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ORIGINAL ARTICLE

Influence of ionic composition in aqueous solution on wettability of rock surface-experiment and economics evaluation



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KEYWORDS

Minerals; Wettability; Monovalent ion; Divalent ions; Dynamics contact angles **Abstract** In this study, the influence of different ionic composition in aqueous solution on the minerals surface wettability was studied. The differences effect of monovalent ion and divalent ions onto the wettability alteration were studied. The anions were Cl⁻ and SO₄²⁻. The SO₄²⁻ could make the minerals surface more hydrophilic. Besides, the influence of NaCl, MgCl₂, CaCl₂, Na₂SO₄, K₂SO₄ and MgSO₄ on the mineral wettability alteration were studied. The results indicated that divalent ions showed significant impact on the minerals wettability alteration, compared with monovalent ion, and divalent ions have higher effect on compressing the electric double layer. The static contact angle and dynamic contact angle were measured. Different heavy oils were studied, including heavy oil with 100 ppm, heavy oil, heavy oil without resins, heavy oil without asphaltenes. The results showed that the asphaltenes would make it difficult for the heavy oils to liberate from minerals, thus decreasing the oil drops contact angle. Then the resins would decrease the heavy oil contact angles. CaCl₂/MgCl₂ and K₂SO₄ have synergistic effect on the change of the minerals surface wettability. Atomic force microscope (AFM) measurement indicated that the ions would

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effectively decrease the interaction force on the surface of heavy oil-minerals, which was beneficial to the heavy oil liberation. The roughness measurement indicated that the different ions would effectively increase the minerals surface wettability.

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1. Introduction

Wettability was the basic property of the surface, which influenced the interface interactions, which was widely used in the emulsions formation (Hou et al., 2022; Jia et al., 2022), oil-water separation (Li et al., 2022; Sun et al., 2022; Oiu et al., 2018), enhanced oil recovery (Zhao et al., 2019), ect. Many researchers focused on the salinity effect on minerals surface wettability, and the detailed research progress was shown as follows. Besides, molecular dynamic simulation was used to study the molecule movement during the enhanced oil recovery process, and the salinity effect on oil recovery were studied (Badizad et al., 2020, Badizad et al., 2020, Badizad et al., 2020, Koleini et al., 2020, Badizad et al., 2021, Koleini et al., 2021, Sun et al., 2021). In recent years, many researchers focused on the salinity effect on rocks surface wettability (Liu et al., 2007, Gandomkar and Rahimpour 2017, Boumedjane et al., 2019, Tetteh et al., 2020, Kasha et al., 2021). Different salinities have different effect on the wettability alteration (Mugele et al., 2015, Alroudhan et al., 2016).

The formation brine would be helpful to the oil recovery. Altering the injected brine would alter the corresponding operation fees. Besides, the salinity alteration would be helpful to the oil recovery increase during the enhanced oil recovery process. Besides, due to the fact that the LSW methods was ion-engineered brine, and the brine composition would influence the heavy oil recovery.

Meriem Boumedjane et al. studied the ions Mg^{2+}/SO_4^{2-} and Ca^{2+}/SO_4^{2-} effect on the oil-wet calcite surfaces wettability alteration (Boumedjane et al., 2019). The results indicated that the determining ions (PDs) would alter the surface wettability, and the surface would be more hydrophilic. When the Mg^{2+} amount was equal to the SO_4^{2-} , the surface wettability would show the highest. Asghar Gandomkar et al. (Boumedjane et al., 2019) studied the monovalent and divalent ions on oil/low salinity brine/ limestone wettability alteration. The results indicated that the divalent ions (Ca^{2+} , Mg^{2+} , SO_4^{2-}) had the similar wettability alteration on the limestone surface. Besides, the asphaltenes acid crude oil had positive effect on the wettability alteration for carbonate rocks. Mg^{2+}/Ca^{2+} have synergistic effect with SO_4^{2-} on the wettability alteration.

In addition, many researchers used the DFT and MD simulation of oil-rock system to study the wettability alteration mechanism (Li et al., 2021). Huifang Li et al. (Li et al., 2020) used the molecular dynamics and first-principles simulation methods to study the interactions between deionized water and brines, and the results indicated that two distinct water adsorption layers are formed by hydrogen bonding, and different ions played different role in the water-calcite (1 0 1 4) interaction. In addition, other researches indicated that the functionalization is no prerequisite of structural ordering and a distinct mass density profile perpendicular to the interface (Li et al., 2021). The corresponding rock surface wettability would influence the oil recovery, and the molecular dynamics simulation would be helpful to study the oil recovery mechanism (Singh et al., 2019).

