

Nanotechnology in the petroleum industry: Focus on the use of nanosilica in oil-well cementing applications - A review

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ABSTRACT

The usage of nanotechnology has gained widespread attention in the petroleum industry in recent years. Existing studies indicate that incorporation of nanoparticles into the cement matrix improves cement properties such as strength, microstructure and durability. This improvement is attributed to the nanoscale size and high specific surface area of nanoparticles. To fulfill the purpose of this paper, previous studies associated with the role of nanotechnology in the petroleum industry, with specific attention to the use of nanosilica in oil-well cementing applications are reviewed. The effect of silica nanoparticles on both fresh and hardened cement properties are investigated and presented. The study has revealed that the addition of nanosilica improves the performance of cement, ensuring adequate zonal isolation and extended well life. These findings highlight the potential use of nanotechnology in the petroleum industry. In terms of future study, more investigation and experimentation into the use of nanosilica in oil-well cementing is strongly recommended.

1. Introduction

Oil-well cementing is a process of injecting cement in the annulus between the casings and the rock formations exposed to the wellbore (Fig. 1) (Nelson and Guillot, 2006; Newlove et al., 1984). The primary functions of the cementing process are; sealing (prevention of fluid communication between different zones), provision of structural support for the casings and protection of the casings from attack by corrosive fluids (Fakoya and Shah, 2017; Newlove et al., 1984). Besides, cement is pumped into the wells for squeeze jobs, plugging operation, etc. (Fakoya and Shah, 2017). Failure of the bonded cement to perform such functions can result in one or more of the following problems; contamination of fresh water aquifers, loss of hydrocarbon reserves in producing wells, loss of injected fluids in injector wells, sustained casing pressure or even a blowout (Agbasimalo, 2012).

However, for a long period, formation fluid leakage has been described as a major challenge in oil and gas wells. The fluid leakage is connected with the flow of fluids through the cemented annulus portion of the wellbore (Fig. 2). High costs of remedy, production, and abandonment, as well as environmental effect are some of the impacts of fluid leakage into the wellbore. Some studies have revealed that poor cement-rock interaction contributes to leakage of formation fluids. Earlier reports on oil-well integrity, globally, indicate that 4.6% of 316,000 wells

examined in Alberta experienced leaking complications (Opedal et al., 2014; Taoutaou et al., 2015). In the Norwegian zone of the North Sea, about 13–19% of the producing wells and 37–41% of the injection wells experienced leakage (Nygaard, 2010; Watson and Bachu, 2007). Additionally, above 8000 wells exhibited gas immigration problems in the Gulf of Mexico (Radonjic and Oyibo, 2014) in the circumstances, leakage was attributed to various causes, including inappropriate well-cementing mechanisms and worsening of cement body during production and after well relinquishment (King and King, 2013).

To obtain a high strength bond between cement and formation, cement performance and properties must be improved. According to Nelson and Guillot (2006), a well that lacks a good cement job may never attain its complete production potential. Hence it is essential to develop high strength and durable cement for long term zonal isolation to obtain maximum value from the well. Moreover, they proposed that oil-well cement must be modified to enhance its functionalities. In that regard, various researchers in recent years have concentrated on the use of materials that can provide high performance to boost the lifespan of the well and reduces costs associated with the renovation or losses due to failures in the cement sheath. One such material is nanoparticles; which has gained currency in cement related researches across the world.

Nanoparticles are the simplest form of structures with dimensions in

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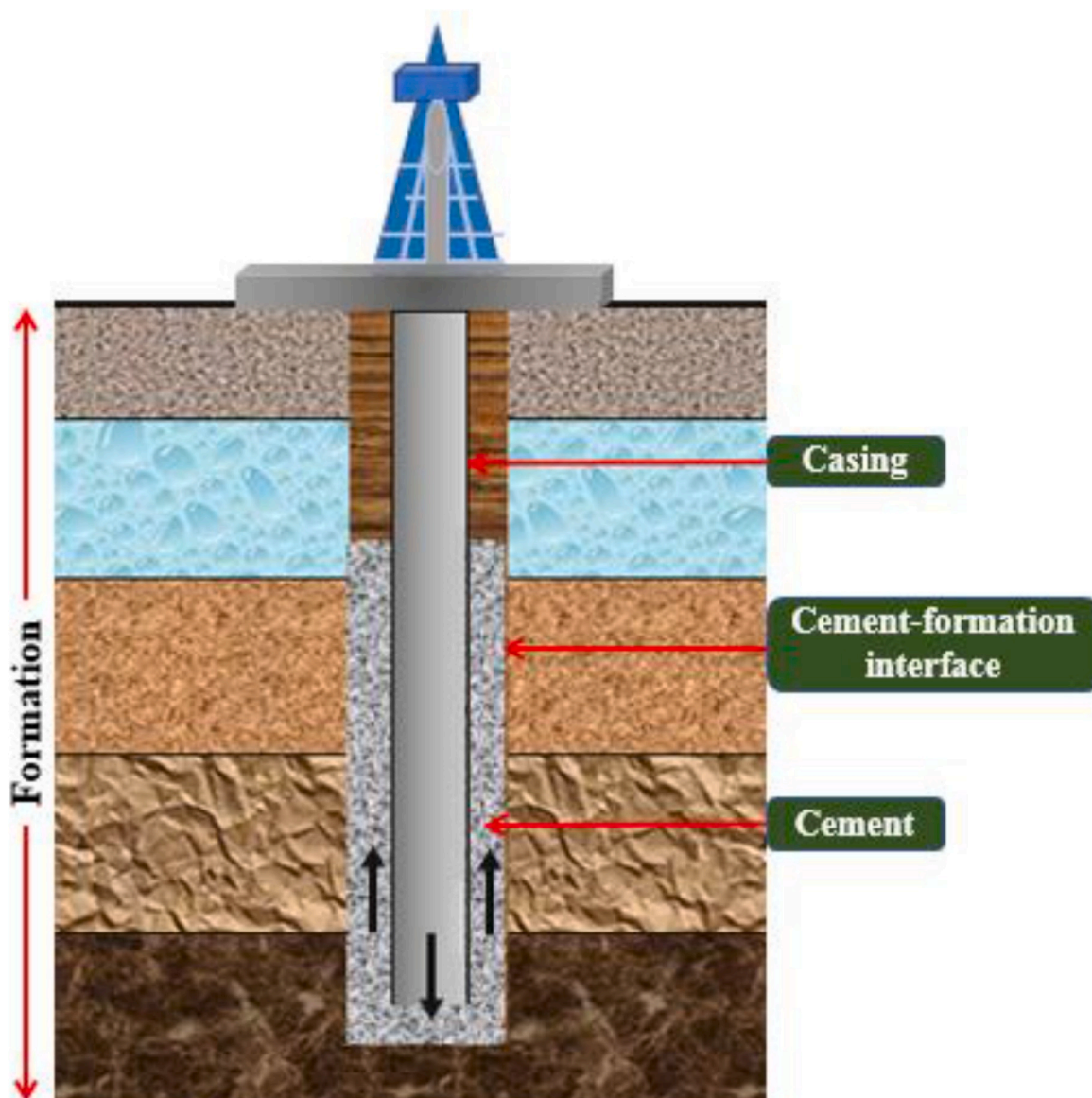


Fig. 1. The schematic diagram for primary oil-well cementing.

