visually observed to assess any significant instability in the behavior of the nanoparticles during the required soaking period. All experiments were carried out under ambient conditions of pressure = 101.3 kPa and temperature of 295.15 ± 3 K.

The process of adsorption of sandstone fines using nanoparticles affixed on glass beads has also been studied by Huang et al. (2008) and Ahmadi et al. (2013) in addition to other researchers (Huang *et al.* 2008b, 2008a; Belcher *et al.* 2010; Huang *et al.* 2010b; Ahmadi *et al.* 2013b, 2013a; Habibi *et al.* 2013; Habibi *et al.* 2014). However, SNPs coating onto the proppant pack quantitatively has only been discussed by a few researchers such as Abhishek and Hamouda (Abhishek and Hamouda 2017). The only reported patent of using nanoparticles for affixing coal fines highlights the use of MgO to affix coal fines (Huang *et al.* 2010a). The selection of the proppant is influenced by the mechanical properties of the formation rock and the properties of the proppant itself. The parameters that impacted the selection of the proppant include crush resistance and size of the proppant amongst others. Hollow glass spheres have been reported to have the least specific gravity of 0.8 to 1.4 in all the lightweight proppants (Parker *et al.* 2012; Liang *et al.* 2016).

The method of affixing nanoparticles onto the surface of proppants is critical to the experiments; thus, in this research, we have soaked the NPs for 24 hours and calculated the nanoparticle coating efficiency via turbidity results. Note that SDBS (an anionic surfactant) is used in all experiments to flow through the slurry with minimal adsorption. The adsorption efficiency is defined in Eq. 1 (Habibi *et al.* 2014). The results of influent and effluent slurry were obtained using the particle counter sizer.

$$\text{Adsorption efficiency } (\%) = 100 - \left[\frac{C_{eff}}{C_{in}} \times 100\right] \qquad \text{Eq. 1}$$

Where,

C_{eff}: Concentration of coal fines in the effluent slurry, gm/cc

C_{in}: Concentration of coal fines in the influent slurry, gm/cc

A glass column was used in all of the experiments of adsorption, in which flow took place merely because of gravitational force, where a schematic of the procedure adopted in the column can be seen in Fig. 1 below.

2.3. Experimental set-up workflow

The laboratory study model and equipment will be adapted from Habibi et al. and Ahmadi al. and used to see the effect of silica nanoparticles on coal fines adsorption (Ahmadi *et al.* 2013b, 2013a; Habibi *et al.* 2013; Arab *et al.* 2014; Habibi *et al.* 2014). In this study, bare silica nanoparticles, as well as treated silica nanoparticles with Sodium Dodecyl Benzene Sulfonate (SDBS), were analyzed with a Malvern Nano Zeta Sizer to determine their stability. Following this, a turbidity calibration curve for silica NPs was made using a HACH 2000 Turbidimeter. The main experiment was the proppant pack tests conducted in the glass column at ambient conditions in which coal slurry flowed into a proppant treated with SNPs. This was compared with proppant without any SNP treatment. The results of the coal slurry were obtained using a particle counter analyzer. The corresponding experimental workflow is shown in Figure 2.

3. Results and Discussion

3.1. Silica Nanoparticles dispersion using SDBS

In order to modify the dispersion stability and surface chemistry of SNPs, an anionic surfactant SDBS was used. In the three dispersion mediums (DI water, 0.01 M NaCl, and 0.6 M NaCl) tested, SDBS reduced the zeta-potential and effectively enhanced the dispersion stability, as shown in Figure 3. Note that the dispersion stability is more effective in DI water and low salinity (0.01 M NaCl) rather than in the high salinity (0.6 M NaCl) suspension.

The zeta-potential measurements of bare SNPs in both ionic strengths are consistent with Al-Anssari et al. (2017) (Al-Anssari *et al.* 2017a). With increases in ionic strength, the absolute zeta-potential value decreases, resulting in reduced dispersion stability. However, the DI water and low ionic strength (0.01 M NaCl brine) provide better stability enhancement when SNPs are treated with SDBS.

