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Article

Experimental Investigation of the Effects of Drilling Fluid Activity on the Hydration Behavior of Shale Reservoirs in Northwestern Hunan, China

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Abstract: The interaction between drilling fluid and shale has a significant impact on wellbore stability during shale oil and gas drilling operations. This paper investigates the effects of the drilling fluid activity on the surface and osmotic hydration characteristics of shale. Experiments were conducted to measure the influence of drilling fluid activity on surface wettability by monitoring the evolution of fluid–shale contact angles. The relationship between drilling fluid activity and shale swelling ratio was determined to investigate the osmotic hydration behavior. The results indicate that, with increasing drilling fluid activity, the fluid–shale contact angles gradually increase—the higher the activity, the faster the adsorption rate; and the stronger the inhibition ability, the weaker the surface hydration action. The surface adsorption rate of the shale with a KCl drilling fluid was found to be the highest. Regarding the osmotic hydration action on the shale, the negative extreme swelling ratio (b) of the shale was found to be: $b_{KCl} < b_{CTAB} < b_{SDBS}$. Moreover, based on the relationship between the shale swelling ratio and drilling fluid activity, shale hydration can be divided into complete dehydration, weak dehydration, surface hydration, and osmotic hydration, which contributes to the choice of drilling fluids to improve wellbore stability.

Keywords: shale reservoirs; surface hydration; osmotic hydration; negative extreme swelling ratio; safety levels of activity

1. Introduction

Wellbore instability is a worldwide challenge in the oil drilling industry [1], causing direct economic losses of more than 1 billion USD annually [2,3]. It has been estimated that about 75% of drilled strata are shale formations and that 90% of wellbore instability occurs in such formations [3–5]. Shale is a sedimentary rock with distinct laminated layers and a high clay content [5,6]. When drilling into a shale formation, the shale comes into contact with the water-based drilling fluid. Thereby, the water molecules in the drilling fluid are adsorbed by the inter-layer of the clay minerals and onto the surface of the clay particles by coordination, electrostatic interaction, and hydrogen bonding;

and surface hydration occurs [3,7]. Additionally, due to the presence of micro-cracks in the shale, free water can easily enter the interior of the rock and induce a series of physico-chemical and mechanical changes [8]. The mechanical changes promote the expansion and enlargement of the original micro-cracks inside the shale, and then macro-cracks form, providing channels for the water molecules to enter the shale, which increases the contact area between fluid and clay particles and consequently leads to ion hydration and osmotic hydration [9]. Shale hydration is responsible for the attenuation of inter-particle interaction and cementation, which result in a decrease in the compressive strength and hardness of the rock [3,10]. Al-Bazali et al. [11] confirmed that diffusion osmosis has a detrimental effect on the mechanical stability of shale by reducing its compressive strength. As the degree of hydration increases, the rock structure is further destroyed, causing the wellbore to expand and fall off, resulting in wellbore instability. Van Oort et al. [12] found that shale–fluid interactions could be controlled to decrease shale hydration, which can enhance wellbore stabilization.

The factors that cause shale hydration are complex: pressure differentials, chemical potential differences [12], the crack structure of shale, and the type and content of clay mineral [13]. Additionally, the difference between the activity of the drilling fluid and the activity of shale formation has been proven to be a crucial factor to affect shale hydration [14]. Al-Bazali et al. [15] found that the membrane efficiency decided by the difference in activity between the drilling fluid and the shale formation was related to the types of cation and anion in water-based fluids, which affect shale hydration. Chenevert et al. [16] proposed the equilibrium activity theory of drilling fluids, and found that the higher the water activity in an oil-based drilling fluid, the more severe the shale hydration. The authors concluded the following: (1) when the shale formation activity is less than the drilling fluid activity, the water molecules in the drilling fluid can flow into the shale formation. Thus, surface hydration or osmotic hydration of the shale formation can be induced, which results in shale inflation; (2) when the shale formation activity is greater than the drilling fluid activity, fluid in the shale formation migrates into the drilling fluid. Thus, dehydration occurs, which induces shale contraction; and (3) when the activity of the drilling fluid is the same as that of the shale, there is no water exchange between the drilling fluid and the shale formation, and the rock remains in its original state [13,17,18]. Therefore, investigating the impact of drilling fluid activity on shale hydration behaviors has practical significance for improving mining efficiency and wellbore stability.

