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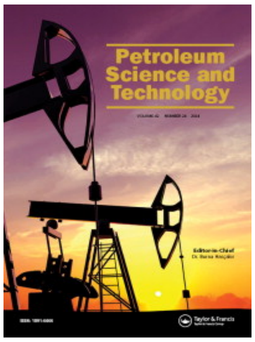

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Modeling the impact of pH and reservoir temperature on the dissolution of quartz mineral due to oilfield chemical treatment

Ebikapaye Peretomode[a], Blessing Eboibi[b], Abarasi Hart[c], and Mike Ajieh[b]

[a]School of Engineering, Robert Gordon University, Aberdeen, UK; [b]Department of Chemical Engineering, Faculty of Engineering, Delta State University, Oleh Campus, Igbide, Nigeria; [c]Department of Chemical and Biological Engineering, The University of Sheffield, Sheffield, UK

ABSTRACT

The petroleum industry frequently uses oilfield chemicals, such as corrosion inhibitors, scavengers, drilling fluid, and chemical enhanced oil recovery methods, to improve oil recovery. However, these chemicals can negatively impact the geochemical characteristics of the formation, potentially leading to facility failure and reservoir formation damage. A study using PHREEQC was conducted to predict the impact of reservoir temperature and pH on quartz rock dissolution under different temperatures and pH levels. The results showed consistent dissolving rates for quartz at different pH levels. The solubility of quartz minerals was slightly affected by the solution pH, suggesting that the process variable pH has little effect on the equilibrium rate. However, precipitation caused quartz to dissolve at a rate of 0.04 mmol.L^{-1}.s^{-1} at 45 °C but increased to 0.07 mmol.L^{-1}.s^{-1} at 65 °C in 0.8 s. As temperature increased, quartz and $NaNO_3$ solution interaction reached equilibrium concentrations in 0.8 and 2.7 s. The study concluded that temperature significantly impacts quartz dissolution during interaction with sodium nitrate oilfield chemicals compared to pH. The PHREEQ software is a useful research tool for simulating rock-fluid interactions resulting in reservoir formation dissolution and precipitation, which can cause formation damage and sand production.

KEYWORDS
Formation failure; mineral dissolution; mineral transport; oilfield chemicals; PHREEQC

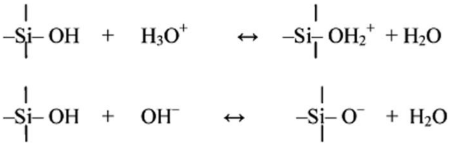
1. Introduction

In the petroleum industry, nitrates are usually employed to treat and manage the activities of sulfate-reducing bacteria, which can cause production equipment to corrode and produce H_2S (Hubert 2010; Voordouw et al. 2011).

CONTACT Ebikapaye Peretomode e.peretomode@rgu.ac.uk School of Engineering, Robert Gordon University, Aberdeen, AB10 7GJ, UK.

Consequently, the petrophysical and mineralogical changes in reservoir rocks are caused by the fluid-rock interaction process, which also releases dissolved minerals (Davidova et al. 2001: Peretomode, Oluyemi, and Faisal 2022a). On the other hand, redox and ion exchange processes, mineral dissolution, precipitation in the fluid, and/or the downstream movement of precipitates containing dissolved minerals are examples of chemical reactions resulting from the fluid-rock contact. However, they precipitate and proceed through ion exchange if the concentrations are higher than solubility (Chang and Civan 1991). The process of solute transport between bulk solutions and mineral surfaces, surface solute desorption and adsorption, ion dehydration and hydration, and solute movement on the surface occur simultaneously during mineral dissolution/precipitation (Appelo and Postma 2005: Peretomode, Oluyemi, and Faisal 2022b).