3.2. Effect of Nanoparticles adhesion onto proppant pack

In order to examine the coating of nanoparticles via the soaking method, we first calibrated SNPs in DI water. The SNPs turbidity increases with an increase in SNP loading, as can be seen in Figure 4. Note that the turbidimeter has a limitation of 1000 NTUs. When on the x-axis, SNP loading is increased while on the y-axis, the turbidity value in NTU increased with the loading resulting in a slope value of 3390.5.

It was observed that higher salinity has lower zeta-potential, which leads to higher retention efficiency of SNPs by soaking method, as seen in Figure 5. The results show that SNPs based in DI water yielded retention efficiency of 61-67%, while 0.01 M NaCl (as comparable to potable water) yielded 60-72% retention efficiency, and 0.6 M NaCl (as comparable to seawater) yielded 81-85% retention of SNPs onto the glass bead proppant pack. This retention is due to the zeta-potential values, as obtained in Section 3.1, which can also be seen in Figure 3. Thus, aggregation of nanoparticles in 0.6 M NaCl ionic strength due to its low zeta-potential, aids it in retaining SNPs in the proppant pack.

Thus, in further experiments after observing better retention of 0.6 M NaCl cases in two coal concentrations (0.01 wt% and 0.1 wt%), we conducted the rest of the experiments of adsorption in 0.6 M NaCl salinity. This is consistent with the DLVO theory, which defines principles based on which the particles (in our case SNPs) aggregation take place at higher salinities.

3.3. Effect of Chemically modified Nanoparticles on coal fines fixation

Chemically modified nanoparticles (in this case, silica nanoparticles treated with SDBS) were injected into the glass-column, followed by the introduction of the glass-bead proppant pack. The results showed that 0.1 wt% of SNPs yielded maximum adsorption of all tested coal fines (0.1, 0.5, and 1 wt%), as can be seen in Figure 6. Note that there is a difference in adsorption of coal fines with weight percentages (even when passing through an untreated proppant pack), as a higher concentration of slurry yields lower dispersion stability, leading to clogging in the proppant pack (Al-Anssari *et al.* 2017a).

The base case has been optimized in our previous work (Awan *et al.* 2019), giving a C_{eff}/C_o of approximately 83%, meaning the adsorption of coal fines is 17% (using 0.1 wt% coal fines treated with 0.001 wt% SDBS). The rest of the coal slurries also have 0.001 wt% SDBS, but their coal fines are of higher weight fraction. It can also be observed that untreated proppant packs in various coal concentrations result in an increasing trend to rising coal concentration, i.e., 17% adsorption for 0.1 wt% coal fines, 27% adsorption for 0.5 wt% coal fines, and 30% adsorption for 1.0 wt% coal fines. This occurs due to the lower stability of the coal slurry as a result of the increase in weight (Al-Anssari *et al.* 2017a), which ultimately causes the straining of coal fines due to their aggregation. Additionally, not all of the coal fines retention in the proppant pack is due to the interaction of attractive forces of nanoparticles with coal fines, there is also an aggregation of coal fines and straining in the proppant pack as has been demonstrated by Zou et al. (2014) (Zou *et al.* 2014) which can be seen in Fig. 6.

With increasing the SNP loading in the proppant pack, its retention efficiency also increases. Thus, the nanoparticle treated glass beads can retard the mobilization of coal fines through the proppant pack and affix them near the source of their generation; thereby, reducing the damage to the pumps and minimizing the possibility of filling of the wellbore.

4. Conclusion

The above sets of experiments conclude that higher salinity yields higher coating efficiency of the proppant pack via nanoparticles due to their aggregation behavior (zeta-potential values of greater than -20 mV). SDBS enhances the dispersion stability of silica nanoparticles in various salinities tested up to 0.6 M NaCl. An optimum concentration of 0.1 wt% SNPs yielded maximum adsorption of coal fines in the proppant pack at higher ionic strength (0.6 M NaCl brine).

Further studies involving a comparison of various coating methods (e.g., calcination, sintering, soaking, oil coating, etc.) for irreversibly affixing nanoparticles on the proppant pack need to be conducted, in order to optimize the coating procedure in various saline environments. Further, coal particles of different ranks need to be studied to understand the behavior of various metal oxide nanoparticles, in order to fixate these coal fines by adsorption in the proppant pack. A comprehensive study to determine the impact of adsorption on the permeability can also be studied, as its implication in the field can be detrimental to natural gas recovery.