Shale hydration is divided into three stages: surface hydration, ion hydration, and osmotic hydration [19]. In previous studies, only the surface hydration of shale was considered as an important factor related to the surface wettability of shale, and was mainly related with fluid adsorption characterized by the change in the contact angle of the fluid on the surface of the shale; the faster the fluid adsorption rate, the faster the rate of change of the contact angle. Meanwhile, osmotic hydration was considered to be a factor inducing the swelling of shale, and was usually characterized by the swelling ratio of shale [20,21]. Huang et al. [19] found that a surfactant compound of polyamine (PA) and twelve alkyl two hydroxyethyl amine oxide (THAO) could enhance the surface hydrophobicity after adsorption onto the surface of shale particles and could restrain osmotic hydration through swapping out inorganic cations in the clay inter-layer. Yue et al. [5] found that the absorption of the cetyltrimethylammonium bromide (CTAB) cation on the surface of shale could increase the contact angle between the drilling fluid and the shale and inhibit the surface hydration of shale. Cai et al. [22] found that a composite surfactant could effectively change the wettability performance of water-based drilling fluid; this allowed the stability of the shale to be enhanced by controlling the wettability of the drilling fluid. Aghil Moslemizadeh et al. [23] reported the effect of silica nanoparticles (NPs) as a physical sealing agent on water invasion into the Kazhdumi shale; the authors showed that the use of the NPs reduced shale hydration and improved the wellbore stability. Liu et al. [24] concluded

that when shale comes into contact with water-based drilling fluid, surface hydration occurs on the surface of the shale clay particles, thus forming a surface hydration film, generating hydration stress, destroying the original mechanical balance, and eventually causing the shale strength to decrease. Using shale osmotic hydration experiments based on the generalized Usher model, Wen et al. [17] established a model related to the activity, swelling ratio, and hydration degree of shale, and classified shale hydration into three stages: dehydration, surface hydration, and osmotic hydration. However, in the above studies, surface hydration and osmotic hydration were not comprehensively considered, nor were they in studies aimed at investigating the relationship between the difference in the activity of the drilling fluid and the shale and shale hydration. Moreover, the relationship between the safety level of fluid activity and shale hydration has not been established. Determining this relationship could allow the impact of fluid activity on shale hydration to be characterized.

The present work takes advantage of the fact that, since the radius of K^+ is similar to that of the hexagonal cavity in the clay crystal unit, CTA^+ can be embedded into the inter-layer space of montmorillonite and SO_3^- can be adsorbed by polar substances on the surface of shale, which can prevent shale hydration. Therefore, potassium chloride (KCl), sodium dodecylbenzene sulfonate (SDBS), and CTAB drilling fluid systems, as typical inorganic and organic drilling fluids, are widely adopted to maintain wellbore stability [25–27]. Thus, in this study, three drilling fluid systems with different activities were selected as test fluids to investigate the effects of the activity of drilling fluids on the surface hydration and osmotic hydration of shale using tests of the contact angle between the drilling fluid and shale and swelling ratio tests. Furthermore, an evolution mechanism of fluid–shale contact angles as a function of fluid activity was proposed. Then, the safety of drilling fluid activity was classified into four levels based on the relationship between the shale swelling ratio and fluid activity.

2. Materials and Methods

2.1. Materials

In this study, the cationic surfactant used was CTAB, the anionic surfactant used was SDBS, and the inorganic salt used was KCl. These materials were obtained from a chemical company in Tianjin, China. The composition of the KCl, SDBS, and CTAB drilling fluid systems is shown in Table 1.

Table 1. Compositions of drilling fluid systems.

Based Fluid Formula	Inhibitor Concentration
0.35% CMC + 0.08% PH	0.5%~2.5% KCl
	0.5%~2.5% SDBS
	0.5%~2.5% CTAB

Note: CMC is carboxymethylcellulose sodium; PH is potassium humate.

The shale samples used in this study (Figure 1a) were collected from the Longshan shale outcrop (Figure 1b). In order to minimize the influence of shale anisotropy on the experimental results, the shale samples were taken from the same outcrop and drilled perpendicular to the bedding direction. The samples were sanded into 15 shale samples with a diameter of $\phi 50$ mm and thickness of 5 mm. Then, the samples were dried and kept at 40 °C for 24 hours in an incubator for fluid–shale contact angle tests. Furthermore, 45 shale samples with a diameter of $\phi 25$ mm and a thickness of 20 mm were prepared for swelling ratio tests. The compositions (Table 2) of the samples indicate that a large amount of brittle minerals and montmorillonite are present in this formation, with montmorillonite comprising 20~27%. The physical and mechanical parameters of the samples are shown in Table 3.

