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Review Paper

Advanced development of chemical inhibitors in water-based drilling fluids to improve shale stability: A review



Qiang Li ^a, Dao-Yi Zhu ^{a, *}, Guan-Zheng Zhuang ^b, Xin-Liang Li ^c

- ^a Petroleum College, China University of Petroleum-Beijing at Karamay, Karamay, 834000, Xinjiang, China
- ^b School of Environmental Science and Engineering, Guangdong University of Technology, Guangzhou, 510006, Guangdong, China
- ^c College of Chemistry and Chemical Engineering, Southwest Petroleum University, Chengdu, 610500, Sichuan, China

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ABSTRACT

Wellbore instability is a critical challenge in drilling operations, especially in shale formations where interaction with water-based drilling fluids can result in significant operational risks and increased costs. To address these issues, shale chemical inhibitors have become a crucial component in drilling fluid formulations to ensure wellbore integrity. Although several researchers have published some reviews on shale inhibitors, the latest advancements in shale chemical inhibitors over the past five years still warrant further discussion and summary. This literature review provided a comprehensive examination of wellbore instability, focusing on the patterns of instability encountered in drilling and the various shale chemical inhibitors employed to mitigate these issues. The review explored the utilization of shale inhibitors in water-based drilling fluids, and the discussion highlights the timeline evolution of these inhibitors, from traditional salts and polymers to advanced ionic liquids and deep eutectic solvents. Additionally, the mechanisms of shale chemical inhibitors are summarized to guide their application. The objective of this paper is to provide a detailed review of the development of shale chemical inhibitors in water-based drilling fluids, aiming to fully appreciate shale hydration inhibition methods and to provide insights into the selection and optimization of shale inhibitors to improve wellbore stability in challenging drilling environments.

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1. Introduction

Energy is the cornerstone and driving force behind the progress of human civilization. The economic growth, social development, and cultural advancement are all deeply dependent on a continuous and reliable energy supply. Currently, fossil fuels dominate the global energy landscape, with oil and natural gas accounting for more than 50% of the primary energy mix as early as 1965 (Pratti, 2024). According to forecasts, global energy demand is expected to increase steadily over the next 25 years, with fossil fuels continuing to dominate the energy supply (Doğan, 2024). The demand for oil and natural gas is projected to grow at an average annual rate of 0.7% and 1.2%, respectively (Li, 2023). On the one hand, advancements in drilling and completion technologies, along with the widespread adoption of hydraulic fracturing, are driving

* Corresponding author. E-mail address: zhudaoyi@cupk.edu.cn (D.-Y. Zhu). increases in oil and gas production (Chen et al., 2024; Dindoruk and Zhang, 2024; Rahim et al., 2024). On the other hand, the accelerated development of society since the industrial revolution has led to unprecedented energy demands in modern society (Akpan and Olanrewaju, 2023; Hassan et al., 2024).

The robust demand for oil and gas has fueled a boom in exploration and development activities, particularly drilling operations. Drilling is the first step in oil and gas extraction, where a hole is created in the earth to reach the target formation, which is believed to contain petroleum-based on geological and exploration data (Lei et al., 2021; Wu et al., 2020). However, wellbore instability is one of the most significant and pervasive challenges in the oil and gas drilling industry, posing risks to drilling efficiency, safety, and economic viability. It is reported that shale formations account for nearly 75% of the world's drilled hydrocarbon formations, with over 90% of wellbore instability problems occurring in these formations, leading to millions of man-hours of non-productive time and economic losses of up to 6 billion dollars each year (Ibrahim, 2021; Kang et al., 2009). Wellbore instability can manifest in various

ways, such as borehole collapse, sloughing, and excessive enlargement, often resulting in stuck pipe incidents, lost circulation, and even blowouts (Ukaeru et al., 2024). The impacts of wellbore instability extend beyond operational disruptions to include the cost of remedial measures, potential environmental hazards, and safety risks for personnel.

Multiple factors contribute to wellbore instability, with the primary cause being the interaction between the drilling fluid and the geological formations encountered during drilling. Currently, more than 80% of drilling activities rely on water-based drilling fluids (WBDFs) due to their low price, environmental friendliness, and ease of preparation (Jiang et al., 2022; Zhao et al., 2017). However, the reactive shale and clay formations are particularly prone to destabilizing interactions, such as swelling and dispersion, when exposed to conventional WBDFs. These formations are rich in clay minerals, which can absorb water and swell, causing the volume of the clay minerals to increase dramatically. This swelling pressure can exceed the mechanical strength of the wellbore walls, which may lead to their collapse (Rana et al., 2020). Additionally, clay particles from the formation may disperse into ultra-fine colloidal particles that are carried into the drilling fluid, directly affecting its performance (Khodja et al., 2010).

Oil-based drilling fluids (OBDFs) are commonly utilized to address wellbore instability issues due to their excellent shale inhibition properties. They do not hydrate with reactive shale formations, which significantly reduces swelling and dispersion (Murtaza et al., 2021). However, the utilization of OBDFs raises serious environmental concerns, including the risk of spills and the challenge of disposing of cuttings and waste fluids. In addition, the high cost of operation also limits the promotion of OBDFs. To mitigate these challenges, the drilling industry prefers to add inhibitors to WBDFs to address wellbore instability issues. Inhibitors are chemical additives designed to prevent or minimize adverse interactions between drilling fluids and geological formations. Their main function is to stabilize the clay particles and reduce their tendency to swell by absorbing water, thereby maintaining the structural integrity of the wellbore (Gholami et al., 2018; Lv et al., 2022)

Over the decades, the progress of inhibitors has evolved significantly, driven by advancements in chemical engineering, and the industry's ongoing push for more environmentally sustainable practices. Initially, the introduction of simple salts such as KCl and CaCl₂ marked the beginning of inhibitor technology (Simpson, 1974). The introduction of polymer-based inhibitors at the end of the 20th century represented a significant achievement. Polymers such as polyacrylamide (PAM) and partially hydrolyzed polyacrylamide (PHPA) offered superior inhibition properties by forming a protective film on the clay particles, thus preventing water absorption and swelling (Wu et al., 2002; Zhang et al., 2006). In recent years, there has been an increased concern for environmentally friendly and efficient inhibitors, a notable evolution being the introduction of ionic liquids (ILs) and deep eutectic solvents (DESs). They usually interact with clay through electrostatic attraction and neutralize the charge on the clay to prevent the clay mineral from swelling (Huang et al., 2020). Their cost-effectiveness and ease of preparation make them attractive alternatives to traditional inhibitors (Ma et al., 2023).

The use of chemical inhibitors in WBDFs is an essential method to improve wellbore stability and inhibit shale swelling. The synthesis, development, and application of shale chemical inhibitors is one of the most attractive research fields. The literature surveys indicate that various types of inhibitors (e.g., polymer, surfactant, nanoparticles, ILs, DESs) have been extensively reviewed to improve understanding of how to prevent clay swelling and wellbore instability (Abbas et al., 2021; Ahmed et al., 2019; Gholami

et al., 2018; Quainoo et al., 2020; Saleh and Ibrahim, 2019). These reviews typically concentrate on the action mechanisms and application status of a specific class of shale chemical inhibitors. For instance, Abbas et al. (2021) reviewed the inhibitory properties of polymers for WBDFs in terms of performance parameters such as electrostatic interaction, hydrogen bonding, thermal stability and its effect on drilling fluids rheology. However, a timeline overview of the shale chemical inhibitors evolution and a comprehensive classification of available chemical inhibitors needs to be discussed in detail. The value and novelty of this review lie in its key differences from existing reviews on this topic. Specifically, these are: 1) brief description for the patterns of wellbore instability in shale formations; 2) illustration of the mechanism of hydrated swelling of clay minerals; 3) presentation of a timeline overview of inhibitor evolution; 4) introduction to the comprehensive classification and mechanism of inhibitors.

The purpose of this review is to provide a comprehensive overview of the development of chemical inhibitors used in WBDFs, focusing on their mechanisms of action, effectiveness in drilling operations, and contributions to wellbore stability. The paper will trace the evolution and current status of widely used chemical inhibitors, emphasizing their critical role in modern drilling operations by discussing the progress from early inhibitor technologies to the latest innovations. In this work, the first part following the introduction summarizes in detail the clay minerals swelling mechanisms and wellbore instability patterns. The second section presents the evolution of inhibitors over time and provides a comprehensive classification based on their mechanism of action. Finally, the prospects and challenges of inhibitors used in WBDFs are highlighted before the conclusion. Fig. 1 illustrates the structure of this review.

2. Wellbore instability patterns

2.1. Clay minerals in shale formations

Shale is a fine-grained clastic sedimentary rock composed of clay minerals and tiny fragments of other minerals (Khadem et al., 2021). It is characterized by ultra-low permeability and is an essential caprock in oil and gas geological formations. The trapped oil or gas can be extracted by drilling wells into the shale reservoirs. The specificity of clay mineral present in a shale formation depends on factors such as the composition of the parent rock, the chemistry of the pore water, and the environmental conditions during deposition (Hou et al., 2022). Clay minerals are most commonly associated with low-energy environments, where fine-grained particles can accumulate over time. Marine shales are often rich in smectite, particularly in deep marine environments (Dayal, 2017). These environments are characterized by low sedimentation rates, and the chemical composition of seawater and organic matter content contribute to the formation of smectite (Basu et al., 2019). Marine shales tend to have higher swelling potential, which can cause issues in drilling operations. Shales deposited in river deltas or estuarine environments typically contain a mixture of clay minerals, including illite and kaolinite. These environments have moderate to high-energy conditions, with higher sedimentation rates and a higher proportion of quartz and feldspar grains (Li et al., 2024b). The clay minerals found in these shales are less reactive compared to those found in deep marine environments, which makes them generally more stable during drilling operations.

The fine-grained character of clay minerals facilitates the formation of a tight matrix, which affects fluid flow and pressure distribution within the shale (Ougier-Simonin et al., 2016). It is crucial to understand the types of clay minerals present in shale formations to predict and manage wellbore stability issues. There

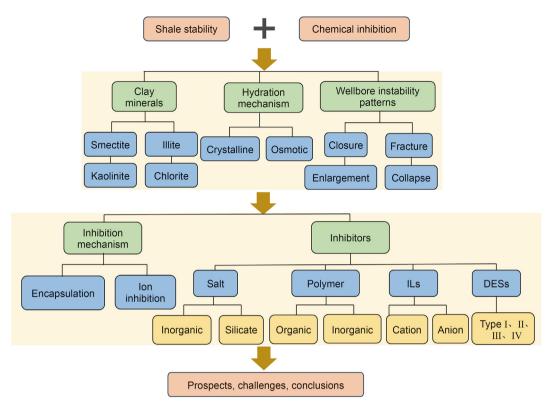


Fig. 1. The logical structure of this review work.

are four main types of shale commonly encountered during drilling operations: hydration shales, brittle shales, tectonically stressed shales, and abnormally pressurized shales (Talabani et al., 1993). The variation in clay mineral content and type in different shale formations means that each formation can present unique challenges. In general, clay minerals account for 10%-80% of shale and are a pivotal component of shale. The mineral content analysis results of shale samples from the Longmaxi Formation in the Sichuan Basin of China and Mississippian Barnett shale in the Fort Worth Basin of North America both show a high proportion of clay minerals (Loucks and Ruppel, 2007; Montgomery et al., 2006; Zhang et al., 2020). The Longmaxi shale has a maximum clay mineral content of 64% and an average clay mineral content of 33.9%, and the average clay mineral content in Barnett shale is 24.2% (Fig. 2). The proportion of clay minerals in shale is so high that researchers have to pay attention to its impact on wellbore

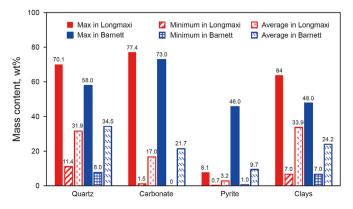


Fig. 2. Comparison of mineral content between shale samples from the Longmaxi shale and the Barnett shale (Zhang et al., 2020).

stability.

The clay minerals involved in the shale consist mainly of montmorillonite, illite, kaolinite, chlorite, and the mixed layer of these minerals. Montmorillonite is a reactive clay mineral with a strong affinity for water and exhibits excellent swelling potential. Swelling of montmorillonite in aqueous solution refers to the movement or separation of clay layers, especially those in a parallel arrangement, resulting in an increase in the interlayer space. Montmorillonite is a silicate clay mineral with a typical 2:1 layered structure consisting of two layers of silica tetrahedral sheets and a layer of alumina octahedral sheet (Huang et al., 2024). Crystalline swelling and osmotic swelling are the main swelling mechanisms of montmorillonite in shale. Illite has a similar layered structure to montmorillonite, but it has a less potential for hydration. The interlayer spaces between individual illite clay crystals are occupied by potassium ions (K⁺), making it difficult for water to penetrate into the interlayer spaces, resulting in weak expansion capacity (Karpiński and Szkodo, 2015). Kaolinite and chlorite are generally considered non-swelling clay minerals, but their ability to disperse in water is noteworthy. Shales with high kaolinite content tend to exhibit brittleness and present challenges to the mechanical stability of the wellbore. There is substantial evidence that shales with high kaolinite content are more susceptible to deposition when exposed to the WBDFs (Anderson et al., 2010). The extent, mechanism and consequence of the interaction between shale and drilling fluid depend on the type and amount of clay minerals in the shale. The main characteristics of clay minerals in shales are summarized in Table 1.

2.2. Clay minerals hydration mechanism

Raw smectite is hydrophilic due to the existence of hydrated exchangeable cations. Smectite can absorb or lose water in response to changes in humidity content in the ambient

Table 1The characteristic parameters of clay minerals in shale.

Clay minerals	Layer arrangement	Chemical formula	CEC, meq/ 100 g	Surface area, m ² /g	Morphology	Reference
Montmorillonite	2:1	(0.5Ca, Na) _{0.7} (Al, Mg, Fe) ₄ [(Si, Al) ₈ O ₂₀]·nH ₂ O	80-150	100-350	Flaky, wavy, wrinkled sheets	Worden and Morad, 2009
Illite	2:1	$K_{1-1.5}Al_4[Si_{7-6.5}Al_{1-1.5}O_{20}](OH)_4$	10-30	60-150	Irregular with granules or elongated spines	Jumaeva et al., 2023
Kaolinite	1:1	$Al_4[Si_4O_{10}](OH)_8$	3-15	10-30	Stacked plate or sheets, booklet	Rimkevich et al., 2010
Chlorite	2:1	$Mg_5Al_2Si_3O_{10}(OH)_8$	10-40	25-40	Pseudohexagonal platelets, scaly flakes	Ennis-King et al., 2017

environment (Faheem, 2018). The swelling of clay minerals in an aqueous solution is also known as hydration. The clay hydration is a function of the extent and location of layer charge, the interlayer cation species, the water activity, the temperature, the external pressure, and the salinity of the bulk solution (Balaban et al., 2015). Hydration is the movement or separation of clay layers, especially those in a parallel arrangement, resulting in an increase in the interlayer space or the expansion of the planes of layers (Fig. 3(a)).

2.2.1. Crystalline swelling

Depending on the extent of the increase in the basal spacing between two clay sheets, crystalline and osmotic swelling are the two main mechanisms reported in the literature to explain clay hydration phenomena (Luckham and Rossi, 1999). Crystalline and osmotic mechanisms involved in clay swelling are particularly associated with WBDFs. Crystalline swelling, also called surface hydration, can occur in all types of clay minerals when they are exposed to concentrated brine or aqueous solutions with high content of divalent or multivalent cations (Alcázar-Vara and Cortés-Monroy, 2019). This result is caused by the hydration of the interlayer cations (Fig. 3(b)). The hydration occurs in steps with one, two, three and four water layers (Ruedrich et al., 2011; Suquet et al., 1975). Furthermore, molecular simulation techniques have been

applied to confirm the previously described stepwise mechanism of crystalline expansion, and it has been observed that adsorbed water molecules form distinct layers in the interlayer region (Hensen and Smit, 2002). In general, the swelling of clay minerals due to crystalline hydration does not normally expand beyond the interlayer spacing of 20 Å, because the repulsive effect of ion hydration is offset by electrostatic attraction between the cation and silicate layers (Luckham and Rossi, 1999).

2.2.2. Osmotic swelling

Unlike crystalline swelling, osmotic swelling is restricted to certain clay minerals containing exchangeable cations in the interlayer region, such as montmorillonite and vermiculite. The principal action force of this mechanism is the diffusion of water molecules caused by the difference in electrolyte concentration (Fig. 3(c)). If the concentration of cations in the interlayer space is higher than that of the surrounding water, water molecules diffuse into the interlayer space to restore cation equilibrium (Anderson et al., 2010). Due to the diffusion effect of water molecules, osmotic repulsion pressure is generated between the clay layers, resulting in a large increase in the distance between clay particles. The osmotic pressure is mainly related to the difference in ionic concentration between the interlayer and the surrounding water

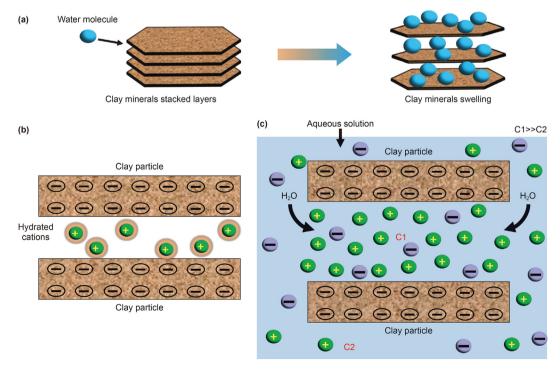


Fig. 3. (a) Schematic diagram of clay mineral swelling; (b) crystalline swelling and (c) osmotic swelling.

(Luis and Ignacio, 2017). In contrast to crystalline swelling, osmotic swelling can abruptly increase the clay mineral interlayer spacing to 30–40 Å and continue to increase to several hundred Angstroms with water content (Luckham and Rossi, 1999). Therefore, osmotic hydration is the primary factor in clay swelling.

2.3. Wellbore instability related to clay minerals

The mechanical and physicochemical properties of shale during drilling operations are considered to be the two most important aspects in the study of wellbore stability (Zeynali, 2012). It is generally accepted that the interaction of highly reactive shale formations with drilling fluids is a major contributor to wellbore instability. Drilling fluids contain various chemicals and undergo a series of physicochemical reactions with shale during the drilling process. Wellbore instability during drilling is a complex problem involving mechanics, chemistry, geology, and mineralogy, and is affected by many factors. These factors include, but are not limited to: water content and activity, membrane efficiency, clay content, mineral composition, drilling fluid type and properties, pore pressure, stress around boreholes and geological settings (Al-Arfaj et al., 2014; Murtaza et al., 2024; Quainoo et al., 2020; Saleh and Nur, 2023). Although the factors contributing to wellbore instability are multifaceted, the patterns of wellbore instability in shale formations are relatively constant and consist primarily of wellbore closure, wellbore enlargements, fracturing and wellbore collapse (Aregbe, 2017).

- (1) Wellbore closure is a time-dependent process of narrowing the borehole size and is common in shale and salt formations (Xie et al., 2019). When the wellbore is stressed, it will result in a decrease in wellbore size. The drilling problems related to wellbore closure include increased torque and drag, and stuck pipe.
- (2) Wellbore enlargement, also referred to as washout, is characterized by the wellbore size becoming much larger than its original size. This can be caused by erosion, abrasion and shale sloughing. Wellbore enlargement complicates cementing operations, impacts hydraulic requirements, and creates challenges during logging operations.
- (3) Fracturing occurs when the hydrostatic pressure of the drilling fluid exceeds the fracture pressure of the formation (Yang et al., 2022). The positive pressure differential acts on the wellbore and subsequently fractures the formation. Fracturing can prevent the circulation of drilling fluids and may allow formation fluids to flow into the wellbore. This can lead to well lost circulation or well kick, which can impact well control operations and wellbore integrity.
- (4) Wellbore collapse occurs when the hydrostatic pressure of the drilling mud is insufficient to maintain the wellbore's integrity. If not properly managed, this can lead to the flow of formation fluids into the wellbore, potentially causing a blowout. Additionally, wellbore collapse may also can result from the hydration and expansion of clay minerals.

Fig. 4 illustrates typical wellbore instability experienced when drilling a wellbore. The patterns of wellbore instability can vary significantly depending on the type and percentage of swelling clay minerals present in the shale formation. For instance, in formations composed primarily of soft shale, wellbore closure is a common issue due to the swelling of the shale. Conversely, the instability behaves differently in brittle shale formations, often leading to wellbore enlargement and collapse. This variation is due to the inherent brittleness of the shale, which makes it prone to fracturing and collapse under the mechanical stresses of drilling operations.

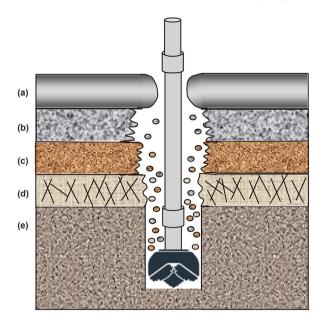


Fig. 4. A schematic of wellbore instability patterns when drilling a wellbore, (a) soft, swelling shale (wellbore closure); (b) brittle-plastic shale (wellbore enlargement and collapse); (c) brittle shale (wellbore enlargement and collapse); (d) naturally fractured shale (collapse), and (e) competent rock formation.

3. Mechanism of wellbore stabilization by chemical inhibitors

WBDFs are extensively employed in oil and gas drilling activities due to their environmental friendliness and low cost. However, they have been widely criticized for their poor ability to control wellbore stability. The application of inhibitors effectively prevents the hydration and expansion of shale. Shale inhibitors may be broadly divided into two categories depending on their mechanism: mechanical inhibitors, and chemical inhibitors. Mechanical inhibitors primarily block shale fractures and pore throats through physical plugging and bridging, but their inhibitory effects often fall short of drilling requirements. Chemical inhibitors are popular additives in WBDFs due to their excellent ability to inhibit shale hydration and swelling through multiple chemical interactions. Shale chemical inhibitors are typically used in the following methods to alleviate wellbore instability problems caused by the interaction behavior between WBDFs and shale, including encapsulation and ionic inhibition. These methods can be used independently or in combination to improve the inhibition of drilling fluids.

3.1. Encapsulation

When cuttings are exposed to WBDFs, shale cuttings are subjected to strong grinding action, resulting in hydrated dispersion as the mud is circulated. It is essential to take measures to prevent the dispersion of shale cuttings from affecting the performance of the drilling fluid. A practical method is encapsulating these shale cuttings particles in a high molecular-weight polymer. The polymer adheres to the surface of the cuttings, slows down water absorption, prevents the cuttings from dispersing and keeps their size unchanged, as shown in Fig. 5. The partially hydrolyzed polyacrylamide (PHPA), as a long-chain polymer, is often utilized as an encapsulant agents to inhibit the dispersion of cuttings in drilling fluid (Sid et al., 2023). It should be noted that the continuous adsorption of the encapsulant agents by the cuttings will lead to a

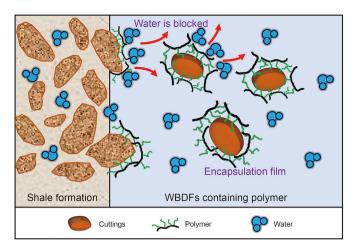


Fig. 5. Schematic diagram of inhibitor encapsulation effect.

decrease in its concentration in the drilling fluid. Therefore, the concentration of the encapsulant agents needs to be monitored in real-time and replenished in time, especially for soft shale cuttings (Mballa et al., 2013).

3.2. Ion inhibition

Hydrated cations in the interlayer space of clay minerals are one of the reasons for expansion and wellbore instability. The purpose of inhibiting the dispersion of clay in the drilling fluid can be achieved by exchanging the original hydrated cations existing in the interlayer space of clay minerals with cations that contribute to the stabilization of the shale. This method is also called ion inhibition. Ionic inhibition is mainly achieved by cation exchange, hydrogen bonding, and electrostatic interaction to inhibit the swelling of clay minerals (Fig. 6) (Tian et al., 2021; Yang et al., 2019). The effectiveness of ion inhibition depends primarily on the type and concentration of the inhibitor used. Still, the pH of the drilling fluid affects the electrostatic interactions of the ions, which may have a

negative impact on the inhibition effect. Potassium and ammonium can inhibit clay hydration due to their low hydration energy and small hydration diameter, and are the earliest drilling fluid inhibitors (O'Brien and Chenevert, 1973). There are other ionic inhibitors used in different situations, including quaternary ammonium salts, sulfonated asphalt, grafted copolymers, polyoxyalkylene amines, and anionic polymers salt (Gholami et al., 2018). With the development of molecular chemistry and polymer chemistry, ionic liquids, deep eutectic solvents and some green natural extracts have also been reported to be used in the practice of ion inhibition in WBDFs (Rana et al., 2022; Rasool and Ahmad, 2023).

4. Chemical inhibitors

Chemical inhibitors are additives added to WBDFs to neutralize the charge on clay minerals for shale hydration inhibition. They can exchange cations with clay minerals or react with clay to counteract their negative charges (Abbas et al., 2021). The inhibition mechanism of chemical inhibitors mainly involves surface adsorption, hydrogen bond formation, electrostatic and van der Waals forces between inhibitors and shale surface (Ahmed et al., 2019). Hydrogen bonding capability, electronegativity, and suppression of electrical double layers are the three most important parameters for evaluating chemical inhibitors (Mohamed et al., 2021). Since petroleum engineers encountered wellbore instability, the evolvement and development of inhibitors have never stopped. Fig. 7 presents the development history of shale inhibitors and their corresponding WBDFs.

The first shale chemical inhibitors to be utilized were inorganic salts. They act through an ion exchange mechanism to stabilize the clay structure. However, their application is restricted because drilling waste with a high concentration of Cl⁻ poses a threat to the environment and their effectiveness is reduced in extreme environments. As polymer chemistry advanced, the petroleum industry started investigating various polymers as shale inhibitors. High molecular weight polymers cover shale formation and prevent water from reacting with shale through hydrogen bonding and electrostatic interaction, which can effectively inhibit the dispersion and

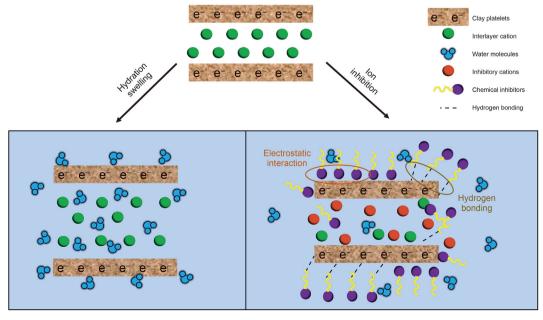


Fig. 6. Schematic diagram of clay mineral hydration and ion inhibition.

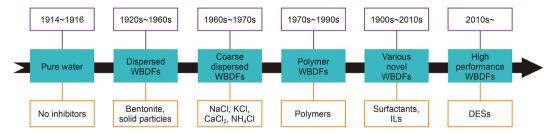


Fig. 7. The development history of shale inhibitors and their corresponding WBDFs.

swelling of clay (Werner et al., 2017; Zhong et al., 2011). However, the degradation of polymers at high temperatures is a challenge for the application of polymer inhibitors. ILs have proven to be excellent candidates for shale inhibitors at high thermal conditions. The favorable thermal stability of ILs has been utilized in geothermal and high-temperature formation drilling practices (Murtaza et al., 2023). Ionic liquids, especially those with large organic cations, not only inhibit shale but also act as corrosion inhibitors, thus reducing the need for additional additives. However, the high cost and toxicity of some ILs still pose significant challenges to widespread adoption. DESs is a mixture of simple, inexpensive salts and hydrogen bond donors. Research on DESs is still at an early stage, but their promising characteristics such as low toxicity, environmental friendliness, and cost-effectiveness, position them as a potential alternative to traditional inhibitors. From inorganic salts to DESs, many inhibitors have been developed to avoid damage caused by clay swelling. According to molecular structure, the classification and typical representatives of shale chemical inhibitors are shown in Fig. 8.

4.1. Salt

4.1.1. Inorganic salts

The earliest and most widely used inhibitors for preventing clay hydration and swelling are thought to be inorganic salts. Some inorganic salts including potassium chloride (KCl), ammonium chloride (NH₄Cl), calcium chloride (CaCl₂), sodium chloride (NaCl), and potassium hydroxide (KOH) are still used as inhibitors to provide temporary inhibition. Inorganic salts have achieved notable

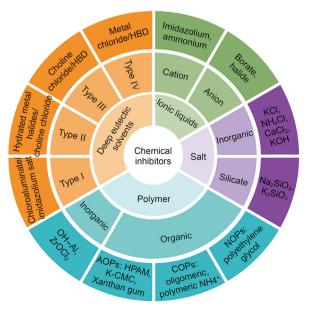


Fig. 8. Classification and typical representatives of shale chemical inhibitors.

success in combating the clay minerals swelling through electrostatic adsorption or exchange with interlayer cations (Wang, 2020). It is generally believed that Na⁺ is beneficial to promote the hydration of clay minerals, but high concentrations of NaCl have a powerful inhibitory effect. Several drilling operations have shown that KCl and NH₄Cl are better options due to their superior inhibition performance and effective cationic radius. K⁺ and NH₄ ions have the proper sizes of 0.27 and 0.29 nm respectively, allowing them to closely fit into the accessible space (0.28 nm) of the clay mineral cleavage spacing (Roehl and Hackett, 1982). Additionally, the K⁺ and NH₄ ions possess the lowest hydration energy, which promote interlayer dehydration and swelling inhibition (O'Brien and Chenevert, 1973). The characteristic properties of some common inorganic salt inhibitors are summarized in Table 2. It is worth noting that high levels of KCl can negatively impact the environment. Reports indicate that WBDFs with KCl concentrations above 1% have failed the mysid shrimp bioassay test (Suter et al., 2011).

4.1.2. Silicate

Another typical representative of inorganic salts acting as inhibitors is silicate. Since the 1930s, WBDFs formulated with silicate have been employed and provided superior shale stability (Baker and Garrison, 1939). These techniques, often referred to as protective silicate muds, were effective for drilling highly reactive shales that were capable of chemical reactions. The most prevalent silicate inhibitor employed in drilling is sodium silicate (Na₂O·*n*-SiO₂), which is partially hydrolyzed in aqueous solution to produce a colloidal precipitate through the following reaction:

$$Na_2O \cdot nSiO_2 + (y+1)H_2O \rightarrow nSiO_2 \cdot yH_2O \downarrow + 2NaOH$$

The sodium silicate solution will form a gel when the pH < 9, so a high pH value needs to be maintained when drilling with sodium silicate as an inhibitor (Li et al., 2024c). The excellent inhibition effect of silicate drilling fluid is mainly derived from the following mechanism: (1) silicate drilling fluids come into contact with multivalent-rich formation water, causing gelation and precipitation to plug microfractures and cracks; (2) gelation and precipitation of clay minerals provide a thin, tough, ultra-low permeability filter cake; (3) silica oligomers attach to drill cuttings surface through chemical bonding to inhibit clay dispersion (McDonald et al., 2007; Soric et al., 2004). In terms of HSE (Health, Safety, and Environment) benefits, silicate-based drilling fluid frequently exhibits optimal inhibition and a high rate of penetration after being improved to work synergistically with particular polymer. The application of hybrid aqueous alkali alumino silicate (AAAS) as shale swelling inhibitor in WBDFs has been reported. AAAS decreased the ζ potential and the capillary suction time was reduced, indicating its high inhibition performance. It not only inhibits shale swelling, but also acts as a shale stabilizer because it has high adsorption on the shale surface, preventing shale/water reactivity and reinforcing shale formation strength (Murtaza et al.,

Table 2 Characteristic properties of some common inorganic salt ion inhibitors (Li et al., 2024c).

Salt	Ion	Charge	Solubility, g/100 mL at 20 °C	Ionic diameter, nm	Hydration number of ion	Ionic hydration energy, kcal/g	Linear swelling inhibitive rate at 3% concentration
NaCl	Na ⁺	+1	36.0	0.20	8.6	114	36%
Ca ₂ Cl	Ca ²⁺	+2	74.5	0.21	10.0-12.0	410	40% at saturated
KCl	K^+	+1	34.2	0.27	4.2	94	64%
KH_2PO_4			22.6				50%
NH ₄ Cl	NH_4^+	+1	37.2	0.29	4.4	87	60%
$(NH_4)_2SO_4$	4		75.4				61%

2020b). Additionally, the use of hybrid silicates in drilling fluids can form a dense quartz crystal coating on the shale surface, which enhances hydration inhibition by preventing formation water filtration (Li et al., 2021).

4.2. Polymer

4.2.1. Inorganic polymers

As polymer chemistry advanced, polymers were introduced as clay inhibitors to address the limitations of inorganic salts. Since the 1960s, inorganic polymers have been utilized in drilling fluids to examine their inhibitory effects (Veley, 1969). Hydroxyl aluminum (OH-Al) and zirconium oxychloride (ZrOCl₂) are typical inorganic polymers employed as inhibitors. The multivalent metal ions in the inorganic polymer are hydrolyzed in the aqueous solution to produce a polynuclear complex with high positive charges that firmly adsorbed on the surface of the negatively charged clay, stabilizing the clay particles for a long time (Wang, 2020). Temperature and pH are the two main factors affecting the hydrolysis of inorganic polymers. Hydroxy aluminum solution is sensitive and harsh to the selection of pH environment. When pH < 3.2, OH–Al will not polymerize, and OH-Al will precipitate as Al(OH)3 at pH > 6. However, OH-Al becomes relatively resistant to acids and alkalis after being adsorbed onto clay (Zhou et al., 1995). In addition, the aluminum hydroxide adsorbed on the clay surface will undergo enhanced polymerization when the temperature exceeds 200 °C (Haskin, 1976). Veley (1969) performed flow tests on formation cores made of bentonite treated with ZrOCl₂, and the results showed that the cores could maintain 100% of their original freshwater permeability, implying that ZrOCl2 is an effective clay stabilizer.

4.2.2. Organic polymers

Compared with inorganic polymers, organic polymers have developed into a large family. Since the 1970s, organic polymers used as additives in WBDFs have played a variety of roles in drilling engineering (Young et al., 1980). Polymers not only modify rheology and filtration properties, but also improve the inhibition properties of drilling fluids. Organic polymers used to stabilize clays can be divided into three groups: anionic organic polymers (AOPs), cationic organic polymers (COPs) and nonionic organic polymers (NOPs) (Ahmed et al., 2019; Muhammed et al., 2021a).

Partially hydrolyzed polyacrylamide (PHPA), carboxymethyl cellulose (CMC), Xanthan gum (XG), and polyanionic cellulose (PAC) are common AOPs inhibitors in drilling fluids. Benefiting from the presence of anionic groups, AOPs molecules can adsorb to some positively charged sites on the clay and form hydrogen bonds with hydrated clay. AOPs act as a protective coating on the shale surface and reducing the invasion of drilling fluid into the shale layer is the main inhibition mechanism (Patel et al., 2007). PHPA employed as an AOPs to stabilize shales is typically utilized synergistically with K⁺ to enhance inhibition. CMC is a polymer with a small molecular size that can be adsorbed to clay particles through hydrogen

bonding and van der Waals forces, thereby reducing the interaction between water and shale. Kuma et al. (2020) investigated the inhibitory ability of PHPA and CMC on shale samples through static immersion test. The PHPA and CMC have similar abilities to inhibit shale expansion. After optimizing the concentration, the inhibitory effect is further improved (Fig. 9(a)). Villada et al. (2017) reported the effects of utilizing PAC and XG in shale formations, and showed that these two polymers can significantly reduce the filtration loss of drilling fluid and form a thin and tough mud cake (Fig. 9(b)). Considering the relatively weak interaction between AOPs and the negatively charged clay, however, AOPs have not been frequently utilized as clay inhibitors (Wang, 2020).

The successful practice of potassium cations as clay inhibitors is widely documented. However, we also need to be aware that potassium chloride has an adverse impact on biological ecosystems (Venâncio et al., 2023). Given that the ammonium and potassium cations have fair equivalent hydrated volumes, their functionalities are assumed to be similar. Therefore, organic polymers containing ammonium cations in their structure are used as additives for clay stabilization and swelling inhibition. According to the structure and chemical properties, the amines utilized for shale inhibition can be divided into three groups: monomeric, oligomeric, and polymeric amine compounds (Patel, 2009). Monomeric amine inhibitors have simple structures and active cationic sites, which can be used to alleviate environmental issues such as marine toxicity and biodegradability, but their applications are limited due to their temperature stability (Gomez and Patel, 2013; Tian et al., 2021). Oligomeric amines have the advantage of structural simplicity and provide more permanent stabilization than monomeric amines due to the presence of multiple active adsorption sites on the clay. The family of oligocationic amines involves polyethoxylated diamines, polyether amines, polyethylene amine salts and ether amines (Gholami

COPs are also used as swelling inhibiting additives in drilling fluid. Quaternary polyamines and quaternary polyacrylamides are the two most prevalent types. Multiple cation adsorption sites provided by the ammonium cations in the repeating unit can immobilize polymer chains on the negatively charged clay surface. This form of adsorption demonstrates great inhibitory potential and much lower replacement risk from individual cations. Bai et al. (2017) discussed the structure and application of amine-terminated hyperbranched polymer shale inhibitors, emphasizing the role of protonated primary amine groups in preventing the hydration and dispersion of sodium bentonite. Polyamine inhibitors have been recognized as crucial in controlling the hydration of the clay surface. COPs with multiple cationic centers have also been identified as effective shale inhibitors in WBDFs, providing a promising solution to the challenges associated with shale reactivity (Zhou et al., 2023a). The cationic nitrogen of COPs binds to the anionic surface of the clay, and the hydrophobic carbon chains form a network structure, preventing the migration of fines (clay particle displacement) and reducing water molecule absorption (Fig. 10) (Dong et al., 2023).

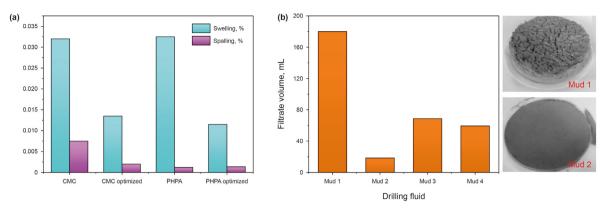


Fig. 9. (a) Swelling/spalling ratio of CMC and PHPA in WBDFs (Kuma et al., 2020); (b) left: filtrate volume of WBDFs containing XG and PAC; right: photographs of mud cake obtained from filtration tests (mud 1: base mud; mud 2: base mud + XG + PAC; mud 3: base mud + XG; mud 4: base mud + PAC) (Villada et al., 2017).

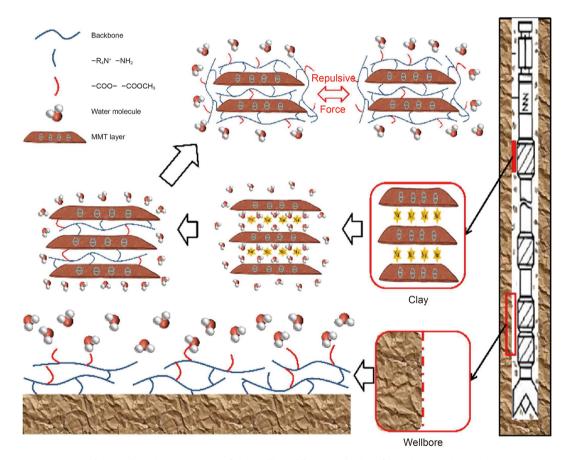


Fig. 10. Schematic representation of clay swelling inhibition mechanism of COPs (Dong et al., 2023).

The application of NOPs in shale inhibition mainly includes polypropylene oxide, polyethylene glycol (PEG) and their derivatives with nanoparticles (Ahmed et al., 2019). Due to its low environmental impact, high lubricity and better shale inhibition properties, PEG is considered as an effective shale inhibitor in WBDFs systems and shows potential to replace oil-based drilling mud (Samaei and Tahmasbi, 2007). Moreover, the low-temperature shale inhibition and gas hydrate aggregation prevention properties of PEG offer a possibility for its application in marine gas hydrate-bearing sediments (Jiang et al., 2011).

Li et al. (2019b) synthesized a novel plugging inhibitor PAS-5, which is based on amine inhibitor, PEG, and asbestos fiber. Their

work highlights the critical need for the development of new inhibitors to ensure wellbore stability. Bai et al. (2021) conducted a comparative analysis of various inhibitors in WBDFs systems, including potassium sorbate, PEG, polyetheramine, and diethylenetriamine, emphasizing the role of PEG in displacing adsorbed water molecules within clay layers through an ion exchange mechanism. Additionally, the application of ethylene glycol-based additives in drilling operations extends beyond shale inhibition. Zhang et al. (2021) carried out experimental research on ultra-low temperature drilling fluid in Arctic permafrost, evaluating ethylene glycol-based additives for their rheological properties, filtration, shale inhibition, thermal conductivity, and biotoxicity. Their

findings highlight the versatility of ethylene glycol in addressing diverse challenges encountered in drilling operations, ranging from rheological characteristics to environmental impact considerations. Thermally activated mud emulsion (TAME) fluids are typical application of polyethylene glycol as a drilling fluid inhibitor. The method of introducing inhibitive ions into PEG is used to enhance the inhibitory performance of the shale inhibitor (Deville, 2022; Villabona-EstupiÑAn et al., 2021). In generally, polyethylene glycol stabilizes the shale through the following mechanisms (Fig. 11). (1) Reducing swelling pressure between clay platelets using inhibitive ions; (2) Displacing the water on the clay surface with ethylene glycol to reduce swelling pressure; (3) Improving drilling fluid viscosity to reduce water flow into shale; (4) Forming a high-viscosity emulsion, polyethylene glycol can plug shale pores, reducing shale permeability and limiting fluid invasion.

The research on polymer nanoparticles in shale swelling has aroused great interest. Polymer nanoparticles used to enhance inhibition in WBDFs typically consist of materials such as polyacrylamide or cellulose derivatives. A typical example is the synthesis of a polyacrylamide grafted polyethylene glycol/silica nanocomposite as a shale inhibitor in WBDFs using free radical polymerization technique. This nanocomposite can form a protective film and block the shale pores, thereby more effectively inhibiting the swelling and dispersion of cuttings. Experimental results show that the shale recovery rate can reach up to 97% (Jain et al., 2015). Similarly, Zhao et al. (2023) prepared a hyperbranched polyethyleneimine/graphene nanocomposite (HPEI-G) using blending method and applied it as a shale inhibitor. The inhibition mechanism of HPEI-G involves both chemical adsorption and physical plugging. Cellulose nanomaterials, renowned for their sustainability, biodegradability, and biocompatibility, have also emerged as effective inhibitors in WBDFs. These materials prevent the hydration and expansion of shale by forming a barrier on the shale surface through electrostatic interactions and hydrogen bonding adsorption. Shen et al. (2021) produced cellulose nanocrystals via the ammonium peroxide method, reducing the linear swelling of shale samples from an initial 6 to 1.41 mm with the addition of 2% cellulose nanocrystals to the drilling fluid.

4.3. Ionic liquids

4.3.1. Evolution from surfactants to ionic liquids

Surfactants are chemicals with a chemical structure that includes a polar water-soluble group (hydrophilic head) attached to a non-polar insoluble chain (hydrophobic tail), so surfactants are amphiphilic in nature (Kamal et al., 2017). Surfactants were first proposed to stabilize shale formations in 2002, primarily by reducing the basal spacing of swelling clays (Quintero, 2002). According to the properties of the hydrophilic head group in the polar part of the surfactant, it can be divided into four categories: anionic, cationic, nonionic and zwitterionic surfactants.

The most prominent performance of surfactants as shale inhibitors in drilling fluids are cationic and nonionic. The interaction between the surfactants and the clay surface is the key to its ability to inhibit shale swelling (Fig. 12). The hydrophilic head group in the surfactant binds to the negatively charged surface of the clay mineral, while the hydrophobic group is aligned towards the aqueous phase. The hydroxyl groups of the surfactant and the oxygen atoms on the clay form hydrogen bonds, allowing the surfactant molecules to adhere to the clay surface, while the hydrophobic tails on the surfactant continue to compete with the outer phase of water molecules (Muhammed et al., 2021b; Ouellet-Plamondon et al., 2014). Some chemical surfactants, including but not limited to cetyltrimethylammonium bromide (CTAB), polypropylene glycol (PPG), hexadecyltrimethylammonium chloride (HDTMA) and sodium dodecylbenzene sulfonate (SDBS), have shown good potential in inhibiting clay swelling in drilling practice (Karimi et al., 2023; Xiao et al., 2024; Xu et al., 2023b).

Gemini surfactants, which contain two lipophilic tails and two lipophobic head groups chemically bonded via a spacer, are a new class of surfactants. The length of this spacer underlies the surface properties of Gemini surfactant (Li et al., 2019a). The swelling performance of piperazine polyether Gemini cationic surfactants with varying grafting groups and alkyl chain lengths on montmorillonite showed that below 5 mmol/L, increasing the length of the surfactant's hydrophobic tail more effectively prevents the hydration swelling of the clay (Zhou et al., 2020). Gemini surfactants with

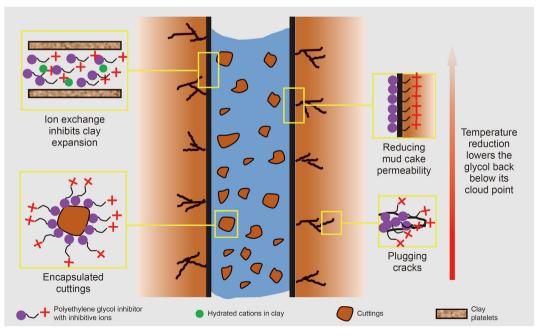


Fig. 11. Inhibition mechanism of polyethylene glycol composite inhibitor in WBDFs.

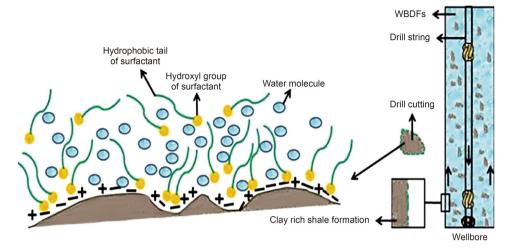


Fig. 12. Schematic representation of surfactant inhibition mechanisms (Shadizadeh et al., 2015).

different spacer lengths exhibit varying abilities to inhibit shale hydration. Gemini surfactants with 12 spacer lengths, referred to as GS12, demonstrate superior inhibition compared to GS8 and GS10, due to their enhanced hydrophobicity (Murtaza et al., 2020a). Although surfactants have shown excellent shale inhibition in some drilling operations, their effectiveness is limited, particularly in high-temperature, high-pressure formations and ultra-deep drilling environments. As the field of chemistry advances, there have been efforts to explore ionic liquids as alternative shale inhibitors to prevent shale swelling.

4.3.2. Classification of ionic liquids

Ionic liquids, liquid electrolytes usually composed of ions with an ionic-covalent crystal structure, were introduced in 1962 (Society, 1962). They are typically considered "green chemicals" and present as liquids at or slightly above room temperature. ILs often have melting points below 100 °C, unlike molten salts, due to the presence of bulky asymmetric cations and weakly coordinating anions (Zhou et al., 2023b). The cationic core, anionic core, and the substituent attached with the cationic core are the three elements that comprise ILs (Cvjetko et al., 2014). According to the ionic composition, ionic liquids can be divided into two categories, namely, anionic and cationic ionic liquids. A more detailed classification of ionic liquids is shown in Fig. 13. As inhibitors, imidazolium, ammoniums (cationic groups) and borates, halides (anionic groups) are favorable as ionic liquids in shale stabilization processes (Fu et al., 2019; Rahman et al., 2020). Extensive experiments and investigations have shown that imidazolium ILs are superior to ammonium ILs due to their higher thermal stability (Das et al., 2023).

4.3.3. Inhibition performance of ionic liquids

ILs can modify their physical properties by varying the alkyl chain length, cation and anion precursors to achieve specific tasks (Hayes et al., 2014). A comprehensive study of ILs applied to shale swelling inhibition revealed that ILs synthesized with more positively charged groups and shorter alkyl chain lengths resulted in better shale inhibition. More positive charge heads enhance the electrostatic forces, allowing ILs to adhere more easily and firmly to the clay surface. Additionally, short-chain alkyl substituents can form hydrophobic shields that effectively prevent water from penetrating interlayer spaces (Yang et al., 2017, 2019). However, the research by Jia et al. (2023) demonstrated different experimental results. Dihydroxy ionic liquids including [C₄DIPA]Br, [C₈DIPA]Br, and [C₁₂DIPA]Br exhibited good inhibition ability, with their effectiveness positively correlated with the length of the alkyl chains. Among them, the [C₁₂DIPA]Br-treated samples presented the lowest linear swelling (42.4%) and attained a high recovery (50.5%) of shale rolling testing at 160 °C. Experimental and molecular simulation results suggested that the inhibitory performance and thermal stability of [C₁₂DIPA]Br should be attributed to the synergistic effects of hydrogen bonding, electrostatic interaction, and hydrophobic shielding (Fig. 14).

Inhibition of shale expansion by ILs involves a series of complex processes. When ILs intercalate the clay interlayer space, they will be adsorbed on the clay surface through electrostatic attraction and

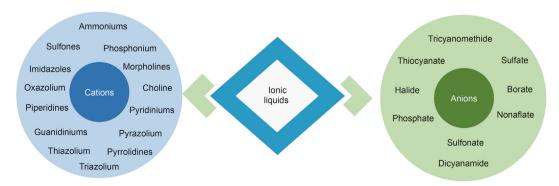


Fig. 13. Classification of ionic liquids based on cations and anions.

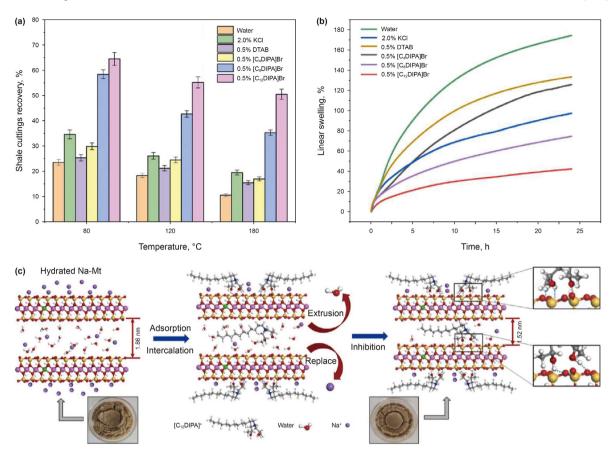


Fig. 14. (a) Hot-rolling recovery of shale fragments in dihydroxy ionic liquids at different temperatures; (b) Linear swelling test of Na-Bentonite pellets in dihydroxy ionic liquids; (c) The mechanism of shale swelling inhibition by dihydroxy ionic liquids (Jia et al., 2023).

hydrogen bonding. The ionic liquid adsorbed on the clay surface neutralizes the negative surface charge and suppresses the repulsive double layer, thus reducing the hydrophilicity of the clay minerals (Jia et al., 2022a). Meanwhile, water molecules are removed from the interlayer space and the clay plates are fixed together by the coordination of hydrogen bonds and electrostatic attraction forces (Zhong et al., 2011). The molecular structure of ILs may play a crucial role in their inhibitory properties. The comparative experiment of the inhibitory ability of the traditional ILs 1-hexyl-3-methylimidazolium bromide (BMH) and the Gemini ILs 1,2-bis(3-hexylimidazolium-1-yl) ethane bromide (HMH) showed that HMH performed a stronger electrostatic interaction and reduced surface tension than BMH, and formed a more hydrophobic shield on the clay surface (Fig. 15) (Jia et al., 2020; Ma et al., 2022).

ILs have been well developed in recent years as excellent shale inhibition additives in WBDFs. Ahmad et al. (2019) emphasized on the utilization of ILs in WBDFs to alter the wettability and hydration properties of shale to improve borehole stability. Ahmed et al. (2020) investigated in depth the mechanism, performance and effects of different anions of imidazole-based ionic liquids as clay swelling inhibitors. In addition, Khan et al. (2021b) explored the use of hydrophobic ILs and Gemini surfactants to develop novel shale swelling inhibitors for WBDFs. He et al. (2022) studied the impact of imidazolium Gemini ILs with different alkyl chains on the swelling and dispersion ability of montmorillonite, and combined with molecular simulation technology to determine the intercalation process and molecular arrangement of ILs in the interlayer space of montmorillonite. Murtaza et al. (2023) proposed the

practice of pyrrolidinium-based ILs as effective shale swelling inhibitors for WBDFs and examined the effect of the inhibitors on the rheology of drilling fluids. The latest literature reports on ILs as shale inhibitors in WBDFs is summarized in detail in Table 3.

4.4. Deep eutectic solvents

4.4.1. Classification of deep eutectic solvents

Deep eutectic solvents are a new class of ILs analogues that have been considered alternatives to ILs since they share many of the same properties and properties. Although the terms DESs and ILs have generally been used interchangeably in the literature, it is necessary to clarify that these refer to two distinct categories of solvent (Smith et al., 2014). In terms of structure and composition, ILs are electrolyte systems composed of one type of ions, while DESs are liquid eutectic salts of two or more components (Chen et al., 2019).

DESs are usually obtained by the complexation of a quaternary ammonium salt with a metal salt or hydrogen bond donor (HBD) (Smith et al., 2014). Compared to any of the individual components, the final mixture has a minor melting point. The first DESs was a combination of cholinium chloride and urea with a melting point of 12 °C, much lower than that of the starting components at 302 and 133 °C, respectively (Abbott et al., 2004a). The primary classification of DESs are based on the type of complexing agent utilized (Fig. 16). Type I DESs formed by quaternary ammonium salts and metal chlorides can be considered an analogous category to metal halide/imidazolate systems (Abbott et al., 2004b). The majority of DESs that have been prepared and studied thus far are also Type I

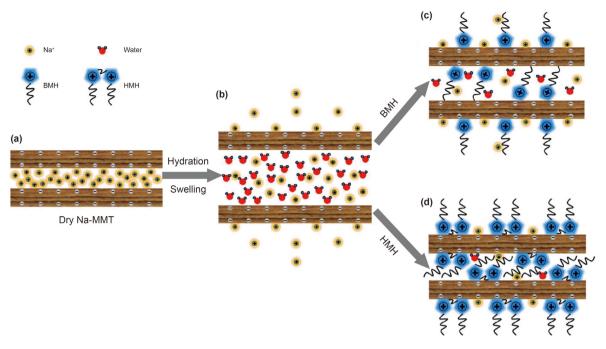


Fig. 15. Inhibition mechanism of BMH and HMH (Jia et al., 2020).

Table 3 Performance evaluation of ILs as shale inhibitors.

ILs	Abbreviation	Concentration, %/Temperature, °C	Linear swelling rate, %	Recovery rate, %	Apparent viscosity, mPa·s	Zeta potential, mV	XRD, nm	Contact angle, °	Reference
Trihexyltetradecyl phosphonium bis (2,4,4-trimethyl pentyl) phosphinate,	Tpb-P	0.1-0.5/65	82-98	15.7 -24.4	7.2-8.9	NA	NA	43.5 -46.3	Khan et al., 2021a
1-Vinyl-3-ethylimidazolium bromide; 1-vinyl-3- ethylimidazolium tetrafluoroborate; 1-vinyl-3- butylimidazolium hexafluorophosphate; 1-vinyl-3- ethylimidazolium bis(trifluoromethylsulfonyl)	VeiBr; VeiBF ₄ ; VeiPF ₆ ; VeiTFSI	0.5-2.0/120	134 -154.5	48-67	6–137	-35.6 21.2	1.34 -1.45		Yang et al., 2021
NH ₂ — and HO—functionalized ionic liquids	[HOEtMIm] BF ₄ ; [C ₂ NH ₂ MIm] BF ₄	1-2/120	62.7 -73.2	61.18 -88.47	NA	-18 9.2	1.29 -1.44	NA	Ren et al., 2021
The fatty acid-based ionic liquids with different anionic groups	FAILs	0.1/80-160	75.39 -112.23	45.32 -75.55	NA	−27.3 −−19.6	1.42 -1.72	57.5 -80.1	Jia et al., 2022b
1-Butyl-3-Methylimidazolium chloride	BMIMCL	0.25-1.0/NA	49.5 at 2 wt%	NA	NA	-23.4 -16.3	NA	NA	Rahman et al., 2022
1-Ethyl-3-methylimidazolium bromide; 1-propyl-3- methylimidazolium bromide; and 1-butyl-methylimidazolium bromide		0.5-2/25-120	89 -117.3	50.5 -81.9	10-120	-8.8 3.3	1.38 -1.43		Kinkeyi et al., 2023
The dihydroxyl ILs	[C ₄ DIPA]Br; [C ₈ DIPA]Br; [C ₁₂ DIPA]Br	0.5/80-160	42.4 -124.3	18-64.5	NA	-35.8 22.9	1.42 -1.7	50-74	Jia et al., 2023
1-ethyl-1-methylpyrrolidinium bromide	1E1MPyBr	0.1-0.3/NA	97-118	NA	NA	-21.9	1.42	NA	Murtaza et al., 2023
The polyionic liquid functionalized polystyrene nanosphere	PSIL	0.5-3/150	38.3 at 3 wt%	41.2 at 3 wt%	7–12	-30.1 24.1	NA	NA	Li et al., 2024a

(Hansen et al., 2021). Type II, composed of quaternary ammonium salts and a metal chloride hydrates, can increase the melting point scope of DESs. Due to its capacity to solubilize a wide range of transition metal species, Type III DESs, which are made from choline chloride and HBD, have received considerable interest. Benefiting from the advantages of low cost, easy preparation, and biodegradability, this type of DESs has been utilized in the removal of glycerol from biodiesel, the treatment of metal oxides, and the

synthesis of cellulose derivatives (Abbott et al., 2004c, 2006). Type IV, consisting of metal chlorides and HBD, a successful example is that ZnCl₂ has been demonstrated to form eutectics with urea, acetamide and ethylene glycol (Zhang et al., 2012).

4.4.2. Advancement of deep eutectic solvents

DESs are regarded as the latest advancement in shale inhibitors for WBDFs, demonstrating significant potential in practical

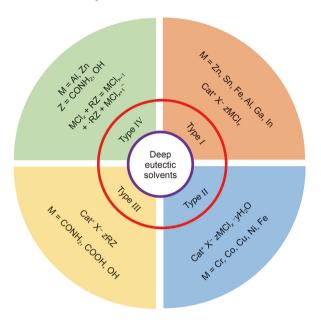


Fig. 16. General formula for the classification of DESs. The general formula of DESs is $\operatorname{Cat}^+ X^- z Y$, where Cat^+ is any ammonium, phosphonium, or sulfonium cation, and X is a Lewis base, generally a halide anion. The complex anionic species are formed between X^- and either a Lewis or Brønsted acid Y. z refers to the number of Y molecules that interact with the anion.

applications. Researchers have been striving to uncover the mechanism through which DESs inhibit shale hydration. The mechanism by which DES inhibits shale swelling involves both physical and chemical effects. The physical effect arises from DES's ability to reduce the hydration of clay minerals within the shale matrix. These clay minerals possess a layered structure consisting of negatively charged hydrous aluminum silicates and positively charged exchangeable cations (such as sodium and calcium ions) in the interlayer space. In the presence of water, these positively charged ions interact with the negatively charged platelets, expanding of the interlayer space (Chen et al., 2022). DESs interacts with these positively charged ions, limiting water penetration into the interlayer space of clay minerals, thereby reducing hydration and shale expansion, as illustrated in Fig. 17 (Beg et al., 2021).

The primary chemical mechanism of DESs as a shale inhibitor is its capacity to generate hydrogen bonds. Hydrogen bonds link the negatively charged clay layers to the DESs, resulting in charge neutralization. Total hydrogen bond count and strength of covalent

polar bond are the two pivotal criteria for evaluating the inhibitory capacity of DESs. A large number of experimental investigations have shown that more total hydrogen bonds and greater covalent polar bond strength will help to enhance the inhibitory ability of DESs (Hammad et al., 2022; Rasool et al., 2021; Skulcova et al., 2018). The specific inhibitory mechanism of DESs can be summarized as follows (Jia et al., 2022a; Ma et al., 2021).

- (1) Forming hydrogen bonds: Hydroxyl (-OH), carboxyl (-COOH), amino (-NH₂) and other functional groups in DESs molecules form hydrogen bonds with -OH or oxygen atoms on the clay surface (Fig. 18).
- (2) Electrostatic attraction: There is a strong electrostatic attraction between the cations from the DESs and the negative charges on the clay surface. DES molecules tightly adsorb onto the surface and interlayers of the clay due to hydrogen bonding and electrostatic attraction, effectively inhibiting the adsorption of water molecules.
- (3) DESs can effectively reduce the surface tension of liquids and weaken the penetration of water into the clay.

DESs are increasingly being employed as shale inhibitors in drilling fluids because they effectively stabilize wellbore during drilling in shale formations. Jia et al. (2019) investigated the use of various DES, including CM-DES, CP-DES, and CIM-DES, as shale inhibitors in WBDFs. The inhibitory properties of DESs were

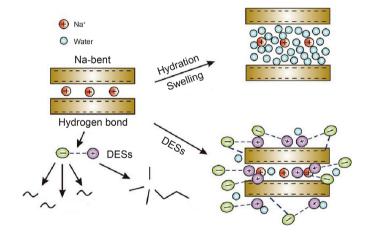


Fig. 18. Mechanism diagram of DESs chemical inhibition of clay hydration expansion (Ma et al., 2021).

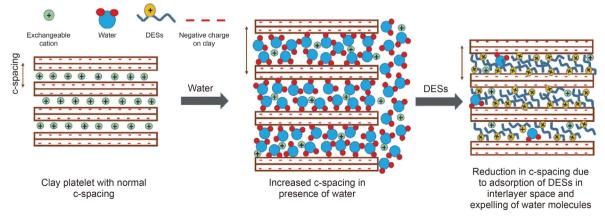


Fig. 17. DESs expel water from the interlayer space to inhibit clay expansion (Beg et al., 2021).

Table 4The recent research advances of DESs for shale stabilization

DESs	Highlights		Mechanism	Reference			
Choline chloride:urea (1:2)	DESs are the cheaper and non-toxic Strong hydrogen bonding alternative to ILs.						
	Improve the rheologic						
	filtration performance of mud. • DESs contribute to mud dispersion						
	 DESS contribute to in DES intercalates with 						
	and causes the declin	5 5					
Choline chloride:urea (1:2)		The electrostatic attraction and the hydrogen	Jia et al.,				
	and polyether diamin		bonding between DESs and clay.	2022a			
	 Good high temperate 160 °C). 	ure stability (at					
	 Low cost and rema properties. 	rkable inhibitive					
The combination of potassium carbonate based DES and modified oxazoline based polymer	 The double action in best inhibition result. 	hibitor gave the	Hydrogen bonding and Vander wall forces (electrostatic attraction force)	Rasool et al., 2022a			
	• The inhibitors imp		·	,			
	activity of the clay slu	•					
	 Inhibitor intercalat interlayer space 	ed IIIto Ciay					
Choline chloride:ethylene glycol (1:1); Choline chloride:glycol		ESs are efficient	The hydrogen bonding and electrostatic	Xu et al.,			
(1:1); Choline chloride:1,4-butanediol (1:1); Choline chloride:	hydration inhibitors.		interactions	2023a			
xylitol (1:1)	Better inhibition perfe	rmance than KCl					
	and polyether.Reduce the surface t	oncion of liquide					
	and the adsorption of						
Ascorbic acid:glycerine (1:10)			Hydrogen bonding and neutralizing negative	Rasool			
	friendly.	,	charges on clay	et al., 2023a			
	 Reduce surface tensi 	on and capillary					
	pressure.Biodegradable and no	n tovic					
Epsom salt:glycerine (1:2)	0		Hydrogen bonding and neutralizing negative	Rasool and			
zpoom saieig.yeerine (112)	of clay surfaces, and o			Ahmad,			
	layers.			2023			
	Improve rheology						
	properties of the drillBetter inhibition perfe	-					
	and ILs.	illidile tildli Kei					
Calcium chloride:glycerine (1:4)		nt and plastic	Hydrogen bonding and modifying the	Rasool			
	viscosity of drilling fl		chemical and physical properties of clay	et al., 2023b			
	Inhibit hydrate forma		mineral surfaces				
	 Cheaper and more friendly. 	environmentally					
choline chloride:citric acid (1:1)		urface hvdration	Hydrogen bonding and adsorption	Bai et al.,			
	of the shale.	j	, 2	2023			
	• Reduce the shale						
	hydrophilicity, and su						
	 DESs and cetyltrin bromide have an exce 						
	effect	ment synergistic					
Potassium chloride:glycerine (1:8)		d rheology and	Hydrogen bonding and neutralizing negative	Rasool and			
	filtration performance		charges on clay	Ahmad,			
	The excellent high stability.	h temperature		2024			
	stability • DESs alter the face-ed	ge orientation of					
	clay.	Se offentation of					
Choline chloride:glycerol (1:1)	• The smaller capillar		Adsorption and hydrophobicity	Xu et al.,			
	minimizes water infil			2024			
	Significantly increase	the water contact					
	angle of shale.Superior hydratic	on inhibition					
	compared to commer						
	compared to commic						

attributed to electrostatic attraction and hydrogen bonding with clay minerals. Additionally, the inhibition performance of the three DESs exceeded that of KCl and polyether diamine. The structure of each DESs resulted in different inhibition capabilities, among which CP-DES containing benzene rings presented the best inhibition performance. Rasool et al. (2021) explored the application of potassium carbonate-based DESs as a potential drilling fluid inhibitor, emphasizing the non-toxic and biodegradable nature of DESs

compared to conventional ILs. Furthermore, the combination of polymer and DESs has been proposed as a double action shale inhibitor to enhance shale stability and prevent wellbore instability. Meanwhile, natural deep eutectic solvents (NADES) based on environmentally friendly compounds such as calcium chloride-based and ascorbic acid based have been studied as potential additives for shale stabilization and inhibition (Rasool et al., 2023a, 2023b). These NADES have shown promising results in improving

the rheological properties of drilling muds and inhibiting shale expansion. Table 4 lists a few recent research advances in utilizing DESs as shale inhibitors.

5. Prospects and challenges

The chemical inhibitors in WBDFs have emerged as crucial measures for enhancing wellbore integrity. Although inorganic salts have historically played a key role in inhibiting shale swelling during early drilling activities, several challenges have emerged with their use as shale inhibitors. A primary issue is their limited effectiveness in preventing shale hydration and swelling in hightemperature, high-salinity environments commonly encountered in deep or ultra-deep wells. While salts like calcium chloride and potassium chloride can reduce water activity and improve shale stability, their performance tends to degrade under extreme conditions. Additionally, the solubility of certain inorganic salts can vary significantly with temperature, leading to inconsistencies in their effectiveness as inhibitors. Among the most promising advancements are the development and optimization of newgeneration inhibitors like ILs and DESs. These compounds, with their unique physicochemical properties, provide tailored solutions for shale inhibition. Unlike traditional salts and polymers, ILs and DESs can be custom-designed at the molecular level to specifically interact with shale components. By adjusting the molecular structure of these inhibitors, researchers can optimize their performance for specific geological conditions, thereby reducing the risk of wellbore instability.

The environmentally friendly properties of ILs and DESs make them sustainable alternatives to traditional inhibitors. Their low toxicity and biodegradable potential of ILs and DESs make them consistent with the goal of sustainable oilfield development. The collaboration between chemists, petroleum engineers and geologists are expected to advance shale inhibition technology. Integrating computational chemistry and molecular dynamics simulations into inhibitor design allows for the prediction and optimization of inhibitor-shale interactions before laboratory synthesis. This approach not only accelerates the development process of shale chemical inhibitors, but also reduces the costs and environmental impacts associated with repetitive testing.

Despite the promising prospects, chemical inhibitors for shale wellbore stability face numerous challenges. These challenges involve technical, economic, and environmental fields, and overcoming them requires the concerted efforts of academia, industry, and policymakers. The heterogeneity of shale formations leads to significant variations in mineralogy, porosity, permeability, and mechanical properties. This heterogeneity complicates the design and application of chemical inhibitors. Even with advanced molecular design techniques, it is still a daunting task to develop inhibitors that perform consistently in different shale formations. The high cost of new inhibitors, such as ILs and DESs, poses another challenge. Although these compounds offer exceptional performance and environmental benefits, they are more expensive to synthesize and produce than traditional salts and polymers, raising concerns about their economic viability, particularly in low-margin operations. From an environmental perspective, although ILs and DESs are promoted as green alternatives, their long-term ecological impacts remain uncertain. The biodegradability and toxicity of these compounds can vary significantly depending on their chemical structure, and some ILs and DESs may accumulate in the environment or degrade into harmful byproducts. Comprehensive environmental impact assessments are necessary to ensure that the widespread use of these inhibitors does not result in unintended ecological consequences.

6 Conclusions

The literature review highlights the intricate mechanisms of shale hydration expansion and the consequential wellbore instability, presents the patterns of wellbore instability and emphasizes the critical role of chemical inhibitors in mitigating these challenges.

- (1) The interaction between highly reactive shale formations and drilling fluids is a major contributor to wellbore instability. The patterns of wellbore instability in shale formations are relatively constant, mainly including wellbore closure, wellbore enlargements, fracturing and wellbore collapse.
- (2) Through a comprehensive historical analysis, it is evident that the evolution of chemical inhibitors—from traditional salts and polymers to advanced ionic liquids and deep eutectic solvents—has significantly enhanced the efficacy of shale inhibition in drilling fluids.
- (3) The mechanism of shale swelling inhibition is various for different chemical inhibitors. Recent advances in ILs and DESs offer promising avenues for more targeted and environmentally friendly inhibition strategies. These inhibitors, through molecular structure design and chemical synthesis, have the potential to provide strong hydrogen bonding interactions and electrostatic interactions to prevent shale swelling while maintaining environmental compatibility.
- (4) Although some progress has been achieved, challenges to effective shale inhibition remain, particularly in synthesizing novel inhibitors to accommodate diverse shale compositions and field conditions, as well as addressing environmental issues associated with chemical usage. Future research should therefore focus on developing sustainable, multifunctional inhibitors that not only improve shale stability but also align with the industry's growing emphasis on environmental stewardship.

CRediT authorship contribution statement

Qiang Li: Writing — review & editing, Writing — original draft, Validation, Methodology, Investigation, Formal analysis. **Dao-Yi Zhu:** Writing — review & editing, Supervision, Methodology, Formal analysis, Conceptualization. **Guan-Zheng Zhuang:** Writing — review & editing, Methodology, Investigation. **Xin-Liang Li:** Writing — review & editing, Investigation.

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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