Conflicts of Interest

None.

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Biographies

Faisal Ur Rahman Awan is a Ph.D. candidate in Petroleum Engineering at Edith Cowan University, Australia. His work focuses specifically on the coal fines fixation using nanoparticles. Mr. Awan did his Bachelor's and Master's degrees in Petroleum Engineering. He has also been serving at Dawood University of Engineering and Technology, Karachi, as an Assistant Professor in Petroleum Engineering for the last seven years. He is a member of prestigious societies such as SPE, SEG, EI, and PEC.

Alireza holds a Ph.D. degree in Petroleum Engineering from the University of Adelaide, an M.Sc. degree in Reservoir Engineering from the University of Tehran (Iran), and a B.Sc. degree in Chemical-Petroleum Engineering from Petroleum University of Technology (Iran). He is presently serving as a Senior Lecturer at the School of Engineering at Edith Cowan University. Before joining ECU, Alireza was a research scientist in the CSIRO-Energy business unit, where he researched enhancing gas production from unconventional resources and CO_2-sequestration. Before pursuing his Ph.D. study, he was a Petroleum Engineer in the National Iranian Oil Company (NIOC) for six years. Alireza's research interests focus on Enhanced Oil/Gas Recovery from conventional and unconventional reservoirs. He is a member of SPE.

Hamed completed his Bachelor's and Master's in Petroleum Engineering. During his Master's study, he conducted numerical research on heavy oil EOR. He used two of the most professional petroleum simulators, CMG and Eclipse, in his studies. He changed his research field to Coalbed Methane in 2016 and received a scholarship for his Ph.D. studies at Edith Cowan University (ECU), Australia. For the time being, as a Ph.D. student at ECU, he is experimentally researching on Coalbed Methane productivity enhancement as his priority, and also partially on enhanced oil recovery and CO_2 geo-sequestration. He is a member of SPE.

Sarmad Al-Anssari is currently a senior lecturer in Chemical Engineering at University of Baghdad, Iraq. He earned a bachelor's and master's degree in chemical engineering from University of Baghdad and he holds a PhD degree in chemical engineering/ Nanotechnology from Curtin University/ Australia. He worked as a faculty member in the university of Baghdad for more than 10 years and recently he is adjunct lecturer at ECU Australia and external supervisor at Curtin university/ Australia. His research interest is on different applications of nanoparticles and nanofluids in different disciplines, including wettability alteration, enhanced oil recovery, and carbon capture and storage.

Stefan Iglauer joined Edith Cowan University (ECU) in 2018 as a Professor to lead the developments in the Petroleum Engineering discipline. His research interests are in petrophysics and interfacial phenomena, mainly at pore-scale with a focus on CO2 geo-sequestration and improved hydrocarbon recovery. Stefan has authored more than 250 technical publications; he holds a Ph.D. degree in material science from Oxford Brookes University (UK) and an MSc degree from the University of Paderborn (Germany). He is a member of SPE.

Table 1: Properties of coal sample

Ash (%, db[a])	Density (g/cm^3)	Volatile matter (%, daf[a])	C (%, daf[a])	H (%, daf[a])	R_v, max[a] (%)	Vitrinite (vol% mmf[a])	Inertinite (vol% mmf[a])	Liptinite (vol% mmf[a])
4.798	1.31	5.042	78.50	5.37	0.91	81.01	12.73	6.26

[a] **db**: on a dry basis; **daf**: dried ash-free; **R_v, max**: maximum vitrinite reflectance; **mmf**: mineral matter free.