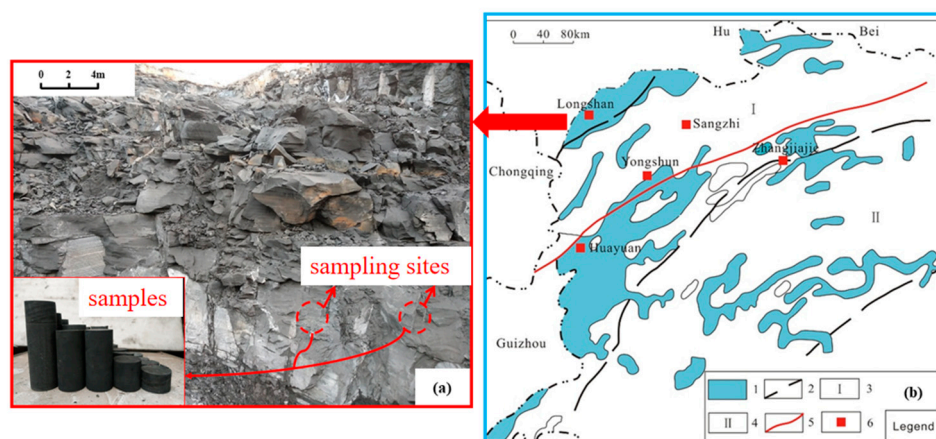


Figure 1. A map of the regional structure and shale outcrop of the Lower Cambrian Niutitang Formation in northwestern Hunan Province, modified after Cao et al., 2018 [28]. 1: Outcrop distribution area; 2: Stratigraphic boundary; 3: Northwestern Hunan area; 4: Xuefeng District, central Hunan; 5: Deep fault; 6: Sampling sites.

Table 2. Contents of mineral components in shale samples.

Quartz (%)	Feldspar (%)	Clay Mineral (%)	Anatase (%)	MICA (%)	Amorphous (%)
21~30	10~15	20~27	5~8	4~9	21~29

Table 3. Physical and mechanical parameters of shale samples.

Elastic Modulus (GPa)	Poisson's Ratio	Brittleness Index	Porosity (%)	Cohesion (MPa)	Internal Friction Angle (°)
1.25~3.86	0.19~0.24	0.20~0.37	2.24~4.47	0.57~0.98	33.82~41.70

2.2. Methods

In this study, tests were conducted for drilling fluid activity, fluid–shale contact angle, and shale swelling ratio.

2.2.1. Fluid Activity Tests

Place 10 ml of the prepared drilling fluid in a glass container, place it in the test box of a calibrated AW-1 water activity meter (Wuxi Huake Instrument and Meter Company, Wuxi, China; measurement range: 0~0.999; precision: 0.01), and then start the test at 23 °C. When the vapor pressure of the sample is the same as that of the surrounding air, the water vapor pressure in the gas space is taken as the value of the sample vapor pressure. Then, the water activity of the drilling fluid is calculated based on the relationship between the water activity and the vapor pressure.

2.2.2. Fluid–Shale Contact Angle Tests

The prepared shale sample was placed on the sample stage of a JCY contact angle meter (Shanghai Fangrui Instrument Co., Ltd., Shanghai, China; contact angle resolution of 0.01°). Then, 5 µl of drilling fluid was dripped onto the shale sample using a micro-syringe. The experimental process was recorded using a MV-1300UM-MIN high-speed camera (Weishi Digital Image Technology Co., Ltd., Beijing, China) with a frame length of 10 ms. Finally, the contact angle of the drilling fluid on the shale surface was obtained using the CONTA3.0 contact angle analysis software (Shanghai Fangrui Instrument Co., Ltd., Shanghai, China). To reduce experimental error, the contact angle of the drilling fluid was measured at three different locations on the shale surface and the average value was taken as the final measurement result.

2.2.3. Shale Swelling Ratio Tests

A displacement sensor (Si Mingwei Technology Co., Ltd., Shenzhen, China) was attached to the prepared shale sample, and the sample was then hung on a pylon. Then, 100 mL of drilling fluid was placed in a liquid cup of a CPZ-II swelling meter (Qingdao Jiaonan Analytical Instrument Factory, Qingdao, China) and the shale sample was immersed in the liquid for eight hours. The swelling meter records the amount of shale swelling displacement in real time. In order to improve the accuracy of the experimental results, three shale samples were used in three replicate experiments for each drilling fluid, and the average swelling ratio was taken as the final measurement result.

3. Results and Discussion

3.1. Effect of Inhibitor Concentration on Drilling Fluid Activity

The relationship between the inhibitor concentration and drilling fluid activity is shown in Table 4.

Table 4. The relationship between inhibitor concentration and drilling fluid activity.