Therefore, the main purpose of this study was to: (i) to study the monovalent and divalent ions effect on original rock surface wettability; (ii) to study the monovalent and divalent ions effect on the oil-wet rock surface wettability; (iii) to study the oil-solid interaction force and zeta potential of different monovalent and divalent ions.

2. Materials and discussion

2.1. Materials

NaCl, KCl, MgCl₂, MgSO₄, Na₂SO₄, CaCl₂, purity > 99 %, were purchased from Aladdin Cop, Shanghai. The lipophilic carbonate asphalt rocks were obtained from Buton, Indonesia. The SNL 10 AFM probe employed was obtained from Brucker, Karlsruhe, Germany. Limestones were procured from Tianjin Liufang Co. Itd., Tianjin, China. The heavy oil was extracted from the Indonesian oil sands, Buton, Indonesia.

2.2. Contact angles measurement

In this study, the contact angles were used to evaluate the wettability of the minerals surface. The static contact angle and dynamic contact angle were measured.

2.3. Oil-solid interaction force measurement

The oil–solid interaction force was determined employing AFM measurements (Brucker, Karlsruhe, Germany). This determination is critical, as the oil-solid separation becomes more challenging when the oil–solid interaction forces increase (Liu et al., 2005, Xiang et al., 2019). During the measurements, the repulsive force would increase when the probe is close to the solid surface; however, when the probe was significantly removed from the solid surface, the force became an attractive force (Ren et al., 2009, Hogshead et al., 2011). This adhesive force was subsequently employed to evaluate the interaction force between the heavy oils and the minerals. The results indicated that the liberation of the heavy oil from the mineral surface became straightforward when less adhesive forces were present (Liu et al., 2003).

Bitumen was obtained from carbonate asphalt rocks through Soxhlet extraction. The bitumen (0.5 g) was subsequently dispersed into a 100 mL toluene solution to produce a 0.5 wt% bitumen–toluene solution, which was further diluted to 0.01 wt%. Next, the SiO₂ microspheres were washed with acetone, ethanol, and distilled water, and the SiO₂ microspheres were hydrophilic. Then, the SiO₂ microspheres were placed into the vacuum desiccator, and there was 0.5 mL octyl trimethoxy silane in the bottom of the vacuum desiccator. In order to create the vacuum environment, the pump was used to vacuumize for 3 min. The then the SiO₂ microspheres were in toe vacuum desiccator for 24 h for vapor deposition. The octyl trimethoxy silane would graft onto the hydrophilic SiO₂ surface, and the SiO₂ surface became hydrophobic (see Fig. 1).

The hydrophobic SiO_2 microspheres produced were subsequently washed with toluene and dried under nitrogen before they were added to the previously prepared 0.01 wt% bitumen–toluene solution for a period of 4 h. Finally, the solution



Fig. 1 The study outline.

was transferred drop-wise onto a glass slide, at which point the hydrophobic SiO_2 microspheres were fixed onto the glass slide. The calcite surface could then be cleaned with acetone, ethanol, and distilled water, before a double-sided adhesive was attached to the calcite surface.

The interaction forces between heavy oil and CaCO₃ surface in different saline solutions were measured using AFM (Bruker, Multimode 8) at room conditions. The speed of the probe was 2 μ m/s, and the interaction forces were measured in 100 different points in order to obtain the average value, and the average value was the interaction force.

2.4. Zeta potential measurement

The charges associated with bitumen and CaCO₃ in different saline water were investigated through an evaluation of the zeta potential (Liu et al., 2002, Ding et al., 2006). Here, the bitumen was dispersed in different ionic liquids employing the ultrasonic method. First, 0.5 g of bitumen was placed into 200 mL of saline solution, which was subsequently sonicated for 30 min. Then the bitumen zeta potentials would be measured. Next, 0.5 g of CaCO₃ would be dispersed into 200 mL saline solution and sonicated for 30 min. All measurements were carried out three times, the average value was the zeta potential value.