Table 2: Properties of glass beads used as proppant packs

Chemical	SiO_2	Na_2O	CaO	MgO	Al_2O_3	FeO/Fe_2O_3	Trace
Wt %	72.5	13.6	9.7	3.4	0.4	0.2	0.2

Table 3: Characteristics of nanoparticles

Nanoparticle	Linear Formula	Primary particle size (nm)	Chemical Structure Depiction	Specific surface area (m2/g)	Purity (%)	Density (g/cc)	Molecular mass (g/mol) @ 298.15 K	Boiling point (K)	Melting point (K)	Additional Description
Silicon dioxide	SiO_2	5-15		140	≥ 99.50	2.20-2.60	60.08	2503	1873	Porous spherical

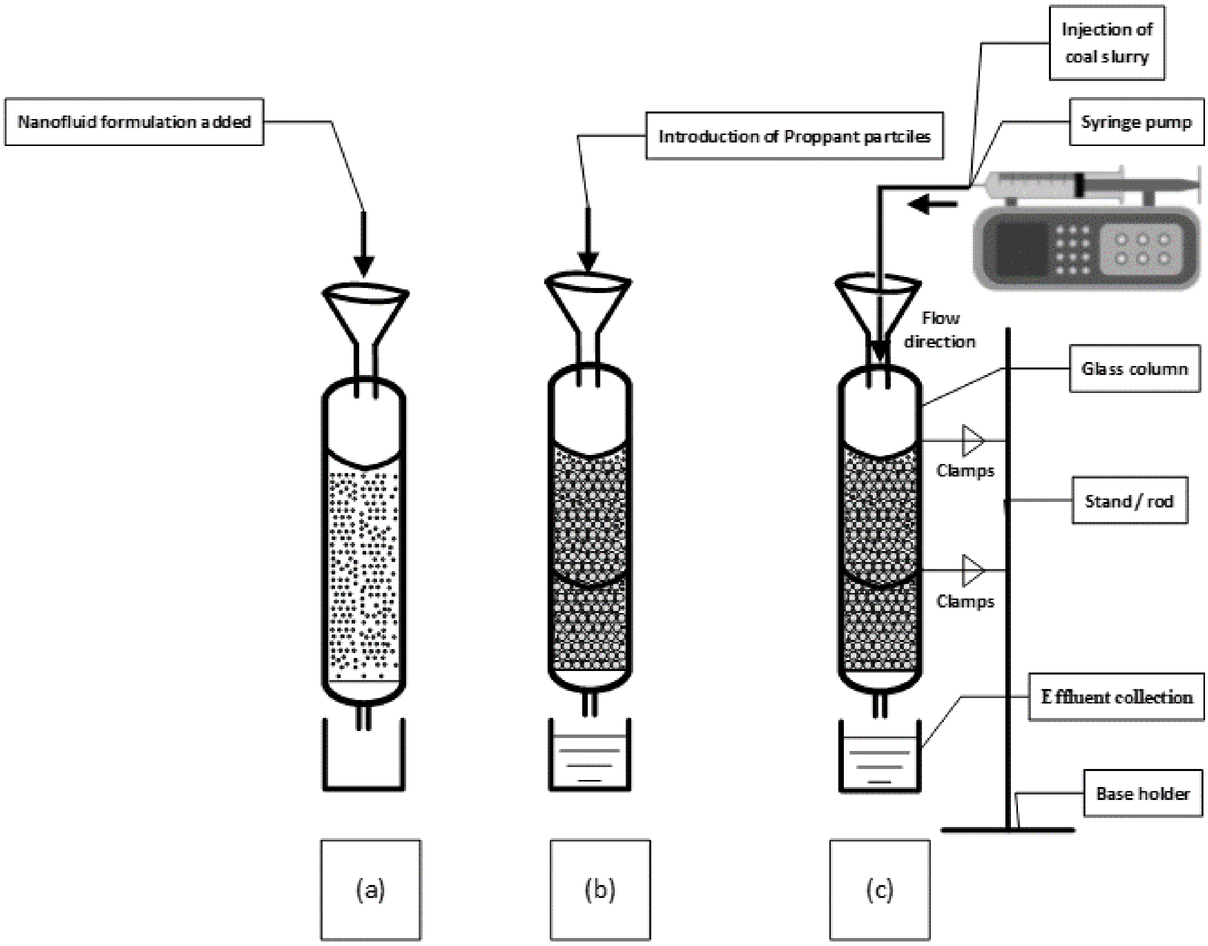

Figure 1: Schematic of (a) Silica nano-formulation added to the glass column, (b) Introduction of proppant particles in the nano-formulation (c) Injection of coal slurry at 1 mL/min