the range of nanometer (nm). Technically, any collection of particles fused together with a structural radius of less than 100 nm, can be considered a nanoparticle (El-Diasty and Ragab, 2013). Recently, the nanoparticles have shown an excessive potential to improve the performance of cementitious materials (El-Diasty and Ragab, 2013; Siddique and Khan, 2011; Singh and Ahmed, 2010; Sinnot and Andrews, 2001; Wong et al., 2015). Due to their size that ranges from 1 to 100 nm, the nanoparticles possess exceptional properties that are at variance with their macro-sized counterparts. Apart from their size effect, nanoparticles have special characteristics including enormous surface area per volume ratios (Fig. 3) and higher chemical reactivity which triggers their ability to modify properties in cement (Amanullah and Al-Tahini, 2009; Patil and Deshpande, 2012; Senff et al., 2010; Siddique, 2011; Stefanidou, 2012). A comparison of the surface area to volume ratio of spherical elements of the same substance with a radius of 1 mm, 1 μ m and 1 nm was reported by Fakoya and Shah (2017). As indicated in Fig. 4, more than a million-fold increase in surface area per volume ratio would be achieved when a particle is converted from a millimeter dimension to a nano-scale. It was summarized that the size effect of nanomaterials compared to micro and macro materials of the same parent substances, provide special properties to the nanos that are

non-existent in the parent substances. By incorporating nanoparticles into oil-well cement, both fresh and hardened properties of cement are modified. Early strength enhancement, improved long-term tensile to compressive strength ratio, reduction in setting time and microstructure modification are examples of the cement property improvements resulting from nano-scale particles (Ravi et al., 2002; Santra et al., 2012). In addition, Santra et al. (2012), demonstrated the impact of nanomaterials in oil-well cement hydration and mechanical strength using nanosilica and nanoalumina of 30 nm and 140 nm particle size respectively. Their study revealed that a set Portland cement mix without any mechanical property modifications has a tensile to compressive strength ratio of about 10%. They further reported that nanomaterials can alter this ratio by increasing it, which demonstrates that strength of the cement sheath can be promoted with nanoparticles for extended well life. Based on that fact, nanoparticles are used either to replace a portion of cement or as an additive in the cement to counter undesirable situations such as high temperature and pressure for ensuring successful zonal isolation (Santra et al., 2012).

Furthermore, several studies report that addition of nanoparticles in cement accelerates the hydration mechanism which reduces cement's wait time; hence saving rig working hours (Chong et al., 2012; Fakoya

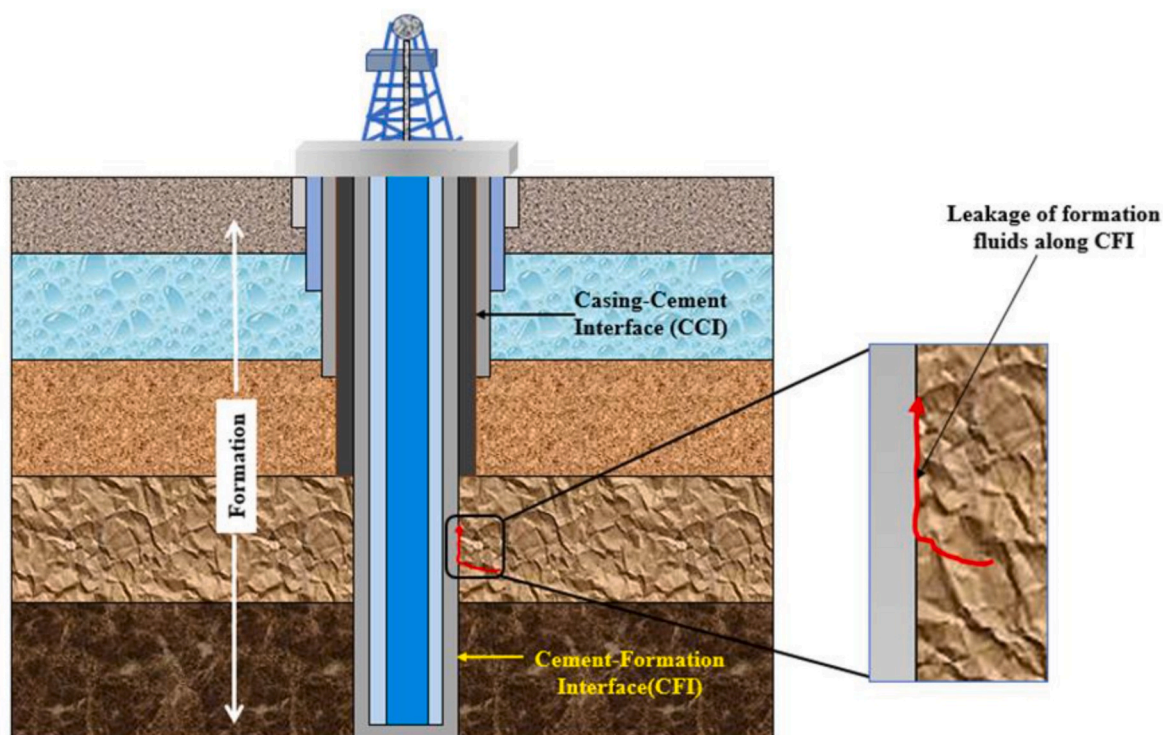


Fig. 2. The schematic diagram for fluid leakage along with the cement-formation interface due to poor cement bonding.

and Shah, 2017; Hou et al., 2013; Kadri and Duval, 2009; Kim et al., 2014; Santra et al., 2012). Compared to other types of nanoparticles, many authors have recommended nanosilica materials to be the best accelerators for cement hydration due to its pozzolanic reactivity (Gai-tero et al., 2008; Heikal et al., 2015; Kong et al., 2012, 2013; Kontoleontos et al., 2012; Kurdowski and Nocun-Wczelik, 1983; Land and Stephan, 2012; Lin et al., 2008; Ltifi et al., 2011; Nabhani et al., 2011; Qing et al., 2006; Quercia and Brouwers, 2014; Quercia, 2014; Wang et al., 2017).

Much of the available literature reports that nanotechnology provides an extensive range of alternatives for technologies and materials to be used in the oil and gas field. This view which is supported by Agista et al. (2018), Fakoya and Shah (2017), Khalil et al. (2017) and Quercia and Brouwers (2010) conclusively show that research and development of nanotechnology in the petroleum industry have been very active in recent years. Currently, the share of nano-articles in the entire scientific periodicals is significantly growing. According to StatNano (2019), more than 166,000 research papers associated with nanoparticles were issued, indicating the addition of more than 7% compared to the same period last year. The share of nanomaterial-based articles among the total number of articles worldwide each year from 2011 to 2018 is indicated in Fig. 5 (StatNano, 2019). Part of these studies is directly associated with the use of nanotechnology in the oil-well cementing. Thus, through nanotechnology advent, the petroleum industry could address some persistent challenges presently facing the discipline through the design of novel products that function at an extraordinary level. In this paper, different studies linked with the use of nanomaterials in the petroleum industry are reviewed. Emphasis is given to the influence of nanosilica to boost cement properties.

2. Nanosilica

Nanosilica, also known as silicon dioxide nanoparticles, or silica nanoparticles is a compound of silicon and oxygen. Its chemical formula is SiO_2 , and it presents in the form of white powder. These compounds are plentiful in the form of quartz or sand; and are therefore widely used

as part of the building blocks of numerous natural and artificial structures (Fakoya and Shah, 2017). The recent introduction of nanotechnology has revealed that materials at small particles can significantly improve properties compared to the same materials at large scale. Petroleum well cementing is among the domains that benefit from the advent of this technology. Numerous types of nanoparticles such as nanosilica, alumina, titania, magnesium oxide and iron oxide have been considered as cement additives (Agista et al., 2018; Jafariesfad et al., 2016; Quercia et al., 2016; Senff et al., 2012). However, several reports have argued that nanosilica is the most investigated nanoparticles for cementing applications, and it has proved to form cementitious material with higher performance (Agista et al., 2018; Santra et al., 2012; Wang et al., 2017).