Inhibitor Concentration		0.5%	1.0%	1.5%	2.0%	2.5%
KCl		0.860	0.871	0.900	0.933	0.957
SDBS	drilling fluid activities	0.904	0.922	0.930	0.935	0.946
CTAB		0.911	0.928	0.952	0.970	0.988

As the concentration of inhibitors in the drilling fluid system increases, the activity of the drilling fluid shows an increasing trend. At the same concentration, the CTAB drilling fluid has the highest activity and the KCl drilling fluid has the lowest activity. This means that at the same concentration, the difference in activity between the shale and the drilling fluid is greatest for the KCl drilling fluid, so when the shale interacts with the drilling fluid, the driving force pushing the water molecules in the drilling fluid into the shale is greatest.

3.2. Effect of Drilling Fluid Activity on Surface Hydration of Shale

The relationship between the drilling fluid activity and the fluid–shale contact angle is shown in Figures 2–4.

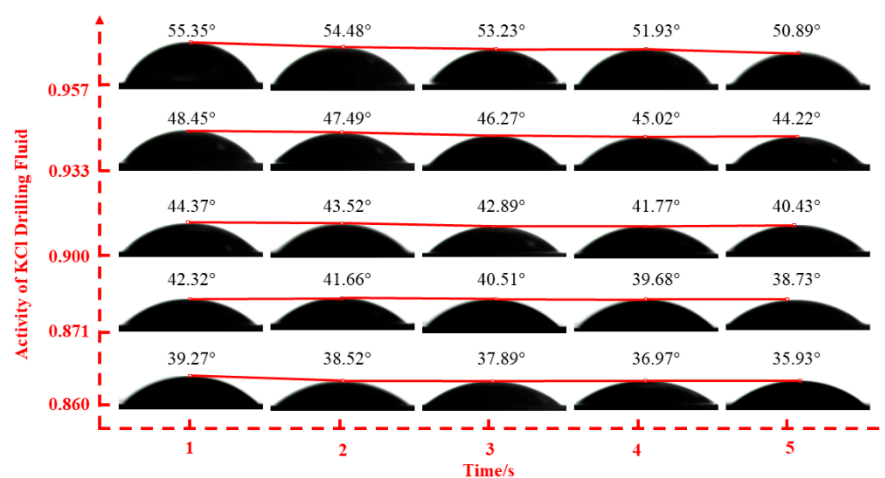


Figure 2. The evolution of fluid–shale contact angles with different activities of KCl drilling fluid.

As shown in Figure 2, as the activity of the drilling fluid increases, the fluid–shale contact angles tend to increase overall. This is due to the fact that the adsorption energy of exchangeable cationic K^+

on the surface of montmorillonite, which has a negative charge, is stronger than the adsorption energy of water molecules on the surface of montmorillonite. Additionally, the exchangeable K^+ is adsorbed on the surface of montmorillonite, forming an ionic film, which greatly improves the hydrophobicity of the surface of shale, which shows strong inhibitory ability of surface hydration. Furthermore, the rate of change of the fluid–shale contact angles decreases as the drilling fluid activity increases, which indicates that with increasing drilling fluid activity, the migration and adsorption rate of K^+ in the fluid increases and the number of layers of ionic film also increases, which can form a dense ionic film that prevents water from entering the shale. This means that the inhibitory effect of the drilling fluid on shale surface hydration is significantly improved when using KCl drilling fluid.

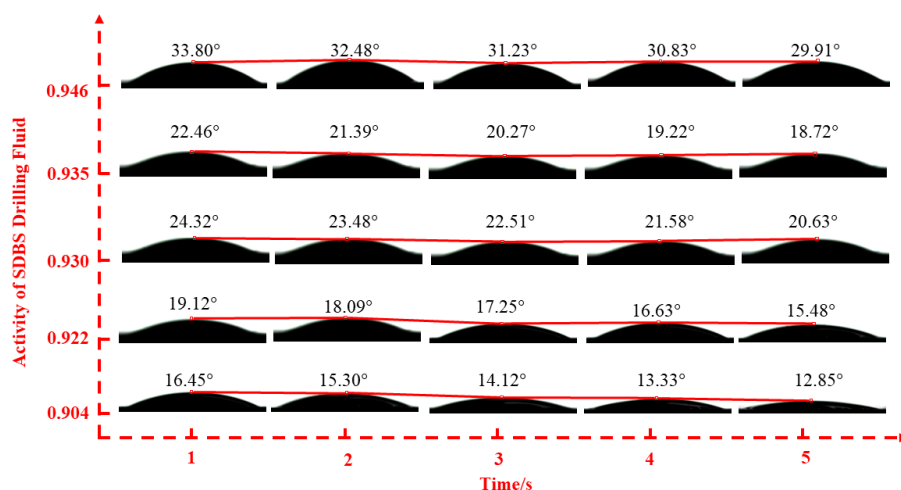


Figure 3. The evolution of fluid–shale contact angles for different activities of sodium dodecylbenzene sulfonate (SDBS) drilling fluid.