The distribution and alteration in the characteristics of the reaction sites can account for the dependence of the dissolution rate on pH (Nangia and Garrison 2008). The hydroxylic group of quartz reacts with water at room temperature to produce an ionized and neutral form, which is represented as follows:



$$-\underset{|}{\overset{|}{Si}}-OH + H_3O^+ \leftrightarrow -\underset{|}{\overset{|}{Si}}-OH_2^+ + H_2O$$

$$-\underset{|}{\overset{|}{Si}}-OH + OH^- \leftrightarrow -\underset{|}{\overset{|}{Si}}-O^- + H_2O$$

where the neutral, protonated, and deprotonated sites are, respectively, SiOH, $SiOH_2^+$, and SiO^- (Choi, Seo, and Chae 2013; Nangia and Garrison 2008). The existence of many sites suggests that there are several different reaction mechanisms taking place on the quartz sites during dissolution. The species distribution at quartz surface sites determines their reactivity, and this distribution varies with pH to impact the total rate of dissolution (Crundwell 2017; Nangia and Garrison 2008). The protonation state is changed by pH variation, which has an impact on the rate of reaction and the mechanism of dissolution.

When chemical injection occurs in a permeable media, precipitation of rock minerals may result in a decrease in permeability. Precipitates and mineral migration are known to clog pore throats depending on particle size, resulting in a decrease in both porosity and permeability (Wu and Sharma 2016).

Mineral dissolution and precipitation caused by chemical-rock interaction in oilfields can be induced by fluid flow, heat transfer, chemical reactions, and mineral transport. The impact of chemical-rock resulting in dissolution and precipitation can be explored experimentally and through numerical models (Barclay and Worden 2000; Fathi 2018; Oelkers et al. 2009; Martin, Manzano, and Dolado 2019). Several experimental studies have been reported on the impact of fluid-rock interaction on rock characteristics. For instance, Behera and Sangwai (2022) experimentally studied the viability of employing a hybrid approach (silica-based nanofluids, anionic surfactant, and polymer) for oil recovery due to the difficulties of extracting oil from an established reservoir with a poor recovery factor. They found that chemical adsorption was responsible for wettability alteration and increased oil recovery. However, for the pore-scale oil displacement phenomenon, some studies utilize a microfluidic chip integrated with an inverted microscope to investigate the role of nanoparticle present in the injected fluid. It was found that the interaction of nanoparticles at the fluid-fluid interface resulted in a drop in interfacial tension. This resulted in an increase in oil displacement as the concentration of nanoparticles increased, and a little decrease at concentrations $>1000\,\text{ppm}$ (Behera, Kumar, and Sangwai 2022). This suggests that the precipitation of nanoparticles as a result of chemical-rock interaction could contribute toward interfacial tension reduction at oil–water interface.

Zhang et al. (2017) looked into the flagstaff Hill chlorate's kinetic dissolution at 100 °C and pH 3.0 and 7.5. The aim of this was to ascertain the degree of undersaturation and the relationship of the chlorite dissolution rate on pH under geologic carbon storage conditions. It was discovered that in severely undersaturated fluids, the chlorite dissolution behaves as though it were near equilibrium. The effect of chemicals ($CaCl_2$, NaCl, KCl, Na_2SO_4, and $NaHCO_3$)-rocks (quartz, K-feldspar, albite, calcite, granite, tuff, and mud) interactions on rock characteristics was studied by Yang et al. (2017). The study explored how chemical injection affected porosity and permeability changes following their interaction with the formation. As a result of the mineral dissolution, it was discovered that the porosity increased by 29%. More recently, Costa et al. (2020) employed coreflooding experimental technique to study oil recovery mechanisms, as well as the effect of pH in salinity solutions, utilizing clastic rocks. The authors observed that more oil is recovered as the pH moves toward 8 as a result of ionic exchange. However, this observed phenomenon was experimentally studied. Thus, these studies indicate that the chemical-rock interaction changes the properties of reservoir rock.

Using numerical and analytical models, several authors (Borazjani et al. 2019; Dang et al. 2013), have investigated the interaction of low salinity water with reservoir rocks. Due to the complexity of geochemical reactions in porous media, fluid-rock interaction has not been adequately accounted for in these models. Owing to this complexity, fluid-rock interactions may now be effectively modeled using geochemical modeling techniques. With PHREEQC, geochemical modeling of the fluid-rock interaction allows for the prediction of these mineral dissolution and precipitation processes. For geochemical processes, some researchers have combined PHREEQC with additional simulators, such as reservoir simulator CMG (Computer Modeling Group) and UTCHEM (3-D chemical flooding simulator) (Chandrasekhar, Sharma, and Mohanty 2018; Shabani and Zivar 2020; Sharma and Mohanty 2018).