There are several possible mechanisms of nanosilica in cement. Firstly, the nano-scale size of silica particles allows them to be used as filler material by filling up voids within the cement matrix, resulting in a dense and compact structure with reduced capillary porosity. Secondly, nanosilica materials have higher pozzolanic activity, making them suitable accelerators for cement hydration (Bhuvaneshwari et al., 2012; Khayati et al., 2015). Silvestre (2015) studied the effect of nanosilica during the hydration of cement. Based on the results, it was conclusively argued that the blending of nanosilica into cement grains creates $\text{H}_2\text{SiO}_4^{2-}$ that reacts with the existing Ca^{2+} to produce an excess calcium silicate hydrate (C-S-H) as displayed in Fig. 6. These C-S-H gel spread in the water between the cement particles and act as seeds for the production of a more compacted C-S-H phase. Therefore nanosilica particles stimulate the pozzolanic reaction. Moreover, nanosilica increases the consumption of $\text{Ca}(\text{OH})_2$ and creates an additional C-S-H gel which is the key component for strength in hardened cement pastes (Chithra et al., 2016; Silvestre, 2015). Other studies posited that the proportion of $\text{Ca}(\text{OH})_2$ is reduced by nanosilica and converted into C-S-H gel hence increasing the hardness of cement (Bahadori and Hosseini, 2012; Gai-tero et al., 2008, 2010; Qing et al., 2007). The usage of silica nanomaterials to promote hydration in oil-well cement was also reported by Pang et al. (2009). The nano-sized silica particles of 4–6 nm were noted to be the best cement hydration accelerators with 30 and 136% increase

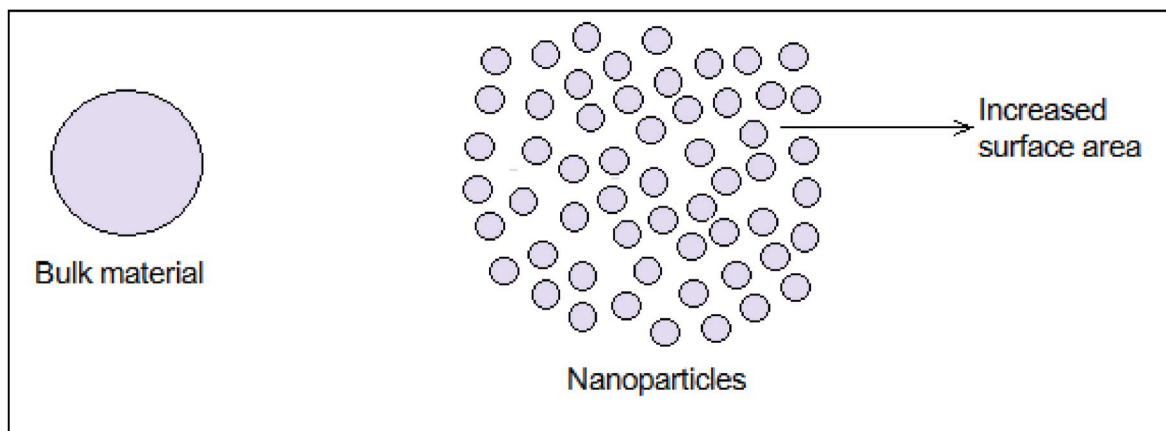


Fig. 3. Increasing the surface area with nanoparticles, modified from (El-Diasty and Ragab, 2013).

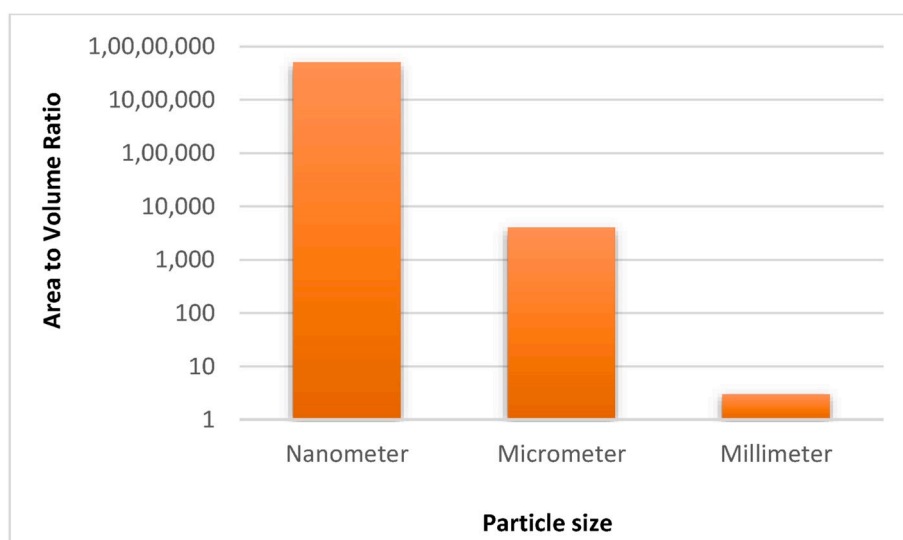


Fig. 4. Surface area to volume ratio of the same volume of material, modified from (Fakoya and Shah, 2017).

in compressive strength within a period of 2 and 7 days respectively.

2.1. Production methods of nanosilica

Different researchers have admitted that nanosilica can be manufactured easily since its physical and chemical properties are well recognized. Others have established that nanosilica particles can be prepared with various characteristics including hydrophilic and hydrophobic both of which are non-toxic inorganic substances (Hendraningrat, 2015; Miranda et al., 2012). The nanosilica technology offers a means to attain significant modifications in the physical, chemical and mechanical behavior at the macro-scale. This is due to the exceptional properties of nanosilica based materials' nano-scale dimension (Quercia et al., 2016). Several methods that have been followed during the invention of nanosilica can be divided into two main techniques, top-down and bottom-up (Klabunde, 2001; Reverchon and Adami, 2006). Top-down (physical approach) is characterized by decreasing the extent of the original dimension through the application of size lessening techniques. On the other hand, bottom-up (chemical approach) contains a common path for manufacturing nanosilica from molecular or atomic size (Rahman and Padavettan, 2012).

Currently, nanosilica is manufactured from different methods. One of the methods is based on a sol-gel technique (organic or water route) at ambient temperatures (Quercia, 2014). In this method, the preparatory

materials mostly (Na_2SiO_4 and organometallics like tetraethylorthosilicate (TEOS)), are added to a solvent and the pH of the solution is altered, obtaining the precipitation of silica gel (Fig. 7). The created gel is aged and sieved to obtain a xerogel (Sakka and Kosuko, 2000). The formed xerogel is then desiccated and burnt or dispersed again, with the stabilizing agents (Na, K, NH_3 , etc.) to create a concentrated dispersion (20–40% solid amount), appropriate for cementitious materials (Sobolev and Ferrara, 2005). The general reactions of TEOS that form nanosilica in the sol-gel technique can be represented as shown in Fig. 8 (Rahman and Padavettan, 2012). In addition, reviewed literature indicates that the sol-gel method is the most used approach for producing silica nanoparticles due to its capability in controlling the size of the particles, as well as its distribution and morphology through methodical monitoring of the parameters in the reaction (Rahman and Padavettan, 2012). Several mechanical strength and water absorption tests were performed using nanosilica powders manufactured by the sol-gel method (Reddy et al., 2019).

