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Review of Geochemical and Geo-Mechanical Impact of Clay-Fluid Interactions Relevant to Hydraulic Fracturing

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Abstract

Shale rocks are an integral part of petroleum systems. Though, originally viewed primarily as source and seal rocks, introduction of horizontal drilling and hydraulic fracturing technologies have essentially redefined the role of shale rocks in unconventional reservoirs. In the geological setting, the deposition, formation and transformation of sedimentary rocks are characterised by interactions between their clay components and formation fluids at subsurface elevated temperatures and pressures. The main driving forces in evolution of any sedimentary rock formation are geochemistry (chemistry of solids and fluids) and geomechanics (earth stresses). During oil and gas production, clay minerals are exposed to engineered fluids, which initiate further reactions with significant implications. Application of hydraulic fracturing in shale formations also means exposure and reaction between shale clay minerals and hydraulic fracturing fluids. This chapter presents an overview of currently available published literature on interactions between formation clay minerals and fluids in the subsurface. The overview is particularly focused on the geochemical and geomechanical impacts of interactions between formation clays and hydraulic fracturing fluids, with the goal to identify knowledge gaps and new research questions on the subject.

Keywords: Shale geochemistry and geomechanics, Clay minerals, Hydraulic fracturing fluids, Shale formation fluids

1. Introduction

Clay minerals interactions with fluids have gained attention in the petroleum industry because of their presence in source rocks, reservoir rocks and seal rocks in petroleum systems. In conventional reservoirs, interactions between clay minerals and fluids have been studied in relation to wellbore integrity and fines migration during production. The inception of enhanced oil and gas recovery, hydraulic fracturing and carbon storage technologies, highlighted knowledge gaps in terms of interactions between clays and fluids injected into the subsurface. Research efforts are focused to understand the impact of clay-fluid reactions geochemistry on shale geomechanics, and deciphering the mechanisms that drive these interactions in order to optimise various technologies adopted by the industry.

In retrospect, studies on interactions between clay minerals and formation fluids have been going on in the petroleum industry well before introduction of the advanced technologies alluded to above. These studies were mainly focused on the relationship between clay minerals interactions with formation fluids during formation, migration and deposition of hydrocarbons. For example, Drits et al. [1] studied clay mineral-fluid interactions in order to gain insight into transformation processes in clay minerals during generation and migration of hydrocarbons.

This review seeks to present a concise overview of published studies on interactions between clay minerals and various fluids in the subsurface with particular emphasis on hydraulic fracturing fluids. Reaction mechanisms as well as geochemical and geomechanical impacts are assessed.

2. History of clay-fluid interactions

Trends of research on interactions between formation clay minerals and fluids over the years have largely been determined by the exigencies of the petroleum industry. The drive for this is the need for in-depth understanding of reactions in order to characterise reservoirs and cap rocks, as close as possible to *in situ* conditions. Summary of research trajectories in major periods is explained in the following paragraphs and captured in Figure 1.

2.1 Petroleum formation and migration

The focus of researchers in the 1940s and 1950s, during worldwide oil and gas exploration, was to investigate the origins of petroleum. Reactions between clays and subsurface fluids were studied extensively. At that time, the major concern was assessment of quality of organic source rocks and the mechanisms involved in generation of oil and gas. In this regard, researchers such as Weaver [2, 3] and Sarkissian [4] recognised that analyses of clay rocks (shales) could be used to track the generation and migration of petroleum in source rocks. Weaver [2] noted that expandable clays are capable of withholding their pore water to greater depths. He therefore inferred that waters in expandable clays at greater depths were responsible for transporting hydrocarbons to reservoir rocks. This inference was premised on

Figure 1. Schematics of typical clay fluid interaction research topics and outcomes relevant to hydraulic fracturing of shale formations.

and supported by earlier studies where over 20,000 samples from major petroleum producing basins in the US showed strong statistical correlation between expandable clay minerals and hydrocarbon production. Similar to Weaver [2], Sarkissian [4] also studied petroleum deposits in the USSR and reported that clay minerals in argillaceous rocks were significant in the formation and deposition of petroleum resources. Premising on earlier works alluded to above, other researchers also used clay rock analyses to determine the hydrocarbon emplacement and migration times and for petroleum system analysis [1, 5–9]. Some of these works are summarised below.

Hamilton [5] used K-Ar dating to assess the formation of illite relative to the timing of generation and migration of hydrocarbons. Considerable correlation was found to exist between these two events. He reported that, in most cases, the timing of the expulsion of hydrocarbons was the same time authigenic illite formation ceased. He concluded that the link between clay-fluid interaction and petroleum generation and migration was therefore established, thus presence of authigenic illite could be used as an indicator of petroleum formation and migration.

Kelly [7] used mineralisation history present in fractures to reconstruct the migration history of hydrocarbons to their current reservoir and found that most petroleum migration paths showed preponderance of illite and clay mineral precipitates. He concluded that illite and other clay mineral precipitates can be used as an indication of petroleum migration pathway.

Jiang [6] examined clay minerals from the oil and gas perspective and drew a lot of parallels between various types of clays, their structural and geochemical transformations as a function of formation and transport of hydrocarbons. Jiang's work is different from earlier works in the sense that he investigates comprehensively the transformations that take place from deposition of rock to when petroleum is formed and expunged.

2.2 Drilling and completion

Adverse economic impact posed by swelling clays during drilling and completion caused intensification in studies of clay-fluid interactions with the aim of understanding the problem and solving it in the shortest possible time. Research was thus aimed at understanding the mechanisms that drive clay swelling during interactions with engineered muds. The conditions of clay swelling and accompanying complications were studied thoroughly with abundant literature to that effect [10–16].