This study explores the use of a geochemical simulation based on the phreeqc.dat database, to model the role of pH (2, 7.5, and 8.5) and average reservoir temperatures (45 °C and 65 °C) in the interaction between nitrate and quartz. This study will help to improve the predictability of quartz behavior in a reservoir rock under varying pH and reservoir temperature conditions. However, this study solely takes into account chemical reactions involving dissolution and precipitation. The integrity of reservoirs and the deformation of rocks are significantly influenced by the dissolution of quartz. Thus, changes in the subsurface porous rock's pore volume and structure are caused by the dissolution and precipitation of minerals following interaction with injected chemicals. For this reason, understanding quartz dissolution because of chemical-mineral interaction is vital to chemically enhanced oil recovery processes and chemical treatment of production facility for corrosion-causing microorganisms.

2. Materials and methods

Using PHREEQC, reactive flow model in porous media and its governing equations are presented in this section to predict the pH (2, 7.5, 8.5) and temperature (45 °C and 65 °C) effects on quartz minerals (dissolution and precipitation) (Parkhurst and Appelo 1999; Appelo, Parkhurst, and Post 2014; Osinowo et al. 2018; Parkhurst and Appelo 2013).

The simulation tool, PHREEQC 3.7, used in this study is an openly accessible inverse geochemical modeling software developed by Parkhurst and Appelo (1999). The software is able to compute complex interactions between aqueous solutions, dissolved gases, mineral assemblages in batches, and one-dimension diffusion-advection transport computations with reversible and irreversible reactions, speciation and saturation index, and kinetically controlled reactions (Al Shalabi and Sepehrnoori 2017). The PHREEQC code computes the saturation index (SI) for minerals and evaluates if a solution has the possibility to precipitate or dissolute a mineral. The saturation indices (SI) are determined by Eq. (1).

$$\mathrm{SI} = \log(\mathrm{IAP}/\mathrm{KT}) \tag{1}$$

where KT is the mineral's solubility product at a specific temperature and IAP is its ion activity product.

A one-dimension model of a cylindrical core (Figure 1) with a geometry of 10 m long and 5 m wide consisting of an inlet face, wall body, and no

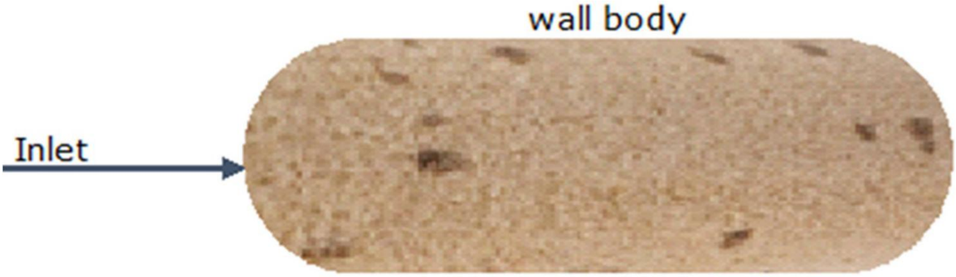


Figure 1. 1D cylindrical core sample used for experiment.

Table 1. Showing input model parameters for the media.

Parameters	Values
Initial surface area (A_o), m^2	22.6
Vol. of solution, m^3	0.162
Initial moles of solid (m_o)	103
Mole, m	1

Table 2. Showing model parameters of the fluid.

Parameters	Values
$NaNO_3$, mol	10
Water, kg	1

outlet face or outlet flow, which mimics "shut-in," is used. The shut-in is to mimic a situation where the oil well is plugged to prevent production for the purpose of maintenance.

The simulation was set at an ambient temperature (i.e., 25 °C). Pure water is defined with solution input and the pure phase assemblage is defined with "equilibrium phases." Nitrate ($NaNO_3$) is added irreversibly to an equilibrium phase (Table 1) of sandstone rock. The parameters of the fluid and medium used in this study are presented in Tables 1 and 2.

The kinetic approach, which explains the reaction's time dependence, is the method utilized for the geochemical modeling. As a result, it can provide data on when a chemical (nitrate) is injected and when the reaction reaches its equilibrium state.