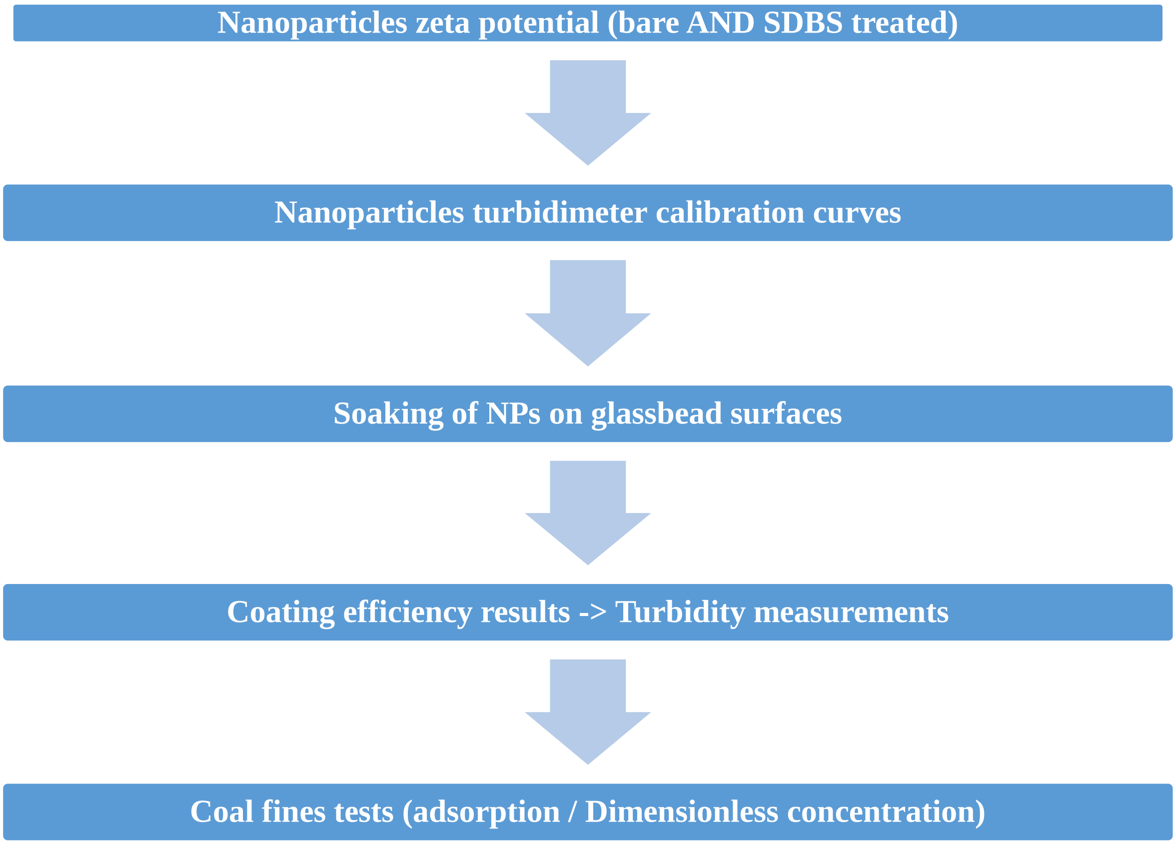

Figure 2: Workflow of experiments for determining the effect of nanoparticles on adsorption of coal fines