As demonstrated by Quercia, one of the substitute nanosilica invention methods involves the vaporization of silica between 1500 and 2000 °C by decreasing quartz (SiO_2) in the heating system (Quercia, 2014). Based on that procedure, nanosilica is manufactured as a by-product of silicon metal and ferro-silicon alloy, where it is obtained by consequent condensation to fine particles in a cyclone (Jonckbloedt, 1997). This method produces very fine nanosilica comprising of

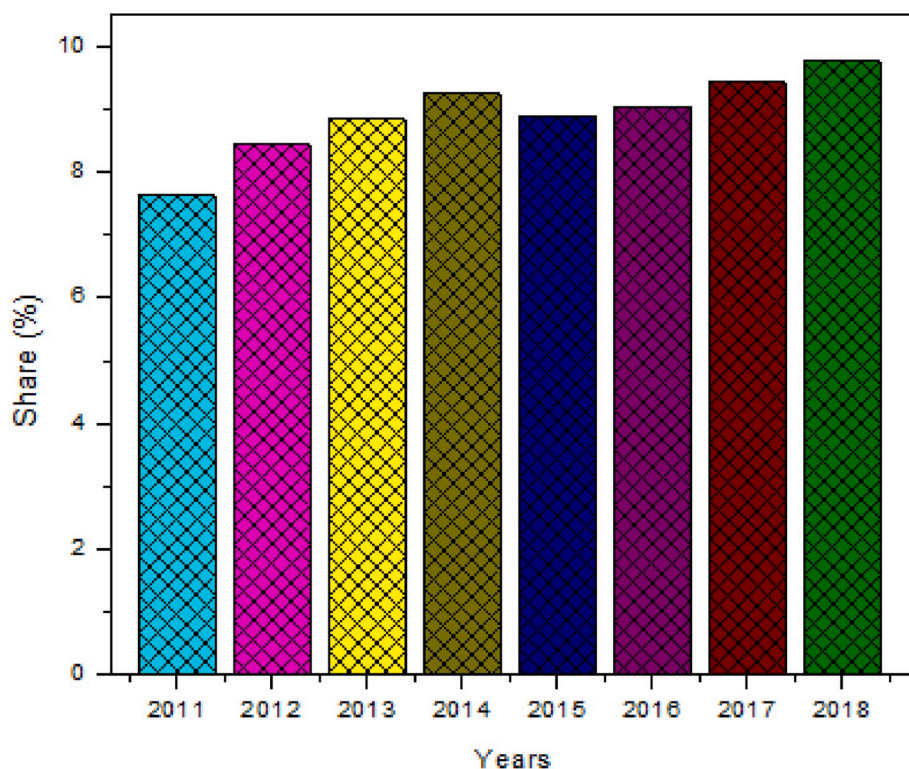


Fig. 5. The share of nano-articles in the global published works from 2011 to 2018. Data source (StatNano, 2019).

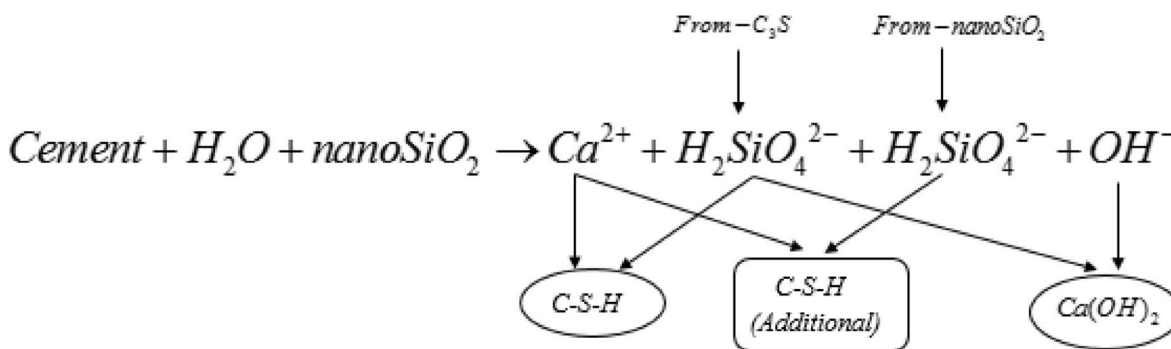


Fig. 6. Production of Calcium silicate hydrate (C-S-H) from nanosilica.

spherical particles with an average diameter of 150 nm and an enormous surface area of 15–35 m²/g (Rahman and Padavettan, 2012). However, the manufacturing of nanosilica using this approach consumes huge quantities of fuel, making the process very expensive and environmentally unfriendly due to CO₂ emissions (Quercia, 2014). Additionally, Esteves et al. designed a biological process that can make a thin and bimodal distribution of nanosilica from the digested humus of California red worms (55–245 nm) based on the temperature of calcination (Esteves et al., 2010). It is argued that this biological method can produce nanosilica particles with a spherical shape with a process efficiency of 88%. As demonstrated by Quercia (2014), these nano-scale particles were manufactured by nourishing worms with rice husk, a biological waste substance having 22% of SiO₂ equivalent. Numerous literature reviews also report that there is a possibility of producing nanosilica by using the precipitation method. In this approach, nanosilica can be precipitated from a solution at the temperature ranging between 50 and 100 °C. Because of that reason, nanosilica obtained from this process is called precipitated silica (Iler, 1955; Lieftink, 1997; Sobolev et al., 2006). This method which was first established by Iler (1955), make use of different precursors such as sodium silicates (Na₂SiO₃), burnt rice

husk ashes (RHA), semi-burnt rice straw ashes (SBRSA) and magnesium silicates (MgSiO₃) (Iler, 1955; Lieftink, 1997; Sakka and Kosuko, 2000; Thuadaj and Nuntiya, 2008; Zaky et al., 2008).

According to the most recent research finding, a new substitute method for producing nanosilica particles using low energy conditions was developed (Jonckbloedt, 1997; Lazaro et al., 2012). This technique includes the dissolution of one of the quickest weathering silicate compounds, olivine, in an acidic environment, at a low temperature ranging between 50 and 90 °C to yield an amorphous nanosilica (Jonckbloedt, 1997; Lazaro et al., 2012). Moreover, Lazaro (2014) reported the possibility of using waste acids such as sulfuric acid to produce olivine nanosilica (OnS); making this method less expensive. He further demonstrated that in comparison with nanosilica particles produced by other methods, OnS has unlike characteristics such as highest fineness (6–30 nm) and purity. However, Lazaro (2014) revealed that the OnS is produced at negative pH, leading to the formation of agglomeration silica nanoparticles with enormous surface area (200–400 m²/kg), besides being concentrated with silanol (OH) surface groups that affect its reactivity. Therefore, further studies on this new technique of producing nanosilica are recommended.

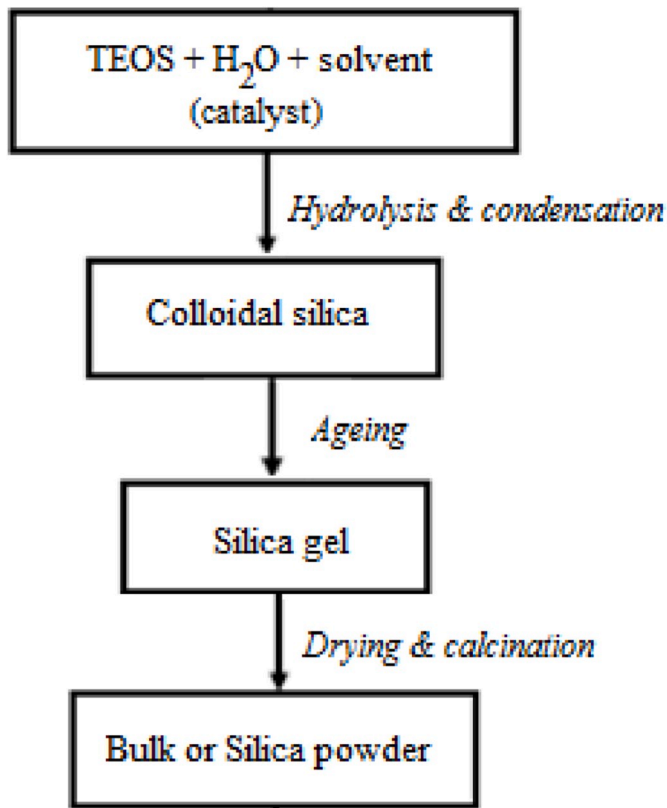


Fig. 7. Flow chart of a typical sol-gel process, adapted from (Rahman and Padavettan, 2012).