2.5. Roughness measurement

In order to explore the different ions effect onto the limestone surface roughness, the roughness measurement was conducted. The detailed experiment procedures were as follows. Firstly, the limestones were treated by different aqueous solutions (with different ions), and then the limestones were naturally dried. Then the AFM measurement (Brucker, Karlsruhe, Germany) was used measure the roughness of the limestone.

3. Results and Discussion.

3.1. Monovalent anion (Cl⁻) effect on minerals wettability

3.1.1. Static contact angles measurement

Fig. 2 showed the different NaCl, KCl, CaCl₂, MgCl₂ effect on the limestone surface wettability alteration. As was shown in Fig. 2, when the salinity increased, the contact angle would decrease. However, when the salinity increased to 10000 ppm, the contact angle remained stable. In other words, the contact angle would not decrease when the salinity was higher than 10000 ppm. When NaCl, KCl, CaCl₂ and MgCl₂ concentrations were the same, the MgCl₂ could make the contact angle show the lowest value, and the NaCl could make the contact angle show the highest value. The reason was due to the fact that divalent ions have more effect onto the minerals surface and compress the electric double layer (Lee et al., 2015, Adapa and Malani 2018).

3.1.2. Dynamic contact angles measurement

Fig. 3 showed the dynamics contact angle of different systems in different ions and different heavy oils contents. The equilibration time for different systems were different. When the heavy oils were original heavy oil + 100 ppm asphaltenes, the equilibrium time was 72 h, which was higher than other heavy oil systems. When the heavy oils were original heavy oil, heavy oil without resins, heavy oil without asphaltenes, the equilibrium time was 48 h, 24 h, 24 h, respectively. The results showed that the lower asphaltenes content would help decrease the equilibrium time, which was due to the fact that



Fig. 2 The contact angle of water drops onto the limestone surface under different ions concentration.

asphaltenes would enhance the heavy oil-solid interaction force (Qao et al., 2018, Hou et al., 2021). Besides, when the heavy oil was the same, the contact angle followed the rule that $\theta_{NaCl} > \theta_{KCl} > \theta_{CaCl2} > \theta_{MgCl2}$. The results indicated that the divalent ions would make the contact angle lower, and the divalent ions have higher effect on the heavy oil liberation.

Fig. 4 showed the contact angles of different oil drops onto limestone rock surface under different ions concentration (96 h). Fig. 4 showed that when the heavy oil was the same, the contact angle followed the rule that $\theta_{MgCl2} > \theta_{CaCl2} > \theta_{KCl} > \theta_{NaCl}$. When the ions were the same, the different heavy oil showed different contact angles, and when the heavy oil was without asphaltenes, the contact angle showed the highest value. And the heavy oil with 100 ppm asphaltenes would make the contact angle show the lowest value. For instance, when different heavy oil was dispersed into the NaCl solution, when the heavy oils were original heavy oil + 100 ppm asphaltenes, original heavy oil, heavy oil without resins and heavy oil without asphaltenes, the contact angles would be 135.1°, 143.4°,152.7°, 158.2°, respectively. For KCl, the contact angles would be 141.6°, 147.2°, 159.4°,



Fig. 3 The dynamics contact angle of different systems under NaCl, KCl, $CaCl_2$, $MgCl_2$ solution (a) original heavy oil + 100 ppm asphaltenes; (b)original heavy oil; (c) heavy oil without resins; (d)heavy oil without asphaltenes.



Fig. 4 The contact angle of oil drops onto the limestone surface under different ions concentration (96 h).

166.7°, respectively. For CaCl₂, the contact angles would be 153.3°, 161.2°, 164.1°, 165.2°, respectively. For MgCl₂, the contact angles would be 157.8°, 164.5°, 170.8°, 172.1°, respectively. The results indicated that different heavy oils would cause the different contact angles.

3.2. Monovalent anion (SO_4^{2-}) effect on minerals wettability

3.2.1. Static contact angles measurement

Fig. 5 showed the contact angle of water drops onto the limestone surface under different ions concentration (SO_4^{-}). For instance, when the different ions were treated the limestone surface, the contact angles followed the rule that $\theta_{Na2SO4} > \theta_{K2SO4} > \theta_{MgSO4}$. The contact angles decreased with the increase of ions concentration, which was due to the fact that the Na₂SO₄, K₂SO₄ and MgSO₄ would adsorb onto the limestone surface, and compress electric double layer, and then the limestone surface wettability alteration.