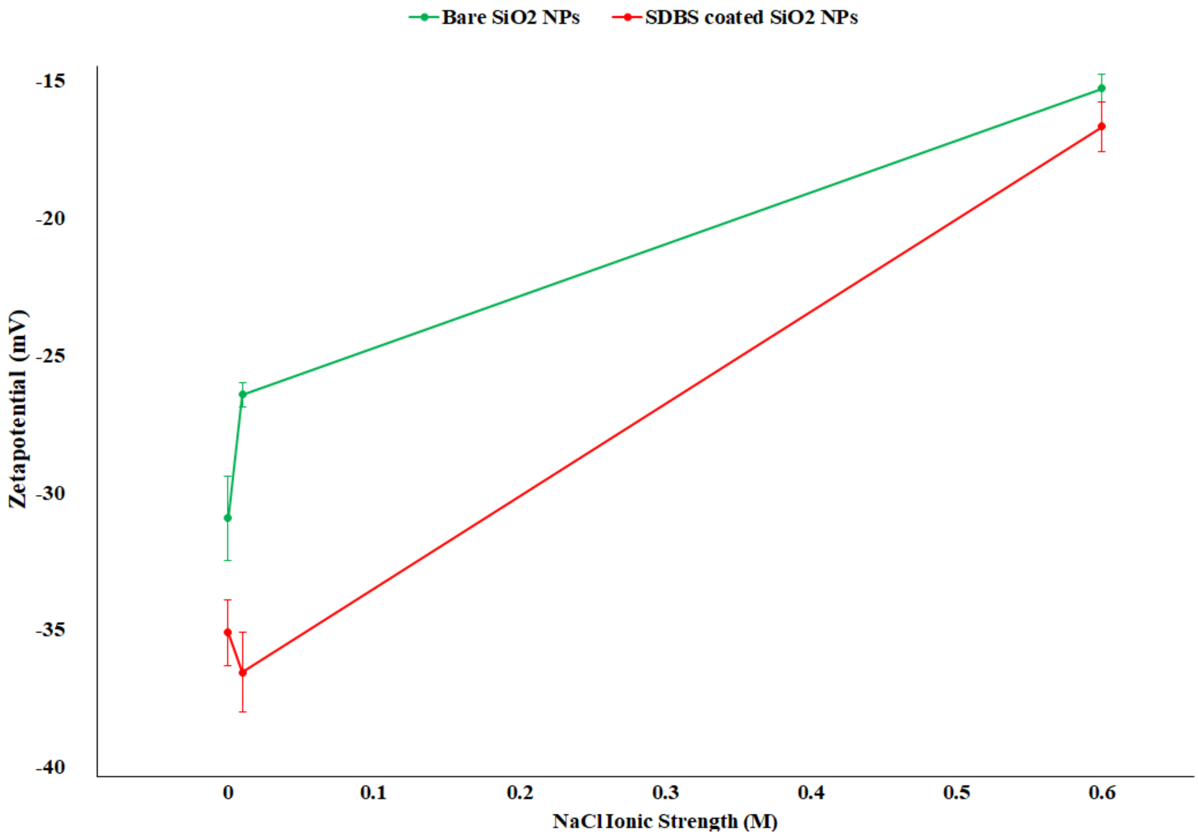

Figure 3: Effect of SDBS in various ionic strength (0, 0.01 and 0.6 M NaCl brines) on zeta-potential for 0.1 wt% silica NPs at pH=9 and temperature=298.15 K.