In comparison to other kinds of nanoparticles, nanosilica has lesser production costs and environmental consequences (Metin et al., 2012); since silicon dioxide nanomaterials are observed as the most economical and environmental friendly (Hendraningrat and Torsaeter, 2014). However, current reports argue that the cost of many nanosilica products is higher hence restricting the actual field application of these potential materials (Reddy et al., 2019). Therefore, future studies should focus on developing new techniques aimed at lessening the raw materials, consumption of energy and the nanosilica production costs. Producing nanosilica at lower costs would enable manufacturers to supply it at an affordable price; hence increasing its practical oilfield applications. Moreover, extensive studies are required to disclose the impact of nanoparticles such as nanosilica on oil-well cement and explain the environmental and health challenges associated with nanomaterials.

2.2. The effect of nanosilica on cement properties

2.2.1. The effect on cement hydration

Many researchers have reported the effects of nanosilica particles on the hydration, microstructures and mechanical properties of cement compared to other properties. The accelerating capability of nanosilica on cement hydration has been established in different studies in which

the peak of hydration was noticeably growing (Bjornstorm et al., 2004; Kadri and Duval, 2009; Kim et al., 2014; Land and Stephan, 2012; Lin et al., 2008; Pang et al., 2009; Quercia et al., 2016; Santra et al., 2012; Thomas et al., 2009). Moreover, these authors argue that nanosilica is the best accelerator for cement hydration due to its pozzolanic reactivity. Its mechanism is mainly associated with the huge specific surface area of nanosilica, because it acts as nucleation sites for the precipitation of C-S-H gel (Pang et al., 2009; Quercia et al., 2016; Wang et al., 2017).

Likewise, Silvestre and Chithra et al. while examining the effects of nanosilica on cement properties, discovered that addition of silica particles into cement grains creates $H_2SiO_4^-$ that reacts with the available Ca^{2+} to form an excess C-S-H (Chithra et al., 2016; Silvestre (2015)). These C-S-H gel spread in the water between the cement grains and serve as seeds for the formation of a more compact C-S-H phase. They further demonstrated that nanosilica increases the consumption of Ca(OH)₂ and creates an additional C-S-H gel formation which is the main component for strength in hardened cement (Chithra et al., 2016; Silvestre, 2015). Other researchers concluded that the percentage of Ca(OH)₂ can be reduced by nanosilica and converted into C-S-H gel, thus, increasing the strength of cement (Bahadori and Hosseini, 2012; Gaitero et al., 2008, 2010; Qing et al., 2007).

The residual amount of Ca(OH)₂ was also examined by Byung et al. while observing the degree of pozzolanic reaction of nanosilica (Byung et al., 2007). In the test, amorphous nanosilica was reacted with Ca(OH)₂ from calcium silicate hydration. To fulfill the purpose of the study, four ratios of nanosilica 3, 6, 10 and 12% by weight of cement (bwoc) were blended with the cement. Based on the qualitative analysis at 7 days, the amount of Ca(OH)₂ in the hardened cement blended with 10% nanosilica was 4.06% while in the control sample, the remaining Ca(OH)₂ was 8.89%. After testing the mechanical strength of the same samples, the specimen mixed with 10% nanosilica showed higher strength, nearly three times greater than the control specimen. It was clearly concluded that the difference in the strength buildup between the two samples was attributed to the pozzolanic reaction of nanosilica. On the contrary, Bagheri et al. reported that nanosilica could promote cement hydration at the early stages, but slow down the cement hydration at later stages (Bagheri et al., 2013).

2.2.2. The effect on mechanical properties of cement

Controversies linger, on the role of nanosilica on the mechanical strength of cement. But numerous literature reported positive feedback about the enhancement of nanosilica on compressive strength of cement. The increase in strength could be attributed to two factors; the filling effect to promote the microstructure, and as an activator to accelerate the pozzolanic reactivity (Wang et al., 2017). Nanosilica was found more effective in improving compressive strength than silica fume (Byung et al., 2007; Qing et al., 2007). The blending of 10% nanosilica with dispersing agents was found to enhance the mechanical strength of hardened cement by 26% in 28 days compared to 15% of silica fume in the same curing time (Li et al., 2004). Similarly, it was found that the addition of small content of nanosilica (0.25%) promotes the mechanical strength. Published results of Sobolev et al. revealed an increase of compressive strength by 10% and flexural strength by 25% after curing the samples for 28 days (Sobolev et al., 2009). It was concluded that the presented results were influenced by the production route and the

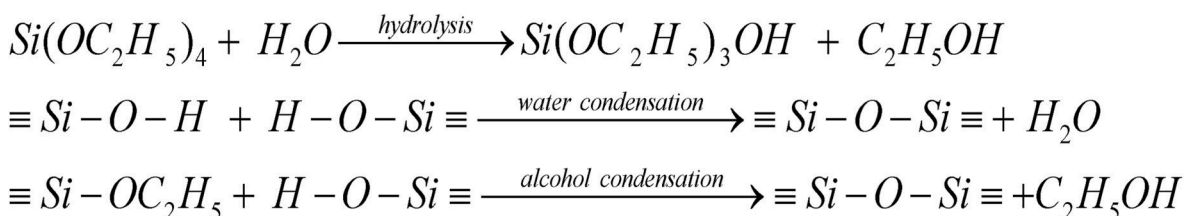


Fig. 8. The general reactions of TEOS that lead to the formation of nanosilica in the sol-gel practice (Rahman and Padavettan, 2012).

nature of synthesis of the nanosilica such as type of reaction media, length of the reaction for sol-gel method and dispersion of the nanosilica in the cement.

Additionally, Chithra et al. (2016) studied the effect of colloidal nanosilica with a particle size of 5–40 nm on the strength of cement. To achieve the objective of the experiment, Portland cement was replaced by nanosilica at 0.5, 1, 1.5, 2, 2.5 and 3% bwoc, and the samples cured for a period of 3, 7, 28, 56 and 90 days. The mechanical strength of all nanosilica blends was found to be higher than the control specimen. The strength of cement cubes increased with the percentage of nanosilica addition as shown in Fig. 9. Moreover, the strength gain is attributed to the method of production of nanosilica and the dispersion of nanoparticles in cement-based materials. The authors concluded that the key function of pozzolanic reaction is strength improvement and lessening the pore size distribution. The role of nanosilica in hydration and mechanical properties of oil-well cement (API Class H) was investigated by Pang et al. (2009) at a low temperature of 59 °F. Nanosilica with the tiniest dimension of 4–6 nm and those with high aspect proportions verified as the most efficient accelerator, with a record of 30 and 130% enhancement in cement mechanical strength for a curing time of 48 h and 7 days respectively. One of the interesting results from the previous studies indicated that cement stone containing nanosilica with a particle size of 10 nm (Li et al., 2004; Li et al., 2004b), displayed lesser strength enhancement compared to cement stone having nanosilica with particle dimension of 40 nm (Byung et al., 2007).

Another earlier report associated with the performance of silica composite coatings recommended that the particle size of nanosilica influenced the bonding strength and toughness of the silica composites (Yuan et al., 2005). However, still a few studies disputed the acceleration of nanosilica on pozzolanic reactions (Byung et al., 2007; Choolaei et al., 2012; Haruehansapong et al., 2014; Kim et al., 2014; Li, 2004; Said et al., 2012). Based on the experimental findings, Zyganitidis et al. demonstrated a decreasing effect of nanosilica on the compressive strength of hardened cement. This was due to the destroyed spatial configuration of cement hydration by the existing nanosilica particles (Zyganitidis et al., 2011). The variance of this investigation outcomes

could be caused by the change of the nanosilica type and dimension as well as the curing environment.