3.2.2. Dynamic contact angles measurement

Fig. 6 showed the dynamics contact angle of different systems under Na₂SO₄, K₂SO₄, MgSO₄ solution. Fig. 6 indicated that the contact angles would increase after the time continued. When the time was the same, the contact angle followed the rule that $\theta_{MgSO4} > \theta_{K2SO4} > \theta_{Na2SO4}$. Besides, different heavy oil phases showed different effect on the contact angle alteration. When the asphaltenes content decreased, the heavy oil liberation would be enhanced, which indicated that the contact angles would decrease.

Fig. 7 showed the contact angle of oil drops onto the limestone surface under different ions concentration (SO_4^2). The results showed that different sulfate ions showed different effect on the contact angles alteration. For Na₂SO₄, the contact angles for original heavy oil + 100 ppm asphaltenes, original heavy oil, heavy oil without resins, heavy oil without asphaltenes were 144.2°, 154.1°, 158.6°, 171.2°, respectively. For K₂SO₄, the contact angles were 154.2°, 160.3°, 161.5°, 173.1°, respectively. For MgSO₄, the contact angles were 166.3°, 162.1°, 166.2°, 174.6°, respectively.



Fig. 5 The contact angle of water drops onto the limestone surface under different ions concentration (SO_4^{2-}) .

3.3. The synergistic effect between Ca^{2+}/Mg^{2+} and SO_4^{2-}

Fig. 8 showed the synergistic effect water drops of CaCl₂, MgCl₂ and K₂SO₄ solution. Fig. 8 indicated that the contact angles would decrease when the ions concentration increased, and when the ions concentration reached to 10000 ppm, the contact angles would become stable. The contact angles would decrease when the ions concentration increased because the ions adsorption would increase and the ions would compress the electric double layer, which would be beneficial to the contact angles decrease. After the contact angles reached equilibrium, when the ions concentrations were the same, the contact angles followed the rule that $\theta_{CaCl2} > \theta_{K2SO4} > \theta_{MgCl2} > \theta_{2CaCl2+K2SO4} > \theta_{2MgCl2+K2SO4} > \theta_{5CaCl2+K2SO4} > \theta_{5MgCl2+K2SO4}$. All the results indicated that the CaCl₂/MgCl₂ has synergistic effect with the K₂SO₄ on the limestone wettability alteration.

Fig. 9. The dynamics contact angle of different systems under different ions and different heavy oil phase. Fig. 9 showed that for different heavy oils, there exist the synergistic effect between $CaCl_2/MgCl_2$ and K_2SO4 , in other words, the $2CaCl_2$ and K_2SO_4 would make the contact angles higher than other aqueous phases.

Fig. 10 showed the synergistic effect oil drops of CaCl₂, MgCl₂ and K₂SO₄ solution. Fig. 10 indicated that there was synergistic effect on the contact angles alteration for the CaCl₂/MgCl₂ with the K₂SO₄. For instance, when the heavy oils were original heavy oil + 100 ppm asphaltenes, original heavy oil, heavy oil without resins, heavy oil without asphaltenes, the CaCl₂ was used as the water phase, the contact angles would be 153.3°, 161.2°, 164.1°, 165.2°, respectively. When K₂SO₄ was used as the water phase, the contact angles would be 154.2°, 160.3°, 161.5°, 173.1°, respectively. When the 2CaCl₂ was combined with the K₂SO₄, the contact angles would be 162.4°, 168.3°, 172.7°, 172.1°, respectively. The results indicated that the CaCl₂ and K₂SO₄ have synergistic effect on the contact angles alteration.

Fig. 11 The contact angle of water drop onto the limestone surface under different ions concentration.



Fig. 6 The dynamics contact angle of different systems under Na_2SO_4 , K_2SO_4 , $MgSO_4$ solution (a) original heavy oil + 100 ppm asphaltenes; (b) original heavy oil; (c) without resins; (d) without asphaltenes.



Fig. 7 The contact angle of oil drops onto the limestone surface under different ions concentration (SO_4^2) .



Fig. 8 The synergistic effect water drops of $CaCl_2$, $MgCl_2$ and K_2SO_4 solution.