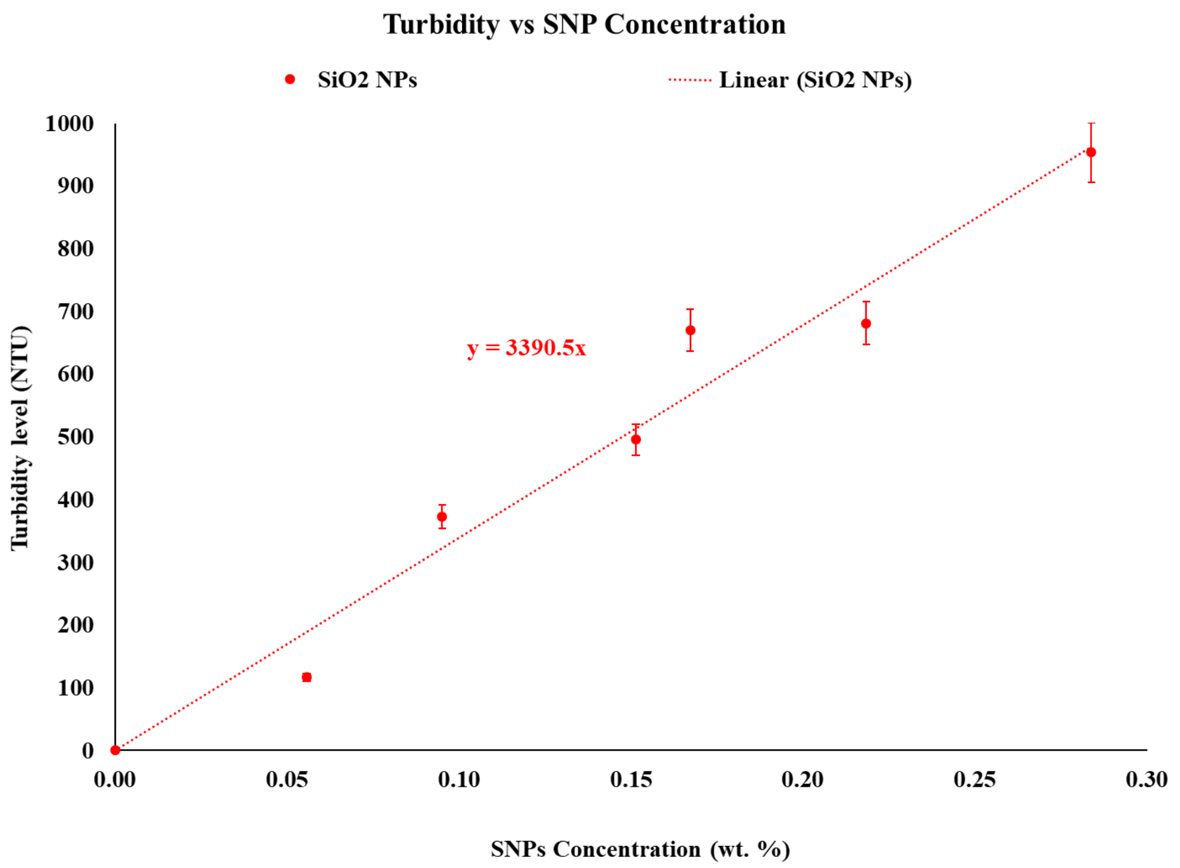

Figure 4: Turbidity results of silica nanoparticles in various concentrations from 0 to 0.3 wt% (R^2 of the Linear SiO_2 is 0.967)