Regarding mineral assemblage and reaction occurrence, the PHREEQC geochemical model makes some suppositions or assumptions. The two most significant of these hypotheses are (a) solids and their compositions precipitate out of solution as secondary minerals and (b) pH is controlled by both internal and external variables (e.g., interactions between surrounding minerals and reservoir rock). These presumptions seem to be important for cation concentration control (Appelo and Postma 2005).

According to Parkhurst and Appelo (2013), PHREEQC uses the mass action equations for aqueous species to estimate the reaction. However, equilibrium is assumed to exist for all ionic strengths. The chemical mass balance equation and the rate law are used to simulate the chemical-rock interaction. Equation (2) depicts the chemical dissolution reaction that occurs between rock (quartzose-SiO_2) and sodium nitrate ($NaNO_3$) solution.

$$2NaNO_3 + SiO_2 \rightarrow Na_2O_3Si + N_2O_5 \tag{2}$$

Equation (3) is utilized to simulate the precipitation and dissolution of formation minerals due to chemical-rock interaction.

$$r_{qu} = k_{qu}\left(1 - \frac{IAP}{K}\right) * \frac{A_0}{V}\left(\frac{m}{m_0}\right)^n \tag{3}$$

The PHREEQC database's compilation of reaction constants is applicable to temperatures between 0 and 300 °C. The pH values selected are based on differing publications (Brady and Walther 1990; Knauss and Wolery 1988; Wollast and Chou 1986). Regarding the impact of pH on quartz dissolving, the fluid-rock interaction is represented as a sequence of exponential equilibria that are computed at every *k*th time step. According to the study, this computation yields an equilibrium composition of aqueous solution and quantities of freshly dissolved and precipitated minerals (Mironenko and Zolotov 2012). With a composition of nitrate and pure water, a batch-reaction model was run. Equation (4) presents rate formulas for the quartz and dissolution processes that incorporate the saturation state, as reported by Saaltink, Ayora, and Carrera (1998), both expressions characterize the reaction kinetics for both processes.

$$r_{qu} = k_{qu}\left(1 - \frac{IAP}{K}\right) \tag{4}$$

At ambient temperature of 25 °C, the solubility product is denoted by K ($g.L^{-1}$), the ionic activity product is represented by IAP ($mol.L^{-1}$), and the saturation state is represented by $\Omega = IAP/K$ given k_{qu} 10–13.7 in $mol.m^{-2}.s^{-1}$. When $IAP/K < 0.1$, the term $(1 - IAP/K)$ has no effect, and the kinetics are linear in time in that domain.

3. Results and discussion

To study the impact of the interaction between quartz rock and sodium nitrate at different pH and temperature levels, sets of simulation results were run. This study is predicated on the assumption that silica, one of the prominent minerals in the mineral assemblage of sandstone rock (quartzose), was chosen to mimic the interaction between rock and the chemical species. As a result, the modeling concentrates on it, with the intention of eventually including other prominent minerals for more research, such as feldspar and lithic.