Fig. 9 The dynamics contact angle of different systems under Na_2SO_4 , K_2SO_4 , $MgSO_4$ solution (a) original heavy oil + 100 ppm asphaltenes; (b)original heavy oil; (c) without resins; (d)without asphaltenes.



Fig. 10 The synergistic effect oil drops of $CaCl_2$, $MgCl_2$ and K_2SO_4 solution.

3.4. Oil-solid interaction force

Fig. 12 indicated that the interaction force between heavy oil and minerals surface in different aqueous solution showed different value. When the heavy oil and mineral surface were in the deionized water, the interaction force showed the highest value (5.27 mN/m). When the heavy oil and minerals surface were dipped into the salt solution, the heavy oil interaction force would decrease. When the anions were Cl-, the interaction force between heavy oil and minerals surface was higher than that for SO_4^{2-} . For instance, the NaCl, KCl, CaCl₂ and MgCl₂ could decrease the oil-solid interaction force to 4.55 mN/m, 4.21 mN/m, 3.62 mN/m, 3.37 mN/m, respectively. The Na₂SO₄, K₂SO₄ and MgSO₄ could decrease the contact angle to 4.12 mN/m, 3.89 mN/m, 2.75 mN/m, respectively. Besides, the AFM results indicated that when chloride salt and sulfate were combined together, the oil-solid interaction force would decrease further. When 5 MgCl₂ was combined with K₂SO₄, the interaction force between heavy oil and minerals surface was 0.85 mN/m, which was the lowest value for different other salinity. The reason was due to the fact that different ions could make the interaction force decrease.



Fig. 11 The contact angle of water drop onto the limestone surface under different ions concentration.



Fig. 12 The interaction force between heavy oil and minerals surface in different aqueous solution.



Fig. 13 The roughness alteration of the limestone surface under different saline ions.



Fig. 14 The price (Yuan/ton) of different chemicals.

3.5. Roughness analysis

Fig. 13 showed the roughness alteration of the limestone surface under different saline ions. Fig. 13 showed that different salinities would make the limestone surface showed different roughness, which was due to the fact that different ions would cause different effect on the roughness alteration. The limestone roughness was 8.2 nm when there were no salt ions. However, when the salt ions increase, the roughness would increase. For instance, when NaCl, KCl, CaCl₂ and MgCl₂ were treated the limestone surface, the limestone surface roughness was 15.4 nm, 20.7 nm, 32.1 nm, 36.4 nm, respectively. When the Na₂SO₄, K₂SO₄ and MgSO₄ were treated the limestone surface, the limestone surface roughness was 24.5 nm, 29.8 nm, 36.6 nm, respectively. The results showed that when the cations were the same, the SO_4^{2-} could make the limestone surface roughness increase. Besides, when different ions combined, the roughness would increase. Fig. 13 indicated that when the ions were combined together, the roughness would increase, the reason was due to the fact. Besides, higher roughness would make the minerals surface more hydrophilic, and the contact angle would decrease (Faibish et al., 2002, Gao et al., 2015).

3.6. Economics evaluation

Fig. 14 showed the price (Yuan/ton) of different chemicals. The results showed that the heavy oil price was 4000 yuan/ton. But the ions concentration was ppm level, so it was economical feasible. In addition, the MgCl₂ and Na₂SO₄ prices were low (450 yuan/ton, 500 yuan/ton), but the MgCl2 and Na2SO4 showed high effect on the wettability alteration, so these salts showed potential application effect.

4. Conclusions

In this study, the different ions effect on minerals surface wettability alteration effect were studied, and the detailed conclusions were as follows.

- (1) The ions would enhance the minerals surface hydrophilicity, and the minerals surfaces would be enhanced.
- (2) The divalent ions have higher effect onto the minerals surface than monovalent ion, which was due to the fact that divalent ions have higher effect on adsorption to the minerals surface and compress the electric double layer.
- (3) The dynamic contact angles measurement indicated that the asphaltenes would make the heavy oil contact angles lower, and then was the resins.
- (4) CaCl₂/MgCl₂ and Na₂SO₄/K₂SO₄ have synergistic effect on the minerals wettability alteration.
- (5) The AFM measurement indicated that the ions would decrease the heavy oil-solid interaction force. The roughness measurement indicated that the ions could increase the minerals surface roughness.

5. Fund

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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