Currently, most studies focus on the modification of oil-well cement after adding nanosilica under normal curing temperature (20–80 °C). Until now, the studies about the role of nanosilica on cement properties under elevated temperatures are lacking. With the rising demand for oil and natural gas, petroleum companies are looking for new areas, as petroleum wells are drilled in a wider range of depths and temperatures than before. The physical and chemical properties of the cement vary significantly when subjected to high temperatures. The reports have shown that above 110 °C, C–S–H gel changes to a phase called dicalcium silicate hydrate (C₂–S–H), which regularly increases the permeability of the set cement resulting in the loss of strength (Nelson and Guillot, 2006; Wang et al., 2017). To prevent strength retrogression, the bulk lime to silica ratio in cement has to be reduced. This can be accomplished by adding 30–40% of silica-based materials to the cement, and this amount of silica is enough to consume large quantities of Ca(OH)₂ and decrease the Ca/Si ratio of C–S–H phase from 1.5 to 1.0 (Nelson and Guillot, 2006; Souza et al., 2012). Nanosilica has therefore been examined recently to determine if it can prevent oil-well cement from losing its strength under high temperatures. Wang et al. examined the development of mechanical strength of the cement stone containing 35% silica flour with 0, 4, and 6% nanosilica under 150 °C/65 MPa curing condition (Wang et al., 2017). Based on the results, it was concluded that the strength of hardened cement containing 35% silica flour improved by the addition of 4 and 6% at different curing periods. The average mechanical strength of the specimen containing only 35% from 1 day to 28 days was 52.38 MPa. When 4 and 6% nanosilica was added, the average values of strength improved to 63.86 and 64.16 MPa respectively. This indicates that the addition of 4 and 6% nanosilica can improve strength in cement containing 35% silica flour under high temperature.

In general, many researchers have mentioned the mechanical properties of cement as the most studied property compared to other properties. The key factors affecting the mechanical strength of hardened cement were reported as the surface area, the micropore volume, the replacement content and the average particle size of the nanoparticles

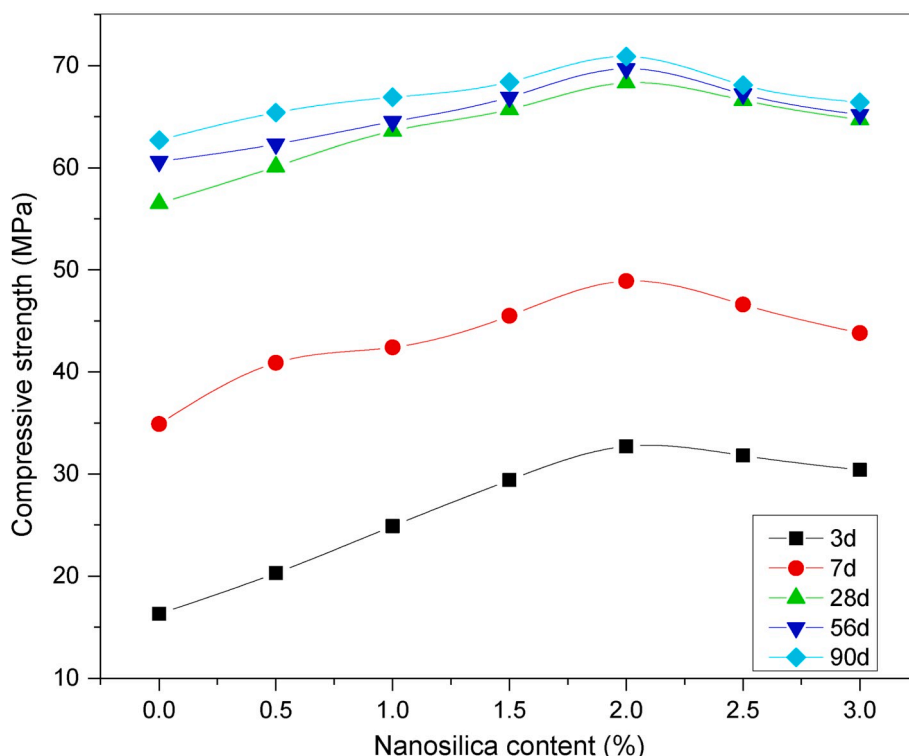


Fig. 9. Compressive strength of cement at various curing times. Source (Chithra et al., 2016).

(Biricik and Sarier, 2014).

2.2.3. The effect on the microstructure of hardened cement

Adding nanosilica into cement also affects the microstructure of the hardened paste. Effective means of dispersing silica nanoparticles in the cement mix was reported to form denser microstructure while promoting the cement hydration process (Silvestre, 2015). Hardened cement with well-distributed nanosilica has a compact microstructure even if nanosilica particles are replaced in cement in a lower ratio. But if nanosilica particles are not distributed accordingly, voids and weak regions may be formed (Li et al., 2004). Different material characterization techniques are being used to investigate the microstructure properties of the cement sheath. Qing et al. (2007), based on the SEM image, found that $\text{Ca}(\text{OH})_2$ formed during hydration remains in cement in a crystal structure which negatively affects cement properties. However, when nanosilica is added to cementitious material, crystallization of $\text{Ca}(\text{OH})_2$ becomes minimal while C-S-H gel formation is promoted, thus improving the strength of cement. An additional C-S-H gel fills the pores system causing denser and well-packed microstructure of cement. Dense and compacted microstructure with a reduced quantity of $\text{Ca}(\text{OH})_2$ crystals were found in samples containing nanosilica compared with the control specimens (Kontoleontos et al., 2012; Singh et al., 2011).

Likewise, El-Baky et al. (2013), investigated the effect of nanosilica with 19 nm particle size on the microstructure of the hardened cement. The content of nanosilica added into cement was 1, 3, 5, 7 and 10% bwoc. The study used the SEM image to compare the microstructure of silica-based cement and conventional cement. It was observed that cement paste containing nanosilica was found to be denser with a homogeneous microstructure. The nanosilica particles react with $\text{Ca}(\text{OH})_2$ to give more C-S-H gel, which fills the pores; promoting densification and modification of microstructure of cement (Bjornstorm et al., 2004; Byung et al., 2007; Ji, 2005; Singh et al., 2011; Tobon et al., 2012). This process decreases the number of pores and thus reducing water permeability of cement (Li, 2004; Mohamed et al., 2014; Ozyildirim and Zegetosky, 2010; Reddy and Meena, 2017; Thomas et al., 2009; Uchikawa and Uchida, 1980). In addition, the specimen containing 5% nanosilica displayed the greater crystals of C-S-H in the range of 1.2 μm , while the sample with 1% nanosilica revealed the C-S-H crystal size of about 600 nm (Stefanidou, 2012). Also, Bahadori and Hosseini examined the effects of supplanting cement with colloidal amorphous nanosilica on the microstructure and durability of concrete. SEM and EDAX tests showed an improved microstructure of concrete in the samples with 1%, 2% and 3% nanosilica as compared to control samples (Bahadori and Hosseini, 2012).

The enhanced microstructure of cement containing nanosilica was also reported by Wang et al. when investigating if nanosilica could prevent strength retrogression in oil-well cement under elevated temperatures. The SEM micrograph of the hardened cement comprising 35% silica flour and 6% nanosilica, indicated that the main products of hydration were C-S-H gels in the form of flake and needle. Besides, lots of nanosilica particles were observed among the C-S-H gels. And due to enormous surface area, these nanosilica particles agglomerated and created groups with a dimension of 50–200 nm (Wang et al., 2017). Pourjavadi et al. demonstrated that the addition of 1.0% nanosilica decreased the porosity of hardened cement due to pozzolanic reactivity and the production of additional quantities of C-S-H gels (Pourjavadi et al., 2012). They further reported that the microstructure was noticeably modified due to the filling effects of nanosilica and crystals of $\text{Ca}(\text{OH})_2$ were decreased in quantity and size as a result of the pozzolanic reaction. According to Ershadi et al. (2011), blending 1.0% of nanosilica bwoc to cement reduced porosity and permeability by 33% and 99% respectively. As observed using the SEM images, the nano-scale particles of nanosilica plugged the pores, resulting in reduced porosity and permeability. It was observed that these nanosilica particles are capable of filling the voids within the cement matrix resulting in a more compact

structure than that of the control.