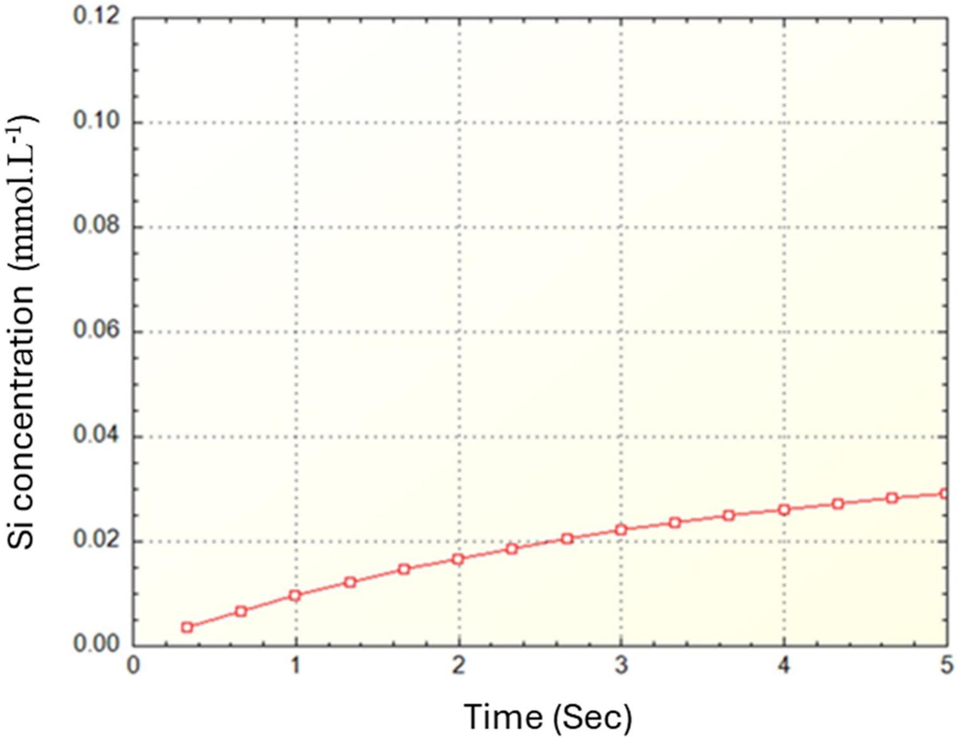


Figure 2. The plot of concentration of Si as a function of time.

The rock dissolution profile caused by nitrate injection over time is shown in Figure 2. It is evident that the concentration of dissolved silica rises dramatically with time. As the equilibrium concentration gets closer, the early increase in Na_2O_3Si concentration decreases. On the other hand, this is noted in the fourth year (measured in seconds), at which point the Si concentration stops being affected by time. Once the saturation indices (SI) begin to approach zero, the equilibrium state is achieved.

Figure 3 illustrates the dissolving of quartz rock as a result of interaction with a sodium nitrate solvent at pH values between 2 and 8.5, fixed at 25 °C over a period of time (s). The selected pH values aligned with the findings documented in previous studies on the impact of pH on quartz dissolving (Ali et al. 2019; Brady and Walther 1990; Knauss and Wolery 1988; Wollast and Chou 1986).

The dissolution trends at 0.002, 0.001, and 0.001 $mmol.L^{-1}.s^{-1}$, respectively, are identical for the three pH values 2, 7.5, and 8.5 that are being studied. Up to 8 s into the dissolution process, the rate increases quickly before starting to slow down when ions begin to precipitate out of the solution. This is consistent with earlier study reported in the literature (Martin, Manzano, and Dolado 2019). Between 8 and 16 s, there is a continuous competitive rate of dissolution and precipitation. After that, the

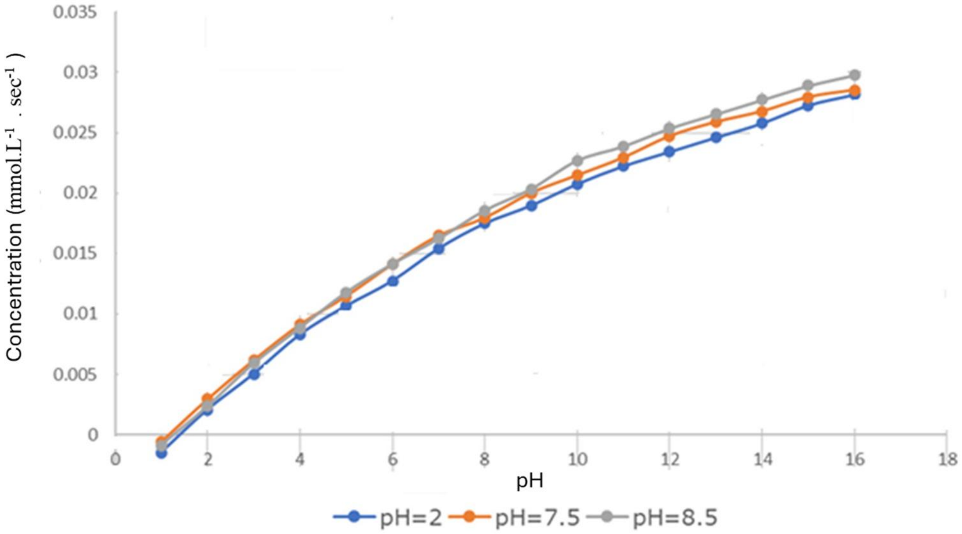


Figure 3. The effect of pH on quartz rock dissolution rate.

concentration-time plot starts to plateau, and the rates of precipitation and dissolution start to approach equilibrium.