The evolution of the fluid–shale contact angle with increasing drilling fluid activity for SDBS drilling fluid is shown in Figure 3. The hydrophilic groups of SDBS molecules can form molecular films on the surface of shale through the van der Waals force due to the large amount of hydrophilic minerals (such as quartz, mica, and montmorillonite) that are present in shale, and which can reduce the fluid–shale interface tension and increase the fluid–shale contact angles. Meanwhile, the polar group SO_3^- in the drilling fluid is adsorbed by polar substances through the van der Waals force, and the SO_3^- can thus form an ionic film on the surface of the shale. As shown in Figure 3, with increasing drilling fluid activity, the adsorption rate of SO_3^- by the difference between the activity of the drilling fluid and the activity of the shale formation is greatly increased, the rate of change of fluid–shale contact angle decreases, and then the thickness of the adsorption layer increases accordingly, improving the hydrophobicity of the shale surface and thus effectively inhibiting the hydration of the shale surface by the fluid.

As can be seen from Figure 4, for the CTAB drilling fluid, the fluid–shale contact angles increase with increasing drilling fluid activity. The hydrophilic groups of the CTAB molecule are adsorbed on the surface of hydrophilic minerals (such as quartz, mica, and montmorillonite) present in the shale via Van der Waals forces, with the hydrophobic end of the molecule facing outward. Meanwhile, the CTA^+ cations in the drilling fluid are adsorbed by O^- anions on the surface of the shale via ionic bonds (Figure 5a). As more surfactant molecules and CTA^+ are adsorbed on the surface of the shale, the thickness and density of the adsorption layer increase, which enhances the hydrophobicity of the shale surface and effectively suppresses the surface hydration of the shale (Figure 5b). As shown in Figure 4, increasing drilling fluid activity accelerates the adsorption rate of CTA^+ and promotes the formation of a hydration-inhibiting layer, which causes the fluid–shale contact angles to increase. The adsorption mechanism of CTAB on the shale surface is illustrated in Figure 5.

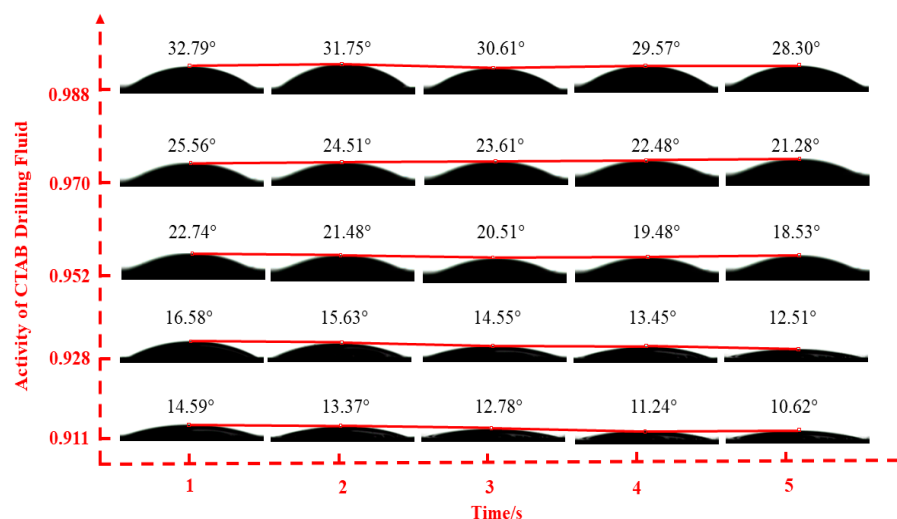


Figure 4. The evolution of fluid–shale contact angle for different activities of cetyltrimethylammonium bromide (CTAB) drilling fluid.

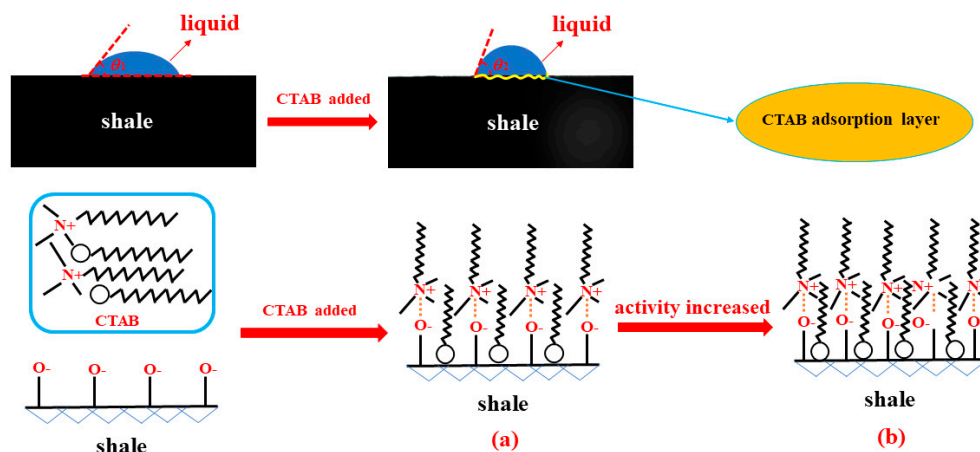


Figure 5. The adsorption mechanism of CTAB on the surface of shale.

The relationship between the drilling fluid activities (α_w) and the change ratio of the fluid–shale contact angle ($\Delta\beta/t$) is shown in Figure 6. It can be seen that with increasing drilling fluid activity the rate of change of the contact angles undergoes a decreasing trend. Increasing drilling fluid activity accelerates the movement of ions or molecules in the drilling fluid, and consequently the rate of adsorption on the shale surface is increased. This effect resulted in the observed reduction of the fluid–shale contact angles in the three drilling fluids over the same period of time. The rate of change of contact angles is the lowest using the KCl drilling fluid, which indicates that the hydration-inhibiting ability of the KCl drilling fluid is the strongest among the three drilling fluids.

3.3. Effect of Drilling Fluid Activity on the Osmotic Hydration of Shale

The relationship between the drilling fluid activity and the shale swelling ratio is shown in Figure 7.

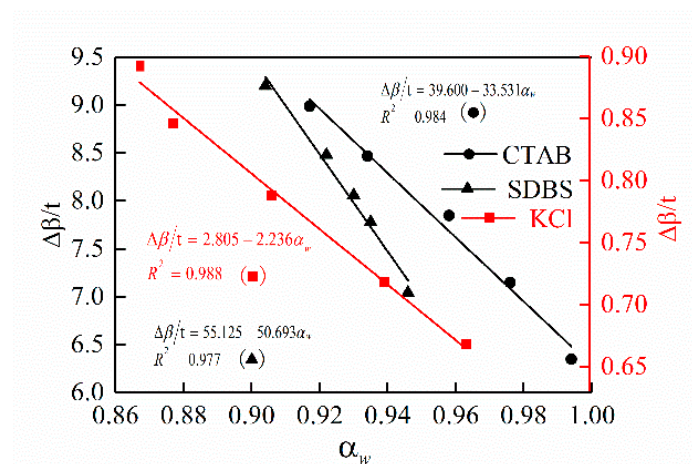


Figure 6. Relationship between drilling fluid activity and the rate of change of fluid–shale contact angle.

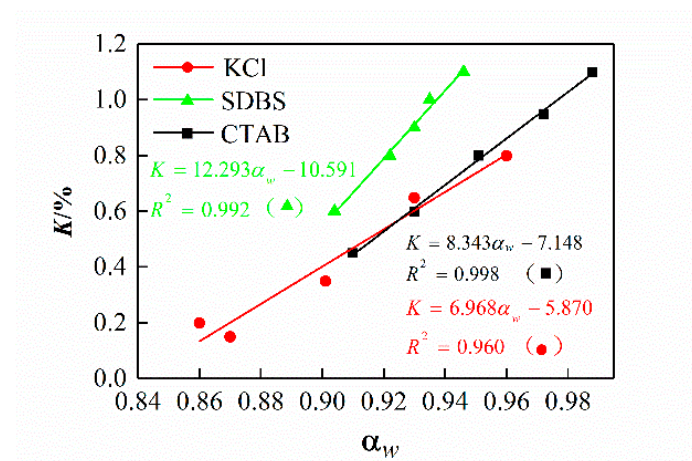


Figure 7. Relationship between drilling fluid activity and shale swelling ratio.

The results demonstrate that the shale swelling ratio increases with increasing drilling fluid activity. As the drilling fluid activity increases, the difference in activity between the drilling fluid and the shale formation increases, meaning that the driving force driving the free water in the drilling fluid into the shale formation increases. Additionally, shale formations contain abundant bedding planes and micro-cracks, which allow drilling fluids to migrate into the formation [29]. The relationship between the drilling fluid activity and the shale swelling ratio can be established by Equation (1):

$$K = a\alpha_w - b \quad (1)$$

where a is the shale swelling ratio per unit activity, which is the slope of the line in Figure 9; and b is the shale swelling ratio when the drilling fluid activity is zero; the swelling ratio, defined as the negative extreme swelling ratio, is equal to the longitudinal intercept of the line. Their units are both 1.

It can also be seen from Figure 7 that there is a significant linear relationship between the activity of the CTAB drilling fluid and the shale swelling ratio, and that there is also a significant linear relationship between the activities of the SDBS and KCl drilling fluids and the swelling ratio. Additionally, when the swelling ratio is close to zero, the drilling fluid activity can be defined as the critical drilling fluid activity (α_{w0}), whose limit is equal to the shale formation activity (α_s). When the drilling fluid activity is lower than the shale formation activity, dehydration of the shale occurs due to the migration of water molecules from the shale formation into the drilling fluid, which can cause the shale to shrink.

Moreover, when the drilling fluid activity is close to zero, the swelling ratio is defined as the negative extreme swelling ratio of shale (b). The values of b for the different drilling fluids have the following order: $b_{KCl} < b_{CTAB} < b_{SDBS}$. For the KCl drilling fluid, on the one hand, due to the difference in activity between the fluid and the shale, the water molecules in the shale are driven to migrate into the drilling fluid, and consequently the shale gradually contracts; on the other hand, as the radius of the potassium ion is similar to that of the hexagonal cavity in the clay crystal unit, potassium ions can be embedded into the cavity, which can prevent water molecules from entering the shale formation. Therefore, the shale shows the lowest negative extreme swelling ratio with the KCl drilling fluid. The principle of potassium ion embedding to prevent shale swelling is illustrated in Figure 8.

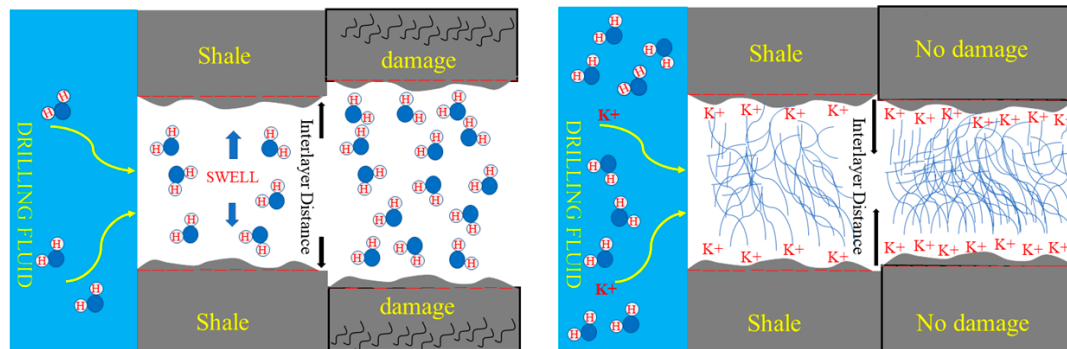


Figure 8. Principle of shale shrinkage due to potassium ion embedding.

3.4. Classification of Safety Levels of Drilling Fluid Activity

The migration of water molecules and ions into a shale formation is greatly affected by the difference between the drilling fluid activity and the shale formation activity (α_s). Therefore, the interaction between the shale formation and the drilling fluid is divided into the following three types [19,29,30]: dehydration action, surface hydration action, and osmotic hydration action. When the drilling fluid activity is higher than the drilling fluid critical activity (α_{wt}), a large amount of water molecules from the drilling fluid migrate into the shale reservoirs, which causes the shale to swell. However, when the drilling fluid activity is slightly lower than the shale formation activity, the resulting slight shrinkage of the shale formation is beneficial to the wellbore stability. Furthermore, when the drilling fluid activity is greatly reduced, the wellbore stability decreases since large amounts of water molecules migrate from the shale into the drilling fluid, causing the activity to reach the dehydration critical activity (α_{wt}). The dehydration critical activity and shale formation activity are defined as the lower limit and upper limit of the safe activity, respectively, and are considered as characteristic parameters that are closely related to the wellbore stability. The range of activities between these two parameters is defined as the safe activity window. Figure 9 is a schematic diagram indicating the divisions of the safety levels of drilling fluid activity.

As shown in Figure 9, the drilling fluid activity and swelling ratio are divided into four zones: the complete dehydration zone (A), the weak dehydration zone (B), the surface hydration zone (C), and the osmotic hydration zone (D). The zones are described as follows:

(1) In zone (A), the drilling fluid activity is lower than the dehydration critical activity. Therefore, the resulting drastic dehydration caused by the loss of free water molecules from the shale formation can result in the serious shrinkage of the shale. As can be seen from Figure 9, the degree of shale shrinkage is largest for the CTAB drilling fluid, followed by the SDBS and the KCl drilling fluids;

(2) In zone (B), the drilling fluid activity is between the dehydration critical activity and the shale formation activity. The dehydration action is weak and the degree of shale shrinkage is minor compared to zone A;

(3) In zone (C), the drilling fluid activity is between the shale formation activity and the critical drilling fluid activity. Due to the fact that water molecules are adsorbed on the surface of the clay through the van der Waals force, shale hydration only occurs on the shale surface, so the swelling ratio of shale is small. As can be seen from Figure 9, the activity window of the surface hydration of the KCl drilling fluid is the largest, followed by the CTAB and SDBS drilling fluids. This means that the surface hydration activity threshold of the KCl drilling fluid is the lowest and that of the SDBS drilling fluid is the highest;

(4) In zone (D), the drilling fluid activity is greater than the drilling fluid critical activity. Therefore, a large amount of water molecules in the drilling fluid are flushed into the shale formation, which causes the clay minerals to swell severely. As can be seen from Figure 9, in this zone, the shale swelling ratio is the largest for the CTAB drilling fluid, which indicates that the swelling-inhibition effect of the drilling fluid on the shale is the worst in this zone.

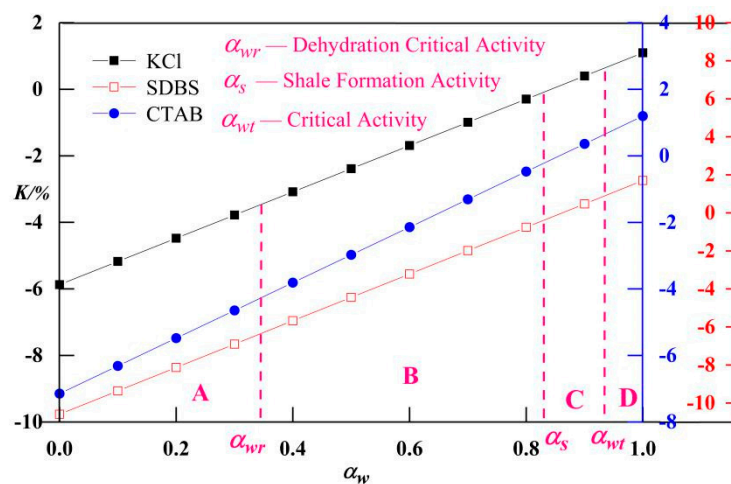


Figure 9. The divisions of the safety levels of drilling fluid activity.

4. Conclusions

In this paper, wettability and swelling tests were conducted to investigate the effects of the activities of three types of drilling fluid on the surface and osmotic hydration of shale. The following conclusions can be drawn:

(1) As the concentration of inhibitors in the drilling fluid system increases, the activity of the CTAB drilling fluid experiences the largest increase, while the activity of the KCl drilling fluid experiences the lowest increase;

(2) As drilling fluid activity increases, a decreasing trend is observed in the rate of change of the contact angles between the fluid and shale, and the shale swelling ratio increases obviously. The KCl drilling fluid showed the best inhibitory effect on shale surface hydration, and also caused the lowest negative extreme shale swelling ratio;

(3) Based on the safety levels of drilling fluid activity, when the KCl drilling fluid was used, the shale underwent the least shrinkage in the complete dehydration zone and the surface hydration activity threshold was the lowest in the surface hydration zone. The largest osmotic hydration zone was observed with the CTAB drilling fluid, which is disadvantageous for wellbore stability.

Author Contributions: H.C. proposed this topic and supervised the work; Z.Z. mainly carried out and completed this work; T.B. provided and analyzed the field data; P.S. and others supplemented and perfected this work in the later stage; all authors read and approved the final manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

KCl	potassium chloride
SDBS	sodium dodecylbenzene sulfonate
CTAB	cetyltrimethylammonium bromide
b	negative extreme swelling ratio
α_w	drilling fluid activity
$\Delta\beta/t$	change ratio of the fluid-shale contact angle
K	shale swelling ratio
a	the ratio of the shale swelling ratio to the drilling fluid activity
α_{wo}	critical drilling fluid activity
α_s	shale formation activity
α_{wt}	drilling fluid critical activity
α_{wr}	dehydration critical activity

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