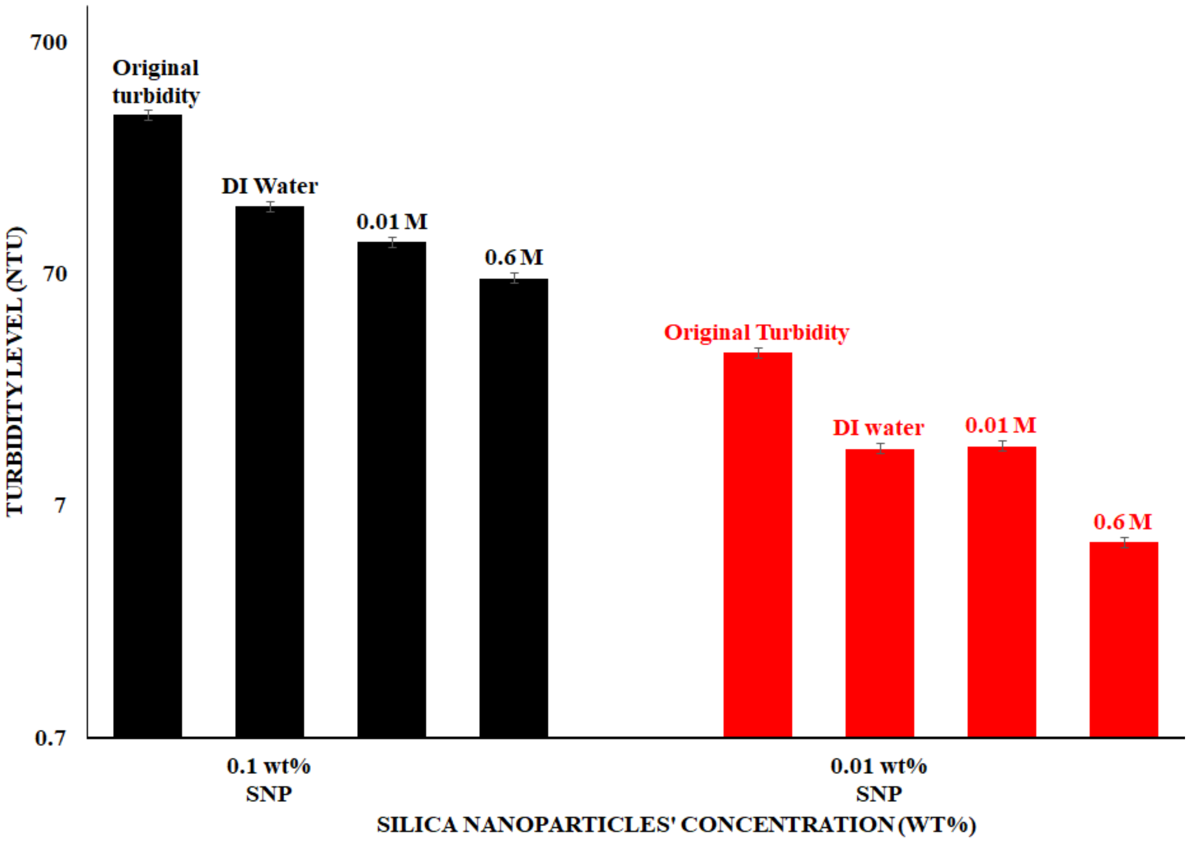

Figure 5: Turbidity results for silica nanoparticles coating via soaking method results in various suspensions (DI water, 0.01 M, and 0.6 M NaCl brines)

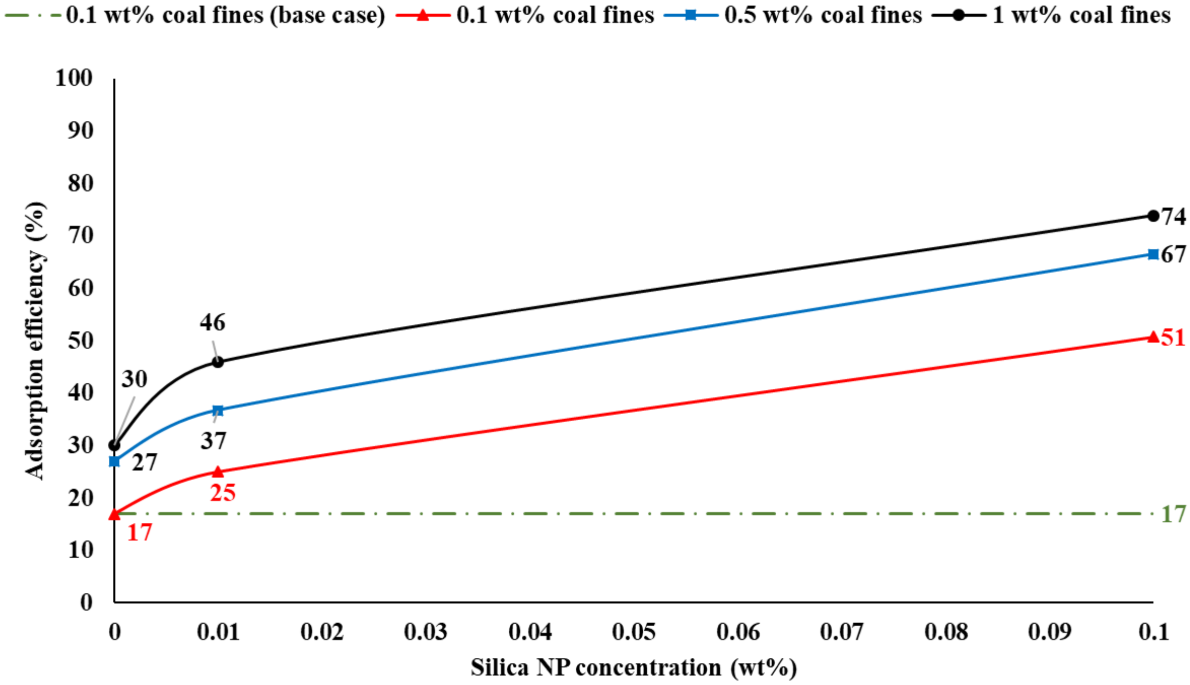

Figure 6: Results of adsorption of the various coal fines concentrations (0.1, 0.5, and 1 wt%) using no nanoparticles, 0.01 wt%, and 0.1 wt% in 0.6 M saline environment in a proppant pack. Note the base case is 0.1 wt% coal fines passed through the untreated proppant pack