Additionally, at pH 2 and pH 7.5, respectively, the concentration-time trend plateaus at roughly 0.03 mmol.L^{-1}. On the other hand, the concentration-time equilibrium is around 0.035 mmol.L^{-1} at pH 8.5. This indicates that the pH value increased from 2 to 8.5, or 6.5 points, due to the dissolution rate. This discovery is in line with literature (Brady and Walther 1990; Knauss and Wolery 1988). It is observed that raising the pH led to an increase in quartz dissolving, especially when the pH was dropped below 2 or raised above roughly 5 (Kamiya, Ozaki, and Imahashi 1974). Figure 3 shows no discernible variation in the rate of dissolution when the pH rises by <6, as in the situations of 2 and 7.5. The solubility of quartz mineral rises as the pH of the solution rises by 6 points, even though the effect of pH on the rate at which equilibrium is attained is negligible when the increase is smaller than 6 (Figure 3). For the disintegration of quartz, Henderson, Syers, and Jackson (1970) and Lasaga (1981) have reported a similar pattern. As a result, when the system pH rises above five points, the solubility of the quartz crystal increases noticeably.

The findings indicate that pH has very little effect on quartz's interaction with the chemical $NaNO_3$. The results show that the dissolution rate due to quartz-sodium nitrate interaction is almost the same for strong acidic pH (2), near neutral pH (7.5), and mild alkaline pH (8.5). Thus, it can be concluded that the dissolution rate due to quartz-sodium nitrate interaction is independent of pH within the region extending from acidic through neutral to mild alkaline solutions. However, reports have it that strong alkali pH solution generally increases quartz dissolution, especially for hydroxides of

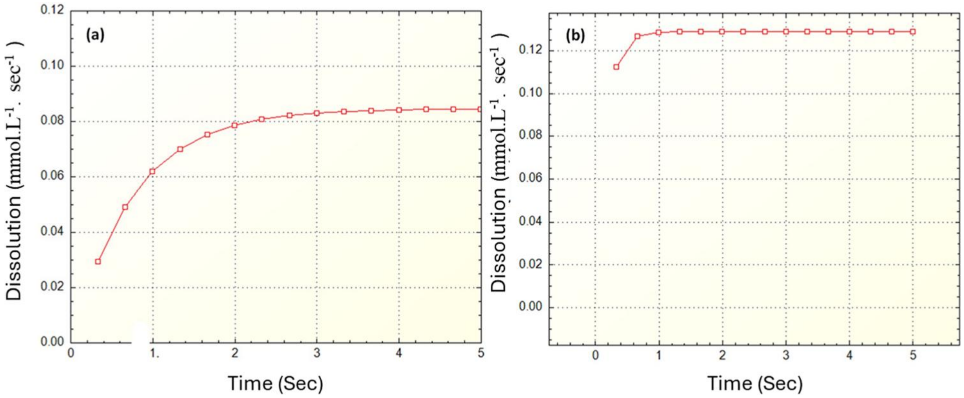


Figure 4. Effect of average reservoir temperature on sodium nitrate-quartz interaction: (a) temperature 45 °C and (b) temperature 65 °C.

monovalent ions like K^+ and Na^+ due to their solvent characteristics (Ali et al. 2019; Knauss and Wolery 1988; Wilson 2020). It is based on this reason that the rate quartz dissolves when it interacts with sodium nitrate follows a similar trend in the region of pH investigated. Hence, quartz often dissolves at different rates depending on the chemical makeup of the aqueous solution. The dissolution occurs at the mineral-solution interface. The aqueous sodium nitrate may adsorb to a silica surface and react with the Si–O–Si bonds on the surface within the pH range investigation. It is the breaking of the Si–O bonds in the surface siloxane groups that induces dissolution of the quartz.

The impact of temperature on quartz rock dissolving resulting from chemical-rock interaction at pH 7 is depicted in Figures 4a,b over a period of time (s). Osinowo et al. (2018) have reported on a comparable investigation. The rate of disintegration at 0.04 mmol.L^{-1}.s^{-1} increases quickly, up to 1 s, at 45 °C. Due to precipitation, it drops between 1 and 2 s and then equilibrates as time grows over 2 s. On the other hand, at 65 °C the dissolution rate rises quickly to 0.07 mmol.L^{-1}.s^{-1} in 0.8 s, slows down in 0.8 and 1 s, and then reaches a plateau in 1 s.