2.2.4. The effect on fresh properties of cement

Apart from the cement hydration, microstructure and mechanical properties, studies have indicated that cement slurry properties such as setting time, rheology, fluid loss and others, can also be modified by the addition of nanosilica into cement. Senff et al. investigated the effect of amorphous nanosilica addition on fresh properties of cement pastes and mortars. The particles were incorporated in cement at the ratios of 1, 1.5, 2.0 and 2.5% bwoc. They reported that an initial and final setting time of cement pastes was reduced after incorporating nanosilica (Senff et al., 2009). It was further revealed that the consistency of the cement paste containing nanosilica increased with the dosage of nanosilica while setting time decreased with the increasing content of nanosilica.

Likewise, Ershadi et al. in their study on the effect of nanosilica on cement matrix permeability in oil-well, where the incorporation of 1% nanosilica bwoc reported reduced the thickening time from 8 h to 1.58 h (Ershadi et al., 2011). Similar findings were presented by Quercia et al. where the addition of 1.0% nanosilica bwoc decreased the thickening time of conventional Class G cement slurry by 2 h 31 min (Quercia et al., 2016). The blending of nanosilica into cement quickens the hydration; turn diminishing the setting time (Bjornstorm et al., 2004; Ltifi et al., 2011). As presented in Fig. 10, the shortening of setting times of cement is also due to huge specific surface area and pozzolanic activity of nanosilica (Chithra et al., 2016). In oil-well cementing the reduced setting time decreases the wait on cement time (WOC), hence saving the operational time and costs. To assure that cement pastes can be blended at the surface and injected into the well with lower pressure drop, rheological properties are put into consideration. Quercia et al. found that the addition of 1.0% nanosilica bwoc increased the specific surface area in the blend hence improving the yield point and plastic viscosity of Class G cement slurry after the initial blending; consequently enhancing the stability (Quercia et al., 2016). Similarly, the increase in yield point and plastic viscosity of Class G cement slurry were also revealed by Ershadi et al. using nanosilica with a particle size of 20 nm between 1.0 and 3.0% bwoc (Ershadi et al., 2011).

Besides, Berra et al. (2012) and Aggarwal et al. (2015), examined the rheology of cement slurries and noted that the blending of nanosilica into cement significantly rises the water requirement in the mix to maintain its workability. Such kind of property indicates that the incorporation of enormous surface area particles into cement results in the high demand for water to maintain the workability of the cement

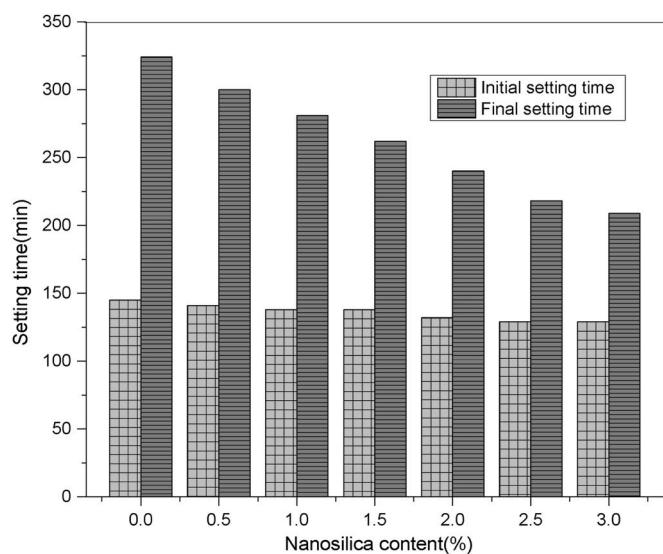


Fig. 10. Setting time of cement containing nanosilica. Source (Chithra et al., 2016).

mixture. Some authors reported that when the content of water in the mixture is maintained, increasing the content of nanosilica will promote the packing of the materials which positively decreases the free water (Aggarwal et al., 2015; Quercia et al., 2016; Senff et al., 2009, 2010; Wang et al., 2017). However, nanoparticles having enormous surface area can needlessly raise the viscosity at a required slurry density, resulting in pumping complications while promoting air content in the slurry (Santra et al., 2012; Senff et al., 2009). Therefore, accurate optimization is required to attain the most efficient porosity shrinkage and rheology.

One of the challenging problems during oil-well cementing is the loss of fluid from the cement slurry. The loss of fluid is detrimental since it can cause dehydration of the cement slurry before its normal setting time (Newlove et al., 1984). When this occurs it prevents the pumped cement from reaching its final position and leaving unwanted cement slurry in the casing becoming one of the primary reasons for cement failure in petroleum wells. Existing reports have posited that addition of nanosilica can control fluid loss in cement due to the effect increased surface area has on reactivity and through filling the nano-pores of the cement paste (Amanullah and Al-Tahini, 2009; El-Diasty and Ragab, 2013; Fakoya and Shah, 2017; Mohseni et al., 2015; Senff et al., 2009; Silvestre, 2015). It has been established that when nanomaterials such as nanosilica are used as liquid loss control agents, they can reduce fluid loss and resist gas migration (Roddy and Duncan, 2008). Moreover, Patil and Deshpande, investigated the effect of nanosilica on fluid loss in oil-well cement slurry using 6 nm nanosilica blended in Class H oil-well cement. The study disclosed that the addition of 0.2 gal/sk nanosilica reduced the loss of fluid as well as lowering the thickening time of cement (Patil and Deshpande, 2012). They evidently concluded that nanosilica can plug the openings between the cement grains, thus promoting the retainability of water. Similarly, the experimental results of Ershadi et al. (2011) revealed that nanosilica promotes compressive strength while reducing fluid loss and thickening time. In this study, spherical nanosilica powder with particle size of 20 nm blended with Class G cement at the ratios of 0.5, 1.0 and 2.0% bwoc revealed efficient properties as water loss control materials. It was suggested that ultra-fine particles of nanosilica are capable of filling the voids within the cement matrix resulting in immobilization of free water. Thus, the fluid in the cement slurry would be restricted from escaping the cement.

3. Evaluation of the current scenario

Nanotechnology has shown the capacity to transform the oil-well cement industry. Numerous research groups have confirmed the effectiveness of nanosilica technology in enhancing the performance of oil-

well cement for improved well productivity (Guo et al., 2017; Kong and Ohadi, 2010). Fig. 11 displays the modified properties of the oil-well cement containing nanosilica. The modification and enhancement of cement properties containing nanosilica are attributed to nanosize effect and high specific surface area of nano-powders that provide the potential for excessive chemical reactivity. The present study revealed that nanosilica has been the most studied oxide nanoparticles since it is more advantageous than other nanoparticles when incorporated into a cementitious system. Many researchers have concluded that the incorporation of nanosilica will impart a more uniform and dense microstructure in cementitious materials. Other findings have pointed out the modifications and enhancement in mechanical properties such as compressive strength of cement containing nanosilica. On the contrary, there are two significant challenges regarding the incorporation of nanosilica in cement paste. To attain even distribution of nanoparticles in the cement matrix remains a challenge. Studies have indicated that the dispersal of nanosilica particles influences the cement hydration, the fresh properties of the slurry and the hardened properties of cement. This helps to maintain the positive influence of nanoparticles into cement paste and thus to completely fill in the voids of the cement structure, resulting in a more compact and denser texture. The second challenge associated with the addition of nanosilica in cement is the reduction of fluidity, due to its higher surface area and enhanced water demand. It has been established that the use of dispersing agents of different types and concentrations along with the particle size and surface area of nanosilica, influences the rheological properties of the fresh cement paste (Biricik and Sarier, 2014). The most common finding discussed by many authors is the optimal capacity of nanoparticles to be used. Numerous authors have reported that the optimum content of nanoparticles to be used is still a challenge, requiring more scientific investigation. Some researchers have argued that the suitable nanosilica dosage in cement must be between 1 and 5% bwoc, to avoid agglomeration of the particles during blending of the slurry. Yet others have averred that the cement properties can also be enhanced with greater contents up to 10% (bwoc); if the nanoparticles are effectively distributed in the cement mix (Biricik and Sarier, 2014).

The effects of the nanosilica size particles and the optimum replacement content of nanosilica on the mechanical strength of cement were presented by Haruehansapong et al. They tested the compressive strength of cement mortars containing nanosilica with different particles sizes of 12, 20 and 40 nm. The obtained results revealed that nanosilica significantly enhanced the mechanical strength of the hardened cement and the increase in strength was similarly reliant on the size of nanosilica particles (Haruehansapong et al., 2014). They further demonstrated that cement specimens containing 40 nm, provided greater

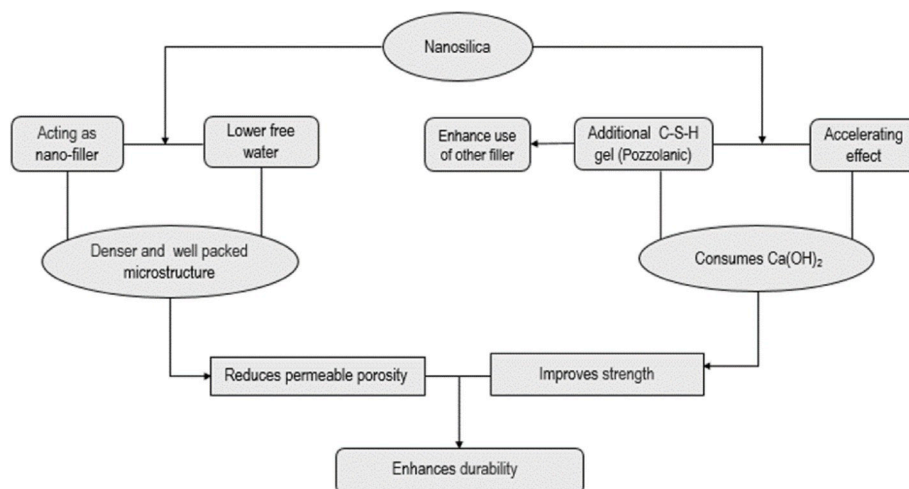


Fig. 11. Summary of the improved properties of cement containing nanosilica.

mechanical strength in comparison with 12 and 20 nm nanosilica due to their ineffective dispersion and agglomeration of nanosilica particles into cement paste. By changing the replacement ratios of 3, 6, 9 and 12% bwoc, [Haruehansapong et al. \(2014\)](#) found that the optimum amount was 9% for all sizes of nanosilica particles-signifying that the particle dimension affected only the mechanical strength of hardened cement while presenting no impact on the optimum content. However, other studies have shown that higher ratios of nanoparticles prevent uniform dispersion in the cement slurry; and that the optimum ratio of nanosilica cannot be fixed with certain percentages ([Singh et al., 2013](#)). They further stated that the optimum content of nanosilica depends on several factors such as the type of nanosilica (dry powder or colloidal). Previous studies revealed that at an early age, colloidal nanosilica in concrete produced more compressive strength than powder nanosilica, but in late stages both colloidal and powder nanosilica produce equal compressive strength ([Biricik and Sarier, 2014](#)). The influence of nanosilica types must be addressed in future research works.

[Bhuvaneshwari et al. \(2012\)](#), found that though many studies had reported the modification of cement properties after incorporation of nanoparticles such as nanosilica, the nature of reaction responsible for the improved properties of cement is not clear. Furthermore, the mechanism to describe the conceptual path between nanomaterials and their reaction in different phases of cement matrix such as C₃S, C₂S, C₃A and C₄AF has not been revealed yet. There is a continuous point of discussion concerning the speed of cement hydration with nanosilica inclusion; whether it is due to its enormous surface activity or its pozzolanic reactivity. Thus, further detailed investigations in this area are recommended. The published articles revealed that the cement industry is responsible for almost 6–7% of the man-made CO₂ produced in the world annually ([Biricik and Sarier, 2014](#)). Thus, there is a huge demand to lessen the amount of cement utilized for construction purposes. In that regard, nanosilica technology has been used to reduce the CO₂ emission associated with the cement industry. Preceding reports in the literature indicate that 1 kg of microsilica can reduce about 4 kg of cement and that the reduction could be higher if nanosilica is incorporated ([Singh et al., 2013](#)). In addition, despite the great advantages of silica nanoparticles in cementitious materials, problems related to lowering cost, environmental and health effects of using nanomaterials should be considered further.

4. Summary and conclusions

This review study focuses on the effect of silica nanoparticles on both the fresh and hardened properties of oil-well cement. The study shows that the inclusion of nanosilica into the cement has a high potential to improve a wide range of fundamental properties of cement to ensure adequate zonal isolation and extended well life. Most of the authors concluded that the incorporation of nanosilica particles into cement enhances mechanical properties and microstructure of hardened cement. In addition, nanosilica particles promote cement hydration which in turn decreases the setting time and increases the rate of compressive strength development. Early strength in oil-well cementing is important because it prevents the influx of formation fluids into the wellbore during cement setting.

Based on the review made, future studies should address the following issues;

1. The optimum nanosilica content to be added into cement needs to be determined, depending on the type and average size of nanomaterials. Currently, there is no formula describing the amount of nanoparticles to be added to oil-well cement, to make nanomaterials beneficial for the properties of cementitious composites.
2. Most of the studies done so far are focused on the influence of nanoparticles such as SiO₂, Fe₂O₃, TiO₂, Al₂O₃, nanoclay and carbon nanotubes to enhance cement properties separately. Researches on the influence of these nanoparticles to modify properties of cement in

a combination of two or more kinds of nanoparticles need to be investigated more.

3. The effective dispersion of nanosilica is a challenge requiring additional study. Nanoparticles are not easy to disperse uniformly into cement paste due to their high specific surface areas.
4. Many works have been done on mechanical properties and microstructural modification. Other durability properties regarding geochemical downhole conditions such as acid resistance, sulfate resistance and corrosion resistance need to be investigated further.
5. To date, many of the petroleum nanotechnology researches and publications are findings of the laboratory experiments. Thus, more field trial works are suggested for further development of nanotechnology in the oil and gas industry.
6. Studies show that a wide range of properties that make nanosilica so useful, such as size, shape and surface characteristics, can also cause potential problems if the material is not properly used. Hence, further research results are needed to clarify some environmental and health-related effects of nanosilica and other nanomaterials.

Declaration of competing interest

The researchers proclaim no contending interests between them.

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Nomenclature

API	American Petroleum Institute
bwoc	By weight of cement
C–S–H	Calcium silicate hydrate
C ₂ –S–H	Di-calcium silicate hydrate
C ₃ S	Tri-calcium silicate
C ₂ S	Di-calcium silicate
C ₃ A	Tri-calcium aluminate
C ₄ AF	Tetra-calcium aluminoferrite
CNF	Cellulose nano-fibres
CNT	Carbon nano-tube
GNP	Graphene nano-platelets
3D	Three dimensions
HDS	Hydro-desulphurization
HTHP	High temperature high pressure
MPa	Megapascal
MRI	Magnetic resonance imaging
OBM	Oil based mud
RHA	Rice husk ashes
RRS	Resonance raman spectroscopy
SEM	Scanning electron microscopy
SBRSA	Semi-burnt rice straw ashes
TEOS	Tetraethylorthosilicate
VES	Viscoelastic surfactant
WBM	Water based mud
WOC	Wait on cement
XRD	X-ray diffraction

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