Journal of Petroleum Science and Engineering 206 (2021) 109057

Contents lists available at ScienceDirect

Journal of Petroleum Science and Engineering

journal homepage: www.elsevier.com/locate/petrol

Emulsions in porous media from the perspective of produced water re-injection – A review

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ARTICLE INFO

Keywords:
Emulsion
Produced water
Permeability
Injectivity
Formation damage
Pore clogging mechanisms

ABSTRACT

During offshore production, it is typical to discharge produced water into the sea after treating it to a level that meets environmental regulations. A growth in the volumes of produced water and increasingly stringent requirements have drawn attention to re-injection of produced water as a more viable disposal method. Produced water re-injection is regarded as an environmentally friendly and cost-efficient approach that allows integration of produced water with reservoir management. However, loss of injectivity, due to the plugging of pores by oil droplets and particles present in produced water, limits implementation of re-injection. This review discusses engineering challenges during re-injection and summarizes the knowledge and the gaps in the literature on the permeability reduction due to the flow of dilute oil-in-water emulsions. First, a short introduction into the origins of produced water, its composition, and the treatment techniques employed offshore are provided. An overview of the water injection strategies as well as a discussion of the past field experience of produced water re-injection is given. This is followed by a detailed review of the flow of diluted oil-in-water emulsions through porous media from the permeability reduction perspective. An overview of models for predicting permeability reduction during the flow of emulsions is provided. The physical parameters affecting the droplet retention and physicochemical interactions between droplets and pore walls are discussed. The studies that investigated co-injection of oil droplets and solid particles are included as well. The review identified that the effect of droplet stability, oil viscosity, wettability, and particles on droplet retention is not sufficiently examined. It was found that drop-to-pore size ratio is a crucial factor for droplet retention, but it is often omitted in the industry. An outlook on the gaps and suggestions to address them was provided. Microfluidics was pointed out as a complementary technique to coreflooding.

1. Introduction

The recovery of oil and gas from underground is often accompanied by production of large volumes of water. Produced water (PW) is a by-product that needs to be disposed with a minimal process and environmental footprint, and in a cost-effective manner. There is a large variety of dispersed and dissolved components of both process and reservoir origin present in PW that can have a negative environmental impact. The amount of PW depends on the structure of a reservoir, recovery stage, water injection strategy, well type and well completion type (Fakhru'l-Razi et al., 2009). Worldwide PW volumes increase as the producing oil fields mature. Nowadays PW makes up in average 70% of total production or, in other words, almost 4 barrel of water per 1 barrel of oil (Dudek et al., 2020).

Typical management of produced water in offshore environment is either discharge into the ocean after sufficient treatment or injection back into the reservoir or another suitable subsurface formation. On the Norwegian Continental Shelf (NCS), more than 75% of PW is discharged into the ocean (Norsk Olje and Gass, 2017). Treated PW must meet environmental regulations regarding its quality before it can be discharged. On NCS, regulations require that the amount of dispersed oil in PW must be less than 30 mg/l when discharged in order to diminish the damage to marine environment.

As the water cut increases from year to year, processing facilities on existing offshore platforms may need to be modified to have enough capacity for PW treatment, which increases the cost of discharge. Additionally, oil companies operating in Europe are pushed by regulatory authorities to decrease the oil content of discharged water from 30 mg/l to 15 mg/l (European Commission, 2019). Nowadays, some companies consider the re-injection of PW back into reservoirs as a more

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https://doi.org/10.1016/j.petrol.2021.109057
Received 28 January 2021; Received in revised form 26 May 2021; Accepted 1 June 2021
Available online 3 June 2021

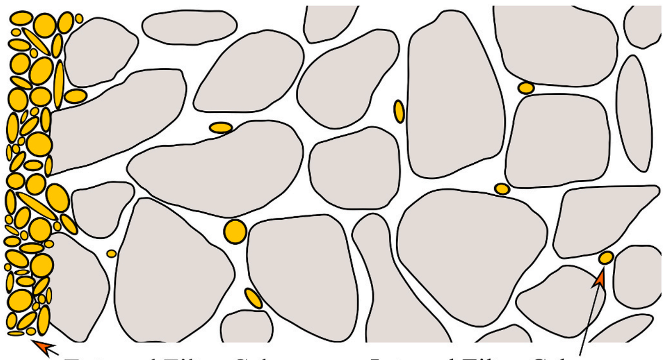

Fig. 1. Conceptualized process of formation damage. Based on Pang and Sharma (1997).

environmentally friendly and potentially cost-effective way of PW management. Produced water re-injection (PWRI) is often considered to be the base case for new fields, as zero discharge is the desired goal of regulatory authorities (European Commission, 2019). Moreover, it allows to incorporate produced water and reservoir management, e.g., pressure support of a reservoir and optimization of the sweep (Bedrikovetsky et al., 2011).