At 65 °C, the system tends toward equilibrium more quickly than it does at 45 °C (Figures 4a,b). It is apparent from comparing the concentration-time graphs that the temperature effect has a major effect on the solubility of silica. With a solubility of around 0.13–0.82 mmol.L^{-1} at 45 °C, quartz is therefore more soluble in sodium nitrate at 65 °C. While quartz in sodium nitrate at 45 °C reached a concentration of 0.072 mmol.L^{-1} SiO_2 and took 2.7 s to equilibrate, quartz at 65 °C reached a concentration of 0.93 mmol.L^{-1} SiO_2 and took 0.8 s to equilibrate. This can be explained by the fact that when the system temperature rises, there are more molecules colliding with one another. This speeds up the dissolution while slowing down precipitation reactions. Moreover, mass movement between quartz

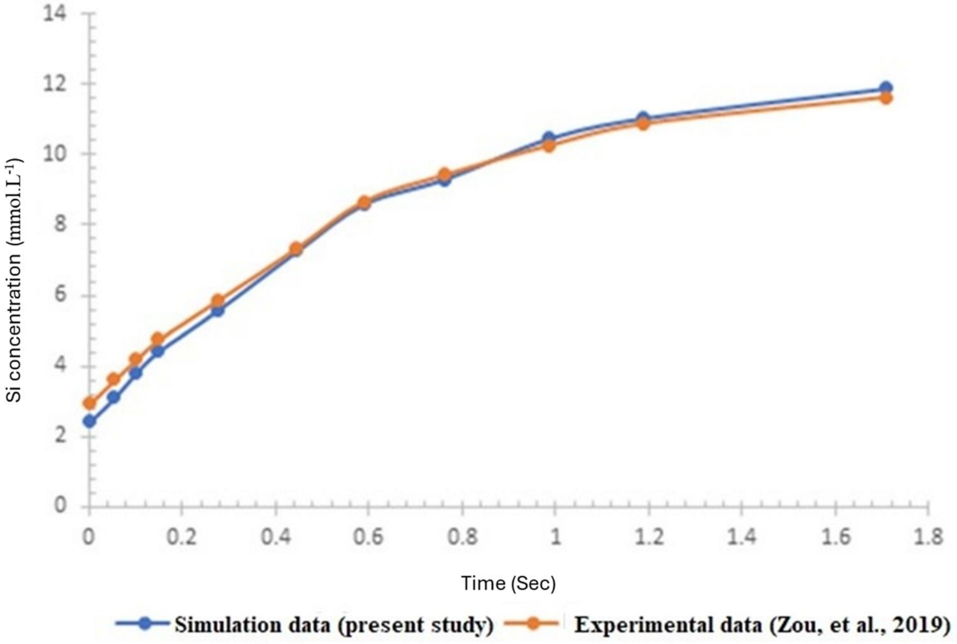


Figure 5. Si concentration vs. time plot of simulation and experimental data at ambient temperature and pH 7.

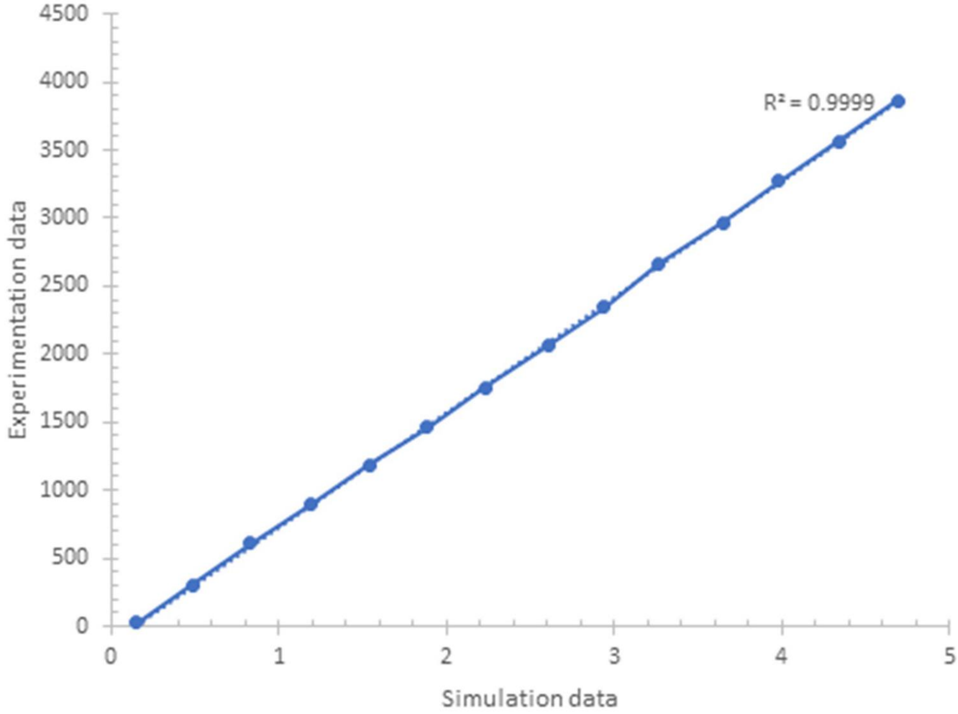


Figure 6. Parity plot for coefficient of determination between experimental and simulation data.

and nitrate solutions is made easier at higher temperatures when $NaNO_3$'s viscosity decreases. The rate of the quartz dissolution reaction is accelerated by this increase in mass transfer. These findings demonstrate that temperature, pH, time, and the chemical makeup of the fluid all affect how quickly quartz dissolves.

The published experimental test dataset from Zhou, Liu, and Tan (2019) at ambient temperature has been compared to the results from the simulations to ensure the results are credible. Figure 5 shows a concentration vs. time plot at ambient temperature and pH 7. The simulation yielded values for the dissolution of quartz with regard to the predetermined time intervals. The anticipated outcomes have a strong correlation with the experimental data. This is displayed as a determination plot (Figure 6) to display the variance between the experimental and simulated findings and demonstrate their match. The plot (Figure 6) shows that the simulation provided a very good description of the experimental data, with a coefficient of determination R^2 of 99.94%.

4. Conclusion

In the petroleum sector, production facility corrosion-causing microbes have been treated and managed with oilfield chemicals. The interactions between various oilfield chemicals and the formation influence formation rock characteristics. PHREEQC has not been considered to evaluate the chemical effects of sodium nitrate in treating reservoir rock. Hence, this work has created a geochemical model using PHREEQC to simulate the effects of sodium nitrate oilfield chemical on the dissolution and precipitation of quartz rock under different pH and reservoir temperature conditions. Kinetically regulated reactions are essential to the modeling approach. The homogeneity of the medium has been assumed when modeling the reaction using the proposed equations. The results demonstrated how minerals reacted with sodium nitrate, dissolving and precipitating them. The results suggest that changes in temperature and pH have an impact on how quickly chemicals dissolve and precipitate. Furthermore, the geochemical model, which has a 99.94% match-accuracy success rate, has been validated using experimental data.

This present study sheds light on the influence of pH and reservoir temperature on quartz minerals dissolution. It is anticipated that reservoir characterization of the phenomenon can be achieved in due course and at minimal cost. The simulation result will be beneficial in saving a great deal of money and time when it comes to the costly laboratory equipment required for validation studies.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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ORCID

Abarasi Hart http://orcid.org/0000-0002-4433-5887
Mike Ajieh http://orcid.org/0000-0003-4571-1634

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Glossary of terms

- Deprotonated: This is the process of removing a proton to an atom, molecule, or ion.
- Equilibrium: When a reversible reaction is occurring, this condition means that there is no net change in the amounts of reactants and products.
- Hydroxylic group: This is a functional group with one atom each of oxygen and hydrogen has been discovered.
- Ion dehydration: This is how much energy an ion needs to give up molecules.
- PHREEQC: Or the pH Redox Equilibrium is a computer application used to perform geochemical calculations.
- Protonated: This is the process of creating an acid conjugate by adding a proton to an atom, molecule, or ion.
- Solute transport: This is the movement of chemicals in a porous media.
- Solubility: This is the state of being soluble.
- Wettability: This is the state of being wettable.
- Salinity: This is the amount of salt dissolved in water.
- Saturation index: This is the computation of the mineral product's dissolved ions' chemical activity using their solubility product.
- Saturation state: At this point, the relative humidity approaches unity.