Van Oort et al. [14] undertook an overview of the mechanisms guiding clay-fluid interactions. Their work identified the mechanisms by which various engineered drilling fluids suppress adverse reactions of clay minerals with water based fluids during drilling and completions. In concluding, the authors simplified their work by categorising drilling fluids into five groups based on the mechanism by which they stabilise clays in shale formations during drilling.

Shukla et al. [17] conducted a review of earlier works on clay mineral swelling in unconventional reservoir systems. They concentrated on the various conditions under which clays swell and the types of clay swelling. They also identified various clay stabilisers and gave a brief on how these work.

2.3 Enhanced hydrocarbon recovery

At inception of shale gas, tight sands and other unconventional petroleum systems development, enhanced hydrocarbon recovery techniques were at advanced stage. The need to fine-tune these technologies to the needs of unconventional reservoir systems spurred another era of research focussed on clay-fluid interactions in

unconventional petroleum systems. One of the earliest works in this area was conducted by Zhou et al. [15] who premised their research on the fact that injected fluids caused formation damage due to clay swelling. They identified two types of swelling due to these interactions; crystalline swelling and osmotic swelling, the later posing significant adverse effects on reservoir quality. Alalli et al. [18] also noted that injected fluids caused disequilibrium in formation which leads to dissolution and precipitation of minerals in an attempt to return to equilibrium state. Dissolution and precipitation patterns were thus examined in order to identify their impact on reservoir quality. Dissolution of minerals, they noted, enhanced the porosity and permeability of formation by creating additional pore volume and linking previously unconnected pores; whereas precipitation of new minerals had adverse impact on reservoir quality due to the occlusion of flow paths, due to mineral growth within the existing pore space.

Buller et al. [19] analysed the Haynesville shale play in East Texas to understand what factors were responsible for efficiency of hydraulic fracturing in this formation. Their work concluded that, in high clay content zones, the efficiency of the fracturing was low due to massive proppant embedment and migration of fines. They postulated that post fracturing diagenetic events could also be initiated due to clay minerals interaction with fracturing fluids.

A similar effort was undertaken by Radonjic et al. [20] in their research focused on Caney shale. They sought to draw the link between mineralogical composition and microstructure of Caney shale to mechanical responses in order to delineate formations suitable for fracturing as well as predict mechanical responses of these formations.

2.4 Geological CO₂ storage

Recent surge in research on interaction between clay minerals and CO₂ in the subsurface is due to the advent of the concept of CO₂ capture, utilisation and storage (CCUS). The CCUS technology may also incorporate enhanced hydrocarbon recovery when CO₂ injection is done in depleted oil and gas reservoirs. The importance of clay-fluid reactions is seen in the fact that most target storage reservoirs and accompanying seal formations have high clay mineral contents. In addition, reactions that cause immobilisation of CO₂ and the ability of caprock to withstand pressures resulting from CO₂ plume beneath are directly related to the amounts and types of clay minerals in the formation [21, 22]. Many studies have thus been conducted to investigate clay-fluid interaction in the context of understanding their implications on combined enhanced hydrocarbon recovery and CO₂ storage projects [23–29].

Olabode and Radonjic [27] studied the reaction between CO₂-Brine and caprock formation to assess the impact of mineral precipitation patterns on caprock integrity at elevated temperature and pressure conditions. They concluded that, precipitation of minerals could cause the sealing of micro-pores in caprocks thus enhancing their ability store CO₂ within the subsurface. Hui Du et al. [30] also studied the sealing properties of caprock at nano and micro-scale with the premise that the durability of the caprock is directly affected by the nanostructures and microstructure of these rocks.

3. Clay minerals

3.1 Chemical composition & crystallographic structure of clay minerals

Clay minerals are a product of rock weathering, and form from decomposition of feldspar minerals in hard rocks such as granite. They are commonly

described as soil particles with sizes below 2 μm , often labelled as nature's nanoparticles. In terms of chemical composition, clays belong to a group of minerals called aluminosilicates. The aluminosilicates are composed of complex arrangement of atoms to form diverse structural configurations with the basic components being silicon, aluminium and oxygen. Silicon and aluminium atoms bond with oxygen to form silicon tetrahedral sheets and aluminium octahedral sheets respectively. These sheets are subsequently bonded by sharing common oxygen atoms, though the oxygen atoms at the edges of both sheets are left unpaired. These unpaired oxygen atoms at the edges of the sheets impose negative charges on clay mineral surfaces rendering them water sensitive and highly reactive to cations [31, 32].

Another factor contributing to high negative charges in clay minerals is the isomorphic cationic substitution within the sandwiched tetrahedral and octahedral sheets, which leads to imposition of excess negative charges on clay mineral surfaces [33]. The mechanism described above contributes to higher levels of clay sensitivity to water-based engineered fluids in subsurface.

3.2 Classification of clay minerals

Hughes [34] was one of the earliest researchers to attempt classification of clay minerals relative in the petroleum industry. This classification was done a few years after commercialization of X-Ray Diffraction (XRD) technology, which was hitherto used by petroleum companies as a method of studying clay minerals. Hughes [34] classified clay minerals into: Kaolinite, Smectite, Illite and Chlorite groups as shown on **Table 1**. He also indicated other classes which are mainly mixed layers of the four groups of clays. Descriptions by Hughes [34] are captured below:

- Kaolinite is composed of one silicate tetrahedral and aluminium octahedral thus a 1:1 clay mineral. This structure makes kaolinite relatively stable due to its low surface area and adsorption capacity.
- Smectites are composed of two silicate tetrahedrals bonded with one aluminium octahedral thus a 2:1 clay mineral. Smectites have a very high rate of expansion and/or shrinkage and are by far the most problematic clay minerals during drilling and production especially with water-base engineered fluids. This behaviour is attributable to the large surface area and high cation exchange capacities of smectites consequently leading to high adsorption capacity.
- Illite is composed of tetrahedral and octahedral plates arranged in a 2:1 format just like smectites. They however have lower adsorption capacities than smectites but higher than kaolinites.
- Chlorites consist of Brucite layers alternating with three-sheet pyrophyllite type layers. Though Chlorite may occur as macroscopic or microscopic crystal, they often occur as mixtures with other minerals in the microscopic state.

3.3 Clay mineral properties

Clay minerals are unique in a number of properties they exhibit; however, the following attributes of clay minerals have significant impacts on their interactions with fluids and are briefly captured below.

Clay Type	Chemical Formulae	Surface Area (m ² /gm)	CEC (meq/100 g)	Configuration
Kaolinite	Al ₄ [Si ₄ O ₁₀](OH) ₈	20	3–15	1:1
Illite	(K _{1–1.5} Al ₄ [Si _{7–6.5} Al _{1–1.5} O ₂₀](OH) ₄)	100	15–40	2:1 Non-Expandable
Smectite	(0.5Ca, Na) _{0.7} (Al, Mg, Fe) ₄ [(Si, Al) ₈ O ₂₀]•nH ₂ O	700	80–100	2:1 Expandable
Chlorite	(Mg, Al, Fe) ₁₂ [(Si, Al) ₈ O ₂₀](OH) ₁₆	100	15–40	2:1 Non Expandable

Table 1.
Four major types of clay minerals relevant to hydraulic fracturing.

3.3.1 Cation exchange capacity (CEC)

Cation exchange capacity (CEC) is defined as the amount of positive ion substitution that takes place per unit weight of dry rock [35] and is expressed in meq/100 g (milliequivalents per one hundred grams) of dry rock. Substitution of ions in minerals is the product of interfacial electrochemical interactions. Some of the most common cations exchanged are calcium (Ca²⁺), magnesium (Mg²⁺), potassium (K⁺), sodium (Na⁺) and ammonium (NH₄⁺). CEC controls contribution of clay minerals and clay-bound water to electrical conductivity of rocks as well as the wettability characteristics of clay minerals during clay-fluid interactions.

Researchers developed various methods of measuring CEC over the years with more accurate methods still being developed. Some of the earlier methods have been exhaustively discussed in literature [36–40]. The most common methods currently used for CEC determination include: wet chemistry method; multiple salinity method and membrane potential method. These are however not without their limitations.

Bush and Jenkins [40] developed a method based on the use of the wet chemistry method in which several samples were investigated and a plot of best fit generated. The main challenge with their method is that, some minerals are capable of adsorbing water in humid environments though they have no CEC. Bush and Jenkins [40] proposed their method as a supplementary method for the wet chemistry method rather than a replacement.

Cheng and Heidari, [41, 42] introduced a new theoretical model of measuring CEC based on energy balance between chemical potential and electric potential energy. This involved the combined analysis of data collected from XRD (X-Ray Diffraction), NMR (Nuclear Magnetic Resonance) and nitrogen adsorption–desorption isotherm measurements with direct evaluation of CEC based on ammonium acetate method and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) measurements used in cross-validation of the results. They however alluded to the fact that their method was yet to be developed for complex rock composition.

3.3.2 Clay swelling

Clay swelling results mainly from fluid intake into the inter-layered structure of clay minerals. Electrochemical interactions between clay minerals and fluids are central to the swelling of clays. The type, quantity and charge of cations in the interlayer zones of clay are the main driving forces in the swelling process. Clay swelling and formation damage during enhanced oil recovery have also been discussed extensively [43, 44].

Two main types of swelling mechanisms have been identified in clay minerals which include crystalline swelling and osmotic swelling [45, 46]. During crystalline or surface hydration mechanism, the water molecules are adsorbed on the crystal surfaces with hydrogen bonding holding the water molecules to oxygen atoms exposed from the crystal surface. Subsequent layers of water molecules align to form a quasi-crystalline structure between unit layers, which results in an increased c-spacing. This type of swelling is common to all types of clay minerals, although to a different degree. In osmotic swelling mechanism, the concentration of cations between unit layers in clay minerals is higher than that in the surrounding water, water is therefore osmotically drawn between the unit layers and the c-spacing is increased. Osmotic swelling mechanism causes a larger swelling relative to the crystalline swelling but only a few clay minerals, such as sodium montmorillonite, swell in this manner [47].

4. Hydraulic fracturing

Hydraulic fracturing entails high rate injection of pressurised fracturing fluids into low permeability formations often targeted at specific horizontal sections of a wellbore in order to induce failure, consequently fracturing rock formation and creating a fracture network that can provide permeability in otherwise almost-impermeable rocks. Studies have shown that the fractures induced by hydraulic fracturing fluids are formed normal to the direction of minimum horizontal stress in the horizontal section of the wellbore. Horizontal wells are normally drilled in trajectories parallel to the minimum horizontal stress in a given reservoir. However, branch-like networks of micro-fractures are formed in all directions, resulting in a hydraulic connectivity that provides permeability from otherwise impermeable shale matrix. The majority of fractures are kept open by proppants which are transported by the injected fluid into the formation. Proppants ensure that fractures remain open thus enhancing the contact area between reservoir and wellbore which consequently serve as a conduit for hydrocarbon recovery, from otherwise low permeability shales [48, 49].

Hydraulic fracturing entails lots of activities, thus, research is fine-tuned on investigating and understanding certain key issues about hydraulic fracturing. For example, Rikards et al. [50] indicated that one of the biggest problems in hydraulic fracturing has to do with ability to find balance between proppant-quality and proppant-transport efficiency. They intimated that high density proppants pose proppant transport challenges whilst low density proppants present issues of strength of the proppants. Also, the importance of fluid viscosity in terms of providing sufficient fracture width to enable transport and proper placement of proppants is another issue in hydraulic fracturing highlighted by Montgomery et al. [51].

4.1 Hydraulic fracturing fluids (HFF)

Since inception of the concept of hydraulic fracturing, a lot of fluids have been developed and experimented as possible suites for various formation types and even geographical locations. These are discussed below.

4.1.1 Water-based fracturing fluid

Water-based fracturing fluids are the most common hydraulic fracturing fluids in use today. This is due to their low cost, availability and their ability to transport

proppants in place to maintain fracture conductivity. Though water-based hydraulic fluids have several advantages over other types of fracturing fluids, they are more susceptible to causing formation damage due to hydration of clays which may lead to lower recovery rates for hydrocarbons. Ribeiro and Sharma [52] contend that water-based fracturing in unconventional wells, most of which contain substantial clay mineral component, presents significant challenges. One of the most effective ways of dealing with this drawback, thus, has been to use energised water-based fracturing fluids in which the fracturing fluid is energised with CO₂ or N₂. This significantly reduces the amount of water needed for fracturing and thus improves the fracturing job in water-sensitive formations. Some water-based fracturing fluid types are discussed below.

Slickwater fracturing fluids are primarily composed of water, sand proppants and other chemicals to deal with friction, corrosion, clay swelling and other adverse reactions due to injection of fluids into the subsurface. These fluids are characterised by lower viscosities and the ability to generate complex fractures which generally reach deeper into target formations. The drawback with this type of water-based fracturing fluid is its poor proppant transport capacity. This is often compensated for with higher pumping rates in order to maintain optimal velocities that prevent settling of proppants.

Linear fracturing fluids were developed as a solution to the poor proppant carrying capacity of slickwater fluids. This was achieved by increasing the viscosity of fracturing fluid through addition of polymers in the fluids. These polymers are capable of turning the aqueous solutions into viscous gels capable of transporting proppants effectively but may also adversely affect the permeability of low permeability formations by forming filter cakes on the walls of fractures. Linear fracturing fluids are good in controlling fluid loss in low permeability formations but prone to higher fluid losses in high permeability formations.

Cross-linked fluids were developed to obtain increased viscosity and performance of gelled polymers without necessarily increasing the concentration of polymers. To develop these fluids, Aluminium, Borate, Titanium and Zirconium compounds may be used to crosslink hydrated polymers in order to increase the viscosity of resulting fluid. The main advantage of these fluids is the reversibility of crosslinks based on pH adjustments. This enables better clean up and consequently improved permeability following fracturing treatment. Borate crosslinked fracturing fluids have been reported to show rheological stability, good clean-up and low fluid loss up to temperatures of over 300°F.

In viscoelastic surfactant gel fluids, increased viscosity and elasticity is obtained by adding surfactants and inorganic salts into water-based fracturing fluids to create ordered structures. These fluids exhibit very high zero-shear viscosity and are capable of transporting proppants with lower loading and without the comparable viscosity requirements of conventional fluids [53].

4.1.2 Oil-based fracturing fluid

Oil-based fracturing fluids are used mostly where a formation is water-sensitive perhaps due to the presence of large quantities of expandable clay minerals. Oil-based fracturing fluids have been found to better preserve fracture conductivity [54, 55] as well as provide better performance in terms of proppant transport due to the generally higher viscosity and lower specific gravity. In their work on wells located in Bakhilov and North Khokhryakov fields in Western Siberia, Russian Federation, Cikes et al. [54] studied the responses from wells after treatment with oil-based fracturing fluids in a depleted oil reservoir. These wells were initially fractured with water-based fracturing fluids but the treatment failed and did not yield

significant improvements in the productivity especially for the long term. Following fracturing with oil-based fracturing fluids, over ten-fold production improvement was witnessed relative to pre-fracturing productivity.

Another advantage of oil-based fracturing fluids as noted by Hlidek et al. [56] is that they are easier to clean-up and can be re-used. Hlidek et al. [56] compared the cost of using water-based fracturing fluids to oil-based fracturing fluids in the Montney (Canada) unconventional gas development. Based on their comprehensive analysis, they concluded that the cost of using oil-based fluids was lower in the long term since all the load oil could be recovered within 4 to 8 weeks and could be reused in fracturing. The main disadvantage of oil based fracturing fluids however, is environmental damage when not properly disposed of.

To enhance the efficiency and recovery of oil-based fracturing fluids, CO₂ has been employed in energising these fluids. Energising oil-based fluids significantly reduces the amount of fluid required to fracture a specific formation as well as aids in fluid recovery following the fracturing process [57, 58]. Vezza et al. [58] studied the impact of energised oil-based fracturing fluid in Morrow Formation in Southern Oklahoma where they used gelled diesel/CO₂ as fracturing fluid. Their results indicated an overall increase in production rate and predicted long-term stability of the wells. Gupta et al. [57] also reported improvements in well productivity and stability after using energised gelled hydrocarbons in fracturing treatments. In their study, they compared the use of conventional gelled fluids to CO₂ energised gelled fluids in formations in Canada. Their conclusions were that: The use of energised gelled hydrocarbon fracturing fluids led to improved production relative to conventional gelled hydrocarbon fluids.

4.1.3 Gas fracturing fluid

Gas fracturing involves the injection of gas at high pressures into the subsurface in order to create fractures within targeted reservoir locations. Nitrogen gas is the most employed gas for fracturing purposes, due to its obvious advantages of availability, its inert nature and of course, cost [59]. The main limitation of gas fracturing is the depth it can be used as a fracturing fluid since it has a low density and thus is restricted to reservoirs of less than 5000 feet deep [60]. Recent advancements in ultra-light weight proppants [50, 61] provides positive prospects that may counter the depth limitation of gas fracturing to some extent.

4.1.4 Foam-based fracturing fluid

Another type of fracturing fluid is a foam-based fracturing fluid which is generated from the combination of two phases of liquid and gas as well as addition of surfactant to ensure stability [51]. The main advantages of this type of fracturing fluid is its efficiency in water-sensitive areas and its relatively better proppant carrying capacity compared to water-based fracturing fluids [62]. High cost and risk of flammability are the main disadvantages of foam-based fracturing fluids.

4.1.5 CO₂-based fracturing fluid

Consideration of the use of CO₂ as a fracturing fluid was mooted due to problems encountered with water-based fracturing fluids in terms of permeability damage. Liquid CO₂ is considered an alternative to water-based fracturing fluids because it causes minimal formation damage plus clean-up is easily achieved. Lower viscosity, miscibility of CO₂ with hydrocarbons, ease of displacing methane from organic matter and the ease of recovery of CO₂ enables it to

create extensive and complex fractures at lower breakdown pressures [63, 64]. However, high pumping rates needed to enhance proppant carrying capacity of CO₂ makes CO₂-based fracturing fluids relatively expensive. Additionally CO₂ is not readily available at all sites. The future applications may change if CO₂ can be sequestered.

5. Mechanisms of clay minerals-HFF interactions

5.1 Hydration imbibition and fluid retention

Imbibition describes the displacement of immiscible fluids from within the formation matrix. In the context of this topic, the fluid within the formation matrix is hydrocarbon and brine, whereas the invading fluid is the hydraulic fracturing fluid, mostly water. The displacement described above occurs at times where the fracturing fluid comes into contact with the formation face creating disequilibrium. In order to gain equilibrium, fracturing fluid is drawn into the matrix spontaneously, without the application of any form of pressure. This phenomenon is known as spontaneous imbibition. Handy [65] defined it as the process in which a fluid is displaced by another fluid within a porous medium due to the effect of capillary forces alone. Other researchers like Bear [66], Bennion et al. [67], Hoffman [68] and Dutta [69] have also interrogated the mechanisms of hydration imbibition.

Imbibition of water into shale matrix has been identified as the major water retention mechanism when using water-based hydraulic fracturing fluids [70, 71]. Research into the controlling factors of imbibition of water in fracturing fluid into formation matrix revealed it to be the function of several parameters which are briefly discussed in the following:

5.1.1 Fluid and rock properties

Fluid and rock properties have been identified as determinants of the amount and rates of imbibition. Ma et al. [72] reported that when water displaces oil and gas within the formation, the rate of imbibition becomes directly proportional to the viscosity of water. Pore sizes of formation inversely affect imbibition since smaller pore sizes generate greater capillary pressures and thus higher imbibition.

5.1.2 Initial water saturation

The initial amount of water present in matrix of rocks has been investigated by several researchers to ascertain its impact on the quantum of imbibition, but findings have been inconsistent, thus making it difficult to draw any conclusions. Whereas Blair [73] and Li et al. [74] found that an initially high water saturation of formation led to lower volumes of imbibed water, Cil et al. [75] and Zhou et al. [76] found the opposite in their experiments. Other works by Li et al. [74], Viksund et al. [77] and Akin et al. [78] also concluded that initial water saturation had no effect on imbibition of water by the formation. They explained that volume of water imbibed is a function of capillary pressure and effective permeability but these show an inverse and direct relation with water saturation respectively. The amount of water imbibed is therefore not controlled by a single parameter, but will depend on which of the two variables is dominant in any given formation. In this regard, they concluded that the influence of initial saturation on imbibition should be ascertained for every formation independently.

5.1.3 Temperature

The impact of temperature on imbibition is not direct however; temperature of a formation has impact on wettability and fluid properties which subsequently impact imbibition. Experimental investigations by Handy [65], Pooladi-Darvish and Firoozabadi [79] concluded that higher temperatures led to faster rates of imbibition.

5.1.4 Clay content

Total clay content directly relates to the effect of pore size on rate and amount of imbibition. Due to small pore size of clay-rich rocks, higher clay content in a formation results in smaller pore sizes thus greater imbibition. This position is confirmed by Zhou et al. [80] who performed several experimental and numerical analyses on the Horn Shale gas formation and concluded that high clay content in a formation leads to high volumes and rates of imbibition respectively.

5.2 Osmosis

According to Zhou et al. [81], earlier researchers viewed imbibition to mainly be the product of capillary pressure but findings from recent studies have challenged this position. Recent research shows osmosis contributes significantly to water imbibition and thus clay minerals and hydraulic fluid interactions especially for unconventional reservoirs which are often characterised by high clay mineral contents.

During osmotic imbibition of water into formation, formation clay minerals act as semi-permeable membranes through which fracturing fluids invade the matrix of the formation. Here, solutes from the concentrated formation fluids try to move into lower solute fracturing fluids but due to the semi permeable membrane formed by presence of clay, the solutes are unable to cross this barrier. Continuous accumulation of solutes near the semi-permeable membrane creates an attraction force that draws water into the formation in order to balance out the concentration differential.

6. Geochemical and geomechanical impacts of clay mineral-hydraulic fracturing fluids interaction

6.1 Water-blocking effect

Inorganic clayey matrix is generally known to be water-wet therefore providing favourable conditions for imbibition of water from fracturing fluid within fractures. In this process, the invading water displaces gas from the surface of clay matrix which leads to the formation of a multiphase flow environment near the fracture surface (**Figure 2**). Development of this phenomenon can create an unfavourable saturation condition, under which gas flow through fractures is hindered, thus lowering yield for wells. The phenomenon is known as water-blocking and it has been described by researchers as one of the most severe damages in reservoirs with ultra-low permeability [82–84].

Recent experimental works on the imbibition of water by shale rocks showed that the imbibed water remains within the pore network, thus reducing the permeability to gas of the reservoir [81]. Simulation and history matching also confirmed that invasion and wetting of clay mineral surfaces by water from fracturing fluid

Figure 2.
Fracture and near-fracture clay-fluid interactions (adapted from [103]).

was responsible for decline in gas production. Reduction in gas flow due to water blocking effect has also been reported by Shanley et al. [85], who observed drastic reduction in gas production when water concentrations in fractures exceeded 40–50%. Detailed study of water-blocking phenomenon has showed that this phenomenon may cause permanent damage for some shale formations while the damage is transient for other shale types [70, 71, 86]. The details of mechanisms and variables that determine whether damage is temporal or permanent are still being investigated.

6.1.1 Water-blocking effect as a transient effect

Water-blocking during fracturing of unconventional reservoirs is explained by the presence of two pore types in unconventional formations. The first pore types are the larger oil-wet pores located within the organic matrix of the formation. The second type of pores are the smaller water-wet pores located within the inorganic argillitic matrix. Pore throats of the larger oil-wet pores are however small. During hydraulic fracturing, high pressure fluids break the formation to form fractures with some fracturing fluids leaking off into near-fracture matrix. Once in the matrix, the fluids first occupy the larger oil-wet pores. However, due to smaller pore throats, fracturing fluid in the formation is segmented within each internal pore with minimal linkage to other pores. This causes water to be domiciled in formation as droplets filling larger oil-wet pores which subsequently makes remobilisation difficult upon resumption of production. This phenomenon significantly reduces hydrocarbon effective permeability. The natural healing process in this phenomenon occurs when fluid is drawn from larger pores into smaller water-wet pores deeper within the reservoir thus dissipating the water blocking effect. This leads to improved permeability and hydrocarbon production [71].

6.2 Mineral dissolution and precipitation

Clay minerals and non-clay minerals (carbonates and quartz) within a formation are susceptible to geochemical attack from the fracturing fluids. Most shale formations were deposited in sea water-rich environments and have established equilibrium of their minerals and fluids over geological time. Once these formations are exposed to engineering fluids, especially water-based fluids, the

geochemical equilibrium is no longer stable. Subsurface temperature, pressure, and pH often enhance geochemical reactivity of scale-forming minerals, resulting in changed porosity, and fracture permeability as a result of mineral dissolution and precipitation [87, 88].

Dissolution of rock forming minerals has been reported at low pH. As pH increases, ions from dissolved minerals recrystallise to form new minerals and/or amorphous precipitate that may have an adverse effect on formation permeability. At very high pH, clay minerals within a formation become unstable and may become mobile. This situation leads to migration of illite samples which may occlude the hydrocarbon flow paths within the formation (**Figure 2**).

6.3 Shale swelling

Shale swelling during hydraulic fracturing results from swelling of clay minerals within fracture face and shale matrix (**Figure 2**). Three mechanisms have been noted to cause clay mineral swelling as water is adsorbed into nano-pores, micro-pores, meso-pores and even macro-pores of clay minerals in the formation. The first mechanism is swelling due to hydration of negatively charged clay surfaces with several water layers depending on the type of clay. This has been observed in various types of clays with different levels of water saturation [89, 90]. The second mode of swelling is similar to what causes imbibition of water into clay minerals, where the clay acts as a semi-permeable membrane. In this case water is moved into the inter-layer spaces of clay minerals causing massive swelling [91]. In the third mechanism, continuous expansion of clay interlayer leads to separation of the clay layers into different clay components thus transforming initially intact clay layers into inter-particle spaces [92, 93].

Swelling of clays within the fracture walls can lead to constricted apertures which severely restrict flow of hydrocarbons during production [94]. Clay swelling may also induce micro fractures in formations which may improve absolute permeability [95, 96].

6.4 Stress development

During fracturing, interactions between shale and invading fluids lead to swelling of clay minerals within the shale causing in-situ stress development. Osmosis has been suggested as the potential transport mechanism which causes swelling pressure build-up within shale rock. The reaction between clay minerals and invading fluid is observed to be a primary cause of damage to reservoir permeability during and after hydraulic fracturing. Previous experiments have shown that a change of permeability impairment following fluid interaction with formation is directly related to the specific clay mineral content of the formation [31, 71, 97]. In the case that the solute concentration in fracturing fluid is significantly lower compared to concentrations in clay interlayer, osmotic swelling is likely to occur, where fluid is drawn into the interlayer with the aim of balancing the solute concentrations. This phenomenon leads to significant expansion of the clay minerals. Expansion of clay minerals therefore exerts pressure on surrounding pores and matrices thus leading to a build-up of stress.

6.5 Mechanical weakening

Geo-Materials mechanical properties are dictated by the amount of pore space present, compositional heterogeneities [98], solids (inorganic and organic) mechanical strength and the presence/absence of pore-fluids and

their composition. Therefore, mechanical weakening of formation rocks due to reaction with newly introduced fracturing fluids has been observed by a number of researchers. Akrad et al. [99] observed that sustained interaction between fracturing fluid and formation can induce softening of the formation rock thus reducing the Young's Modulus of the rock, therefore producing mechanical weakening. Du et al. [98] investigated mechanisms of fracture propagation due to hydraulic fluid injection and concluded that the mechanical response of formations due to interaction with fracturing fluid is directly linked to the mineral composition and geochemistry of rocks. In their studies of proppant embedment efficiency, Corapcioglu et al. [100] found that exposure of the formation to fracturing fluid leads to decrease in Young's Modulus of the rock. Research on the impact of fracturing fluid on formation mineralogical components has also showed that non-clay minerals (carbonates, quartz, feldspar and various sulfides/sulphates) as well as clay minerals, like chlorite, are susceptible to dissolution in fracturing fluids, leading to a reduction in the structural strength of the formation.

LaFollette and Carman [101] reacted Haynesville shale samples in fracturing fluid at temperature of about 300°F for periods of 30, 60, 120 and 240 days respectively and observed changes in Brinell Hardness of the samples. The highest reduction occurred between 60 to 120 days after which there was a marginal increase in hardness. Carman and Lant [102] also reacted rock samples with different fracturing fluids at temperatures close to subsurface formation temperatures. Their results showed that Brinell's Hardness for all the rock samples decreased after reacting with fracturing fluids.

6.6 Impact of geochemistry of shale-HFF reactions on toxicity of produced water

Flowback water is the fluid produced immediately following treatment of a well. This fluid is generally made up of mixed compositions of fracturing fluids, products of reactions between fracturing fluids and formation, and formation fluids. High salinity and concentrations of dissolved metals have been reported in flowback waters [104, 105]. Potential toxicity of produced water to humans and the environment remains high and of great concern; therefore, researchers have been studying these fluids to assess their risks to the environment. Most studies of this nature have largely been undertaken in producing unconventional hydrocarbon fields in the USA as well as black shales from Germany and Denmark [106, 107]. A summary of these works is presented below:

Chapman et al. [104] sought to characterise the flowback waters from Marcellus Formation in the Appalachian Basin with strontium isotopes in order to help detect and trace contamination of surface and ground waters by flowback water. The geochemical characterisation of the flowback fluids showed elevated levels of Bromide, Calcium, Strontium (up to 5200 mg/L), Sodium, Chloride (up to 12000 mg/L) and Barium. They also reported high total dissolved solids in the fluids in excess of 200000 mg/L. They concluded that high elemental concentrations were the result of interactions between fracturing fluids, formation minerals and formation fluids.

For their part, Wilke et al. [107] studied the rate of release of metals, salts anions and organic compounds from shale rocks in Denmark and Germany. They reported that, concentration of ions in solution largely depended on the composition of the black shale and did not show dependency on pH of the fluids used in experiments. There was however a correlation between the buffering capacity of specific mineral components of the rock, such as pyrite and carbonate on amount of dissolved ions. They also reported decrease in ionic concentrations over time due to precipitation

of new minerals. Findings from this research provided an understanding of possible flowback water composition and toxicity from these unconventional fields in Denmark and Germany respectively.

Experimental studies by Macron et al. [106] on geochemical interactions between fracturing fluid and formation showed evidence of clay and carbonate dissolution as well as precipitation of new minerals. Dissolution leads to elevated elemental concentrations in fluids some of which show a drastic reduction when precipitation of new minerals begins. Results from this experimental work were validated using measured concentrations of sodium chloride (NaCl) in Marcellus shale which showed similar trends. They concluded that results from their work can be used to form a basis of assessing controls of geochemical reactions in the reservoir, in other words, flowback water compositions.

7. Summary of advances and research gaps topics for future consideration

In this section, recent research advancements are summarised. Review of recent studies shows that geochemical and geo-mechanical impacts from interactions between clay minerals and/or formation and fracturing fluids are being assessed more closely to help solve problems associated with reduced permeabilities during post-fracturing flowback. Geo-mechanical response of formations due to differences in temperatures of fracturing fluid and subsurface formations have also become a focus area for researchers. These advancements have unlocked new areas of research which will be explored in the near future. Other researchers have also focused on developing methods to measure the extent to which geochemical and geo-mechanical impacts are controlled by certain mechanisms during interactions between formations and fracturing fluid. Recent studies assessed for the purpose of this review are as follows:

7.1 Fracture face damage

Though water-blocking effect is known to be one of the causes of permeability loss following hydraulic fracturing, mechanisms by which this occurs are not well understood. Elputranto et al. [108] simulated this phenomenon to study the main forces that drive it. They concluded that the fundamental driver of high water saturations held near the fracture-matrix interface may be due to capillary end effects. These act near the interface between fracture and formation matrix to increase the water saturation beyond the saturation caused by imbibition. Elputranto et al. [108] therefore suggested that capillary end effect is a significant mechanism that must be considered when assessing potential of water-blocking effect in a formation. Future research may focus on experimental validation of this simulation work.

In order to effectively diagnose the predominant mechanism of face damage in fractures in tight sands, Li et al. [109] proposed a new experimental method. In their work, two mechanisms are suggested as mostly being responsible for fracture face damage; high capillary pressure and swelling of water sensitive clays. Li et al. [109] integrated pressure transmission and pressure decay methods to determine the predominant cause of fracture face damage. They concluded that their method is able to distinguish the cause of the key mechanism in fracture face damage. Though the method was effective in tight sand, it has yet to be tested on shales and other unconventional reservoir rock samples with high clay mineral content. Future research should focus on investigating the scope of application of this method for shale formations and other unconventional reservoir rocks with high clay compositions.

7.2 Fines migration

Fine particle migration is a major cause of fracture aperture blocking yet a very difficult phenomenon to study. Muggli et al. [110] introduced a simple, time saving experimental method to assess particle migration potential for different fracturing fluid compositions. They premised their method on the fact that behaviour of fine particles in the subsurface is a function of fracturing fluid composition. In their method, turbidity, capillary suction time behaviour and particle size distribution relative to the time and depth of particles are observed and recorded. Results from observations are then used to draw conclusions on the potential of particle migration and pore throat blocking. They tested their method using Eagle Ford Brine with and without additives. Low turbidity and capillary suction time were observed for brine without additives while higher values were recorded when additives were used. They explained that high turbidity was due to the inhibition of flocculation caused by the additives. Low capillary suction times therefore, may not always be desirable. In conclusion, they indicated that this experiment can be repeated using other fracturing fluid compositions to determine their impact on migration of fines.

7.3 Mineral dissolution and precipitation

Significant geochemical reactions are expected to occur during the shut-in period when fracturing fluid is in contact with formation. Wang et al. [111] studied rock-fluid interactions during the shut-in period to assess the water chemistry over the period using 15% HCL and water. They also assessed the possibility of scale formation in the reservoir based on these reactions. Their results showed ability of fracturing fluid to react with and dissolve formation minerals to increase permeability. However, these reactions also lead to release of ions into solution which may cause precipitation of scale-forming and permeability reduction minerals. Wang et al. [111] proposed their study as a way of understanding the long-term implications of rock-fluid interactions. Future research to expand the frontiers of this work should consider using different fracturing fluid compositions.

Furthermore, the impact of microstructural and geochemical interactions between fracturing fluids and fracture face in organic rich carbonates was studied by Liang et al. [112]. They concluded that 2% Potassium Chloride, though used to inhibit adverse reactions between fluids and clays minerals, may in fact increase the dissolution rates of carbonates thus increasing absolute permeability. This increase was observed to be more pronounced for slickwater relative to synthetic sea water. Their study is evidence that water-based fracturing fluids could be beneficial when used in some formation types such as organic-rich carbonates. Future research in this area should therefore be focused on understanding the mechanisms that cause faster dissolution in presence of Potassium Chloride.

7.4 Changes in mechanical properties

Juan et al. [113] investigated the relative impacts of slickwater and linear gel on mechanical properties of different rock types in the Permian basin. Reactions for these set of experiments were conducted at elevated temperature and pressure conditions, 190°F and 1000 psi respectively. Their findings indicated that: linear gel caused more mechanical (Young's Modulus) reduction, about 27% compared to slickwater, about 14%; Samples with higher contents of carbonates sustained more damage relative to low carbonate samples, with carbonate etching being the primary damage mechanism in slickwater whilst that in the

linear gel is aggressive dissolution; Most carbonate dissolution happened within the first five hours of the reaction. This experiment provides critical information on reactions between rocks and fluids at elevated temperatures and could be repeated for other types of fracturing fluids in different formations to observe the responses.

Temperature differences between injected fracturing fluids and formation have been reported to exert significant mechanical impacts during fracturing. This observation has gained attention and has become focus area for researchers. Vena et al. [114] studied the impact of large temperature differentials between formation and invading fracturing fluids. They took particular interest in changes such as clay swelling, imbibition and other mechanisms that adversely affect formation permeability. Their results indicate an initially pronounced impact on stress regimes within the formation leading to development of micro-fractures which are sealed over time. Similar findings were obtained by Elputranto et al. [115] when they used high resolution simulation methods to assess the response of formation to fluid with high temperature and salinity differentials to formation. Since perpetual propagation or opening of these micro-fractures will greatly enhance permeability of a reservoir, future research should be focused on understanding the mechanisms that can sustain these micro-fractures.

Elputranto et al. [115] used high resolution simulated models to investigate the mechanical impact on the interface between hydraulic fracture and matrix due to reactions emanating from cold and low salinity fracturing fluid invading rock formation. They simulated the responses during the well shut-in period and flowback and production periods. Their results show that thermo-elastic effects are generated in the formation that lead to increased permeability which is short lived. Based on results from this work, future research will focus on how to sustain and possibly allow better propagation of these short-lived fractures created due to thermo-elastic effects of fracturing fluid interaction with formation. Achieving this will lead to significantly improved permeability and production.

7.5 Alternative fracturing fluids

Li et al. [116] conducted an experiment on the use of CO₂ as pre-fracturing fluid during hydraulic fracturing in tight gas formations. They aimed to confirm that the combination of CO₂ and water during hydraulic fracturing operations could help harness benefits of both fluids especially at locations with low water availability. This research was undertaken for subsequent application in the Loess plateau of Ordos basin in China. Results from experiments showed improved permeability due to dissolution of carbonates and clay minerals by CO₂. They also found that CO₂ interacts with hydrocarbons and provides additional impacts in terms of improving hydrocarbon properties to enhance relative permeability, thus providing increased productivity. More experiments should be conducted to ascertain the optimum use of CO₂ and water combinations for fracturing.

Adverse environmental footprints of hydraulic fracturing operations have also necessitated research to find innovative ways of mitigating these impacts. Ellafi et al. [117] investigated the possibilities of re-using produced water as fracturing fluid. They justified their research by drawing attention to the large volumes of fresh water used in hydraulic fracturing operations. Some helpful statistics quoted to buttress their points include the following: Texans waste about 2% of water demand on fracturing jobs [118]; the amount of water withdrawn from the Missouri River for hydraulic fracturing in 2018 alone, was about 1.269×10^{10} gals, an estimated 10.1% of North Dakota water consumption [119].

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