However, implementation of produced water re-injection entails several challenges. Even after the treatment, PW contains particles and droplets which can clog porous media in the near wellbore region when re-injected and cause formation damage (Sharma et al., 1997). Therefore, PWRI can be associated with a risk of unacceptable injectivity decline. Other phenomena associated with produced water re-injection are reservoir souring resulting from microbial activity, scaling and corrosion (Zuluaga et al., 2011). The reservoir souring and scaling can contribute to the injectivity decline; however, unlike for plugging, they can be evaluated, controlled, and mitigated. Maintaining the injectivity of the wells and good sweep is the key to successful PWRI projects. The design of topside and/or subsea facilities for the water injection system happens at the planning stage of the field development. It can be very costly to make or difficult to implement changes to the injection system later in the field life (Palsson et al., 2003). Moreover, contingency planning must be made for the scenarios when PWRI is not available due to injectivity decline, excessive fracturing, mechanical failures, etc. (Evans and Robinson, 1999). It is common practice to treat produced water to at least 30 mg/l of oil concentration (unless better quality is required) to comply with discharge regulations in case of downtime on PWRI system. Good understanding of the formation damage mechanisms by produced water components is required to define injection water quality specifications for cost-effective produced water re-injection. Specifications of water quality and PW treatment system must be determined individually for every oil field as the quality of produced water and reservoir properties depend on the geological formation of the reservoir (Evans, 1994).

For decades, seawater (SW) has been the most widely used source of injection water in the offshore environment. Therefore, causes of injectivity decline during SW injection has been widely investigated. Water quality specifications and treatment techniques to minimize formation damage during SW injection are well developed. The process of formation damage is illustrated in Fig. 1. Shortly, during the injection, suspended particles build an internal and external filter cake on the formation face (Pang and Sharma, 1997). There are several models describing transport of suspended solid particles through porous media with the deep bed filtration model believed to be the most robust (Tolmacheva et al., 2019). The principal difference between SW and PW with regards to formation damage is the residual oil concentration present in produced water. It was experimentally shown that the formation damage caused by droplets can be as severe as the one caused by particles, while the damage caused by droplets and particles co-injected was even more prominent (Ochi and Oughanem, 2018). Another analysis of PWRI showed that in some cases oil content was the main contributor to the damage (Rossini et al., 2020).

In the oil and gas industry, the transport of droplets in porous media is an engineering concern during produced water re-injection and enhanced oil recovery (EOR) by means of emulsions. Therefore, the phenomenon was studied by researchers working on both issues, mainly on emulsions EOR. The studies were conducted separately and, thus, there is a limited intelligibility of data. Therefore, to this date there is a lack of understanding of the droplet retention mechanisms for produced water considering the complexity that arises from its components, e.g., particles, crude oil. Simulations of deformable drop motion in porous media and granular materials was reviewed by Zinchenko and Davis (2017). Perazzo et al. (2018) reviewed the flow of emulsions through porous media with the emphasis on the enhanced oil recovery. The aim of this paper is to review the droplet retention in porous media and point out the gaps in the fundamental knowledge in the perspective of injectivity decline during PWRI. Section 2 of the paper presents the origins of PW, its composition and formation damage caused by the components, and treatment methods. Section 3 presents field experience of PWRI reported in the literature to provide an understanding of the water quality specifications and criteria considered in the industry for re-injection projects. Section 4 of the paper covers models of permeability reduction caused by emulsions, factors affecting droplet retention, and experimental techniques utilized in the literature to study the flow of emulsions through porous media. Gaps in the knowledge of droplet retention and an outlook are presented in Section 5.

2. Produced water

2.1. Origins of produced water

Typically, reservoirs comprise a gas zone (gas cap) on the top, an oil zone in the middle, and a water zone (aquifer) at the bottom. Production of hydrocarbons takes place in three stages: primary recovery, secondary recovery, and tertiary recovery. Primary recovery relies on the natural flow of oil through wells due to the pressure difference between the reservoir and the wellhead. Usually, up to 10–15% of original oil in place (OOIP) is recovered during the primary recovery stage before pressure depletes, and additional support is required to maintain economically feasible production rates. In the secondary recovery stage, treated SW and/or PW or gas (sometimes combined) is injected into the reservoir to maintain the pressure and drive the oil production. Around 35–50% of OOIP is recovered at this point, and in order to produce more oil, various improved/enhanced oil recovery (IOR/EOR) techniques are implemented – tertiary recovery stage.

The aquifer is a source of energy for oil recovery in the natural water drive mechanism. The decrease of reservoir pressure due to oil production causes aquifer water to expand and flow into the reservoir, displacing oil towards production wells as the water-oil contact rises. This eventually leads to water breakthrough and production of water along with oil and gas (Bailey, 2000). Furthermore, insufficient sweep efficiency due to viscous fingering (Homsy, 1987) and permeability anisotropy of the reservoir can cause an early breakthrough of water into production wells during waterflooding operations (Bailey, 2000). Therefore, PW comprises both produced formation water and back-produced injection water. The amount of water produced increases as the wells age and water cut (total water to produced fluids volume ratio) can reach up to 95% (Kaur et al., 2009).

2.2. Produced water components

This section lists components of PW and briefly discuss the potential formation damage of each component (Dudek et al., 2020; Fakhru'l-Razi et al., 2009):

2.2.1. Dispersed oil

Crude oil is present in PW in the form of micron sized droplets. The pressure drop across the chokes and valves in the production pipeline provides a lot of energy and creates shear forces in the system, which mixes oil and water. The presence of oil droplets in PW constitutes the principal difference between SW and PW injection specifications. A detailed review of the permeability reduction due to retention of oil droplets is presented in Section 4.

2.2.2. Dispersed solids

PW can contain fine clay, sand grain, etc., mobilized by hydraulic drag forces and large flow gradients near wellbore. In addition, corrosion and scale products from the production facilities, as well as dead microorganisms are present in PW. Several authors have suggested that formation damage caused by suspended particles in the presence of oil droplets is more severe than by solid particles alone (Ochi and Oughanem, 2018; van den Broek et al., 1999). However, the underlying mechanism is not well understood (Section 4).

2.2.3. Dissolved organics

Benzene, toluene, ethyl benzene and xylenes (BTEX), polycyclic aromatic hydrocarbons (PAH), naphthenic acids, have partitioned from oil into the water over millions of years, causing its toxicity. Moreover, partitioning happens during the production of fluids, as the temperature or pressure changes. To the best of our knowledge, there were no reports that dissolved organics directly cause formation damage. Potentially, dissolved organics could change the wettability of rock and influence plugging of pores by oil droplets and particles.

2.2.4. Dissolved minerals

Cations such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Ba^{2+}, Sr^{2+}, Fe^{2+} and anions such as Cl^-, SO_4^{2-}, CO_3^{2-}, HCO_3^- make up PW chemistry. Additionally, traces of heavy metals, for example, cadmium, chromium, copper, and naturally occurring radioactive materials (NORM) are present in PW. The formation of scales is associated with sulfate and carbonate ions. Deposition of carbonate-based scales in porous media happens due to pressure, temperature, and pH changes on the way to the formation. When SW supplements PW during re-injection, sulfate-based scaling can occur if there is physicochemical incompatibility between SW and formation water. Precipitation of scales can significantly reduce the permeability of porous media (Mahmoud et al., 2015).

2.2.5. Bacteria

Sulfate-reducing bacteria (SRB) can be present in PW. Additionally, other kinds of bacteria can be introduced into PW if mixed with SW for injection. Hsi et al. (1994) showed that PW containing bacteria caused significant permeability reduction of core plugs. Bacteria clog the pores in a different way than particles; biofilms formed by bacteria in response to shear stress physically adsorb on the surface of the pores and partially or entirely restrict flow.

2.2.6. Schmoo

The viscous tar-like substance called "schmoo" is formed as the result of agglomeration of organic and inorganic components of PW, including production chemicals. The reader is referred to Eroini et al. (2015) for a more detailed overview of schmoo. Schmoo is known to cause severe plugging of injection wells if PW is poorly treated before the injection. Removal of schmoo is an extensive and complicated process (Bader, 2007).

2.2.7. Production and EOR chemicals

Injection of EOR chemicals, such as surfactant and polymers, improve the recovery; however, when back produced, they add to the complexity of the PW composition. Moreover, a large variety of chemicals are added into the oil production system to maintain its normal operation and improve the separation process: scale, corrosion, wax, asphaltenes, hydrate, and bacterial growth inhibitors, flocculants, emulsion breakers and antifoam chemicals. Production chemicals are often surface-active compounds and can indirectly affect the plugging of injection wells, e.g., corrosion inhibitors, by promoting the formation of schmoo (Ly et al., 1998). Their potential effect on formation damage by oil droplets has been reported in the literature (Coleman and McLelland, 1994). It has also been reported that adsorption of scale inhibitors can cause damage to core plugs (Jordan et al., 1994).

2.2.8. Dissolved gas

Gases such as CO_2, O_2, and H_2S can also be present in PW. The presence of oxygen in PW can be avoided if it is managed in a closed system; thus, the formation of iron particles due to corrosion problems can be reduced. Carbonate- and sulfate-based scale deposition during PWRI can be related to CO_2 and H_2S dissolved in PW as these gases cause pH changes with pressure and temperature. Moreover, CO_2 and H_2S are known to be highly corrosive, which can increase the solids content of the injected PW, e.g., iron sulfide, iron carbonate, or iron oxide.

2.3. Produced water treatment

Selected PW treatment techniques that are commonly used are briefly discussed in this section. Only the most common produced water treatment technologies used offshore are included, i.e. gravity separation, hydrocyclone treatment, gas flotation, media and membrane filtration (Judd et al., 2014). The reader is referred to the following reviews for comprehensive overview of the existing technologies. (Fakhru'l-Razi et al., 2009; Igunnu and Chen, 2014; Jiménez et al., 2018).

All fluids produced from the reservoir flow from production wells to processing facilities where they are separated into three phases: gas, oil, and water. First, the produced fluids undergo gravity separation typically in a gravity separator train. Afterwards, water from separators is diverted into its respective treatment stage. The treatment is usually divided into three steps: primary and secondary treatment, and a water polishing step. The water stream, before it enters the produced water treatment (PWT), can contain up to 1000 mg/l of oil-in-water and 350 mg/l of suspended solids (Mueller et al., 1997). Most of oil droplets are in the range of tens of microns and can be as large as 100–150 μm (Arnold and Stewart, 1999), while solids usually do not exceed 50 μm (Rawlins, 2013).

Hydrocyclones are commonly used at offshore facilities as a primary PWT method. They remove oil drops dispersed in the water, with the help of centrifugal force, down to 5–20 μm (Arnold and Stewart, 1999; Judd et al., 2014). Another primary PWT method is the use of skimmers. Gas flotation is typically a secondary PWT approach that utilizes attachment of gas bubbles, often dispersed into the system, to remove oil droplets. The requirements associated with offshore environment, such as space, weight, sensitivity to motion, simplicity of operation, etc., led to the development of the compact flotation unit (CFU). The CFU technology applies flotation and centrifugal force to accelerate the separation process. The performance of gas flotation units may allow to reach residual oil concentrations below the discharge limit (Piccioli et al., 2020). The gas flotation units are suitable for removal of droplets larger than 10 μm and down to 10–25 mg/l concentration (Judd et al., 2014; Saththasivam et al., 2016). Media filtration (nutshell filter) offers a very effective oil and particles removal. The downside of media filtration is the need for regeneration every few hours (Judd et al., 2014). Membrane technology has also been proposed as a method for secondary PWT (Dickhout et al., 2017). However, the risk of reduced performance due to clogging of membranes by oil droplets, particles and biofouling still hampers implementation (Baker, 2012; Guo et al., 2012; Ng and Kim, 2007; Shi et al., 2014). The secondary stage can be followed by a water polishing step where various techniques are utilized to completely remove oil droplets and some dissolved organic components (Fakhru'l-Razi et al., 2009); however, these techniques are mostly used

Table 1
Summary of droplet sizes and concentrations range after various treatment stages (Arnold and Stewart, 1999; Judd et al., 2014; Mueller et al., 1997; Saththasivam et al., 2016).

Technology	Typical Oil concentration ranges (mg/l)	Typical droplet size ranges (μm)
Gravity separation	100–1000	100–150
Hydrocyclones	20–80	5–20
Gas flotation	10–25	10–25
Media filtration	2–5	2
Membrane filtration	~0	<1

at onshore water treatment facilities. The droplet sizes and oil concentrations after the main technologies as well as media and membrane filtration are summarized in Table 1.

3. Produced water Re-injection

3.1. Water injection strategies

Injection of water into the reservoir can be performed under two conditions: *matrix injection* and *fracture injection*. The decision on what strategy to follow depends on the goals of water injection, reservoir properties and associated risks. Matrix injection assures optimal efficiency considering the heterogeneity of the reservoir, which is important to optimize the reservoir drainage (Tipura et al., 2013). On the other hand, it is dominated by particle plugging of the formation (Todd et al., 1984), causing severe injectivity decline. Fracture injection provides a way to mitigate the matrix plugging and gives higher long-term injectivity (Clifford et al., 1991). Fracture injection allows poorer quality of the injection water, which reduces the costs of water treatment. On the other hand, there is a possibility that poor quality of water will cause excessive fracture growth. This can compromise the sweep depending on the well configuration (Clifford et al., 1991), and still costly water treatment will be required to prevent this.

3.2. Field experience of injectivity loss

A short review of PWRI projects in the North Sea before 1993 was done by Evans (1994). In this section, some of the recent PWRI experience around the globe is presented to showcase parameters that are considered in the industry when preparing for and during re-injection activities. The presented cases are examined because they report variety of information about the injection planning and execution from the angle of suspended solids and oil. Analytical tools for gauging injection well performance are not in the scope of this paper and will not be reviewed. For an overview of injection well testing and well injectivity analysis the reader is referred to Dunn-Norman and May (1997) and Rossini et al. (2020).

Tipura et al. (2013) reported PWRI under matrix conditions into a homogeneous, highly permeable reservoir at the Grane field in the North Sea. The PW contained 18–65 mg/l of oil-in-water (OiW) and 2–9 mg/l of total suspended solids (TSS). Prior to the injection, a measurement of particle sizes, leak-off test, and numerical models indicated a limited potential for a filter cake build up and fracturing. Injectivity of the well started to decrease several weeks after the injection commenced; consequently, the injection rates were reduced to stay below the fracture pressure. For the next two years, the injectivity continued to drop but then stabilized, which was explained by some pressure peaks during the injection, which might have caused fractures around the wellbore.

Mainguy et al. (2019) reported PWRI under matrix conditions into high quality Miocene reservoirs of two oil fields offshore Angola. The OiW concentration of the re-injected PW was on average 18 mg/l for field A and 57 mg/l for field B. The TSS of the re-injected PW was 29 mg/l and 34 mg/l for fields A and B, respectively. The injection water was a combination of PW and ultra-filtered SW. The injectivity declined once PWRI started and varied depending on the injected PW-to-SW ratio. The variation could be associated with temperature changes of the mixture when the SW share increased, which induced thermal fracturing. An injection of chemicals improved the injectivity to some extent, but it declined once PWRI was restarted. Improvements were more significant at field A than at field B, which was related to the fact that the quality of PW is better at field A. Overall, the authors claim that "cleaner" injection water could wash away retained particles and droplets. On the other hand, the injectivity gains became smaller and smaller as PWRI continued, possibly due to permanent damage of the near wellbore area by the particles. The analysis of the wells at field B, which switched from matrix to fracture injection, showed that the injectivity increased once fracture injection was started. However, a minor injectivity decline over time was observed even under fracture condition.

Martins et al. (1995) reported PWRI under thermally induced fracturing conditions supplemented by SW at Prudhoe Bay oil field onshore Alaska. The waterflooded zones had moderate reservoir quality. With no exceptions, all the wells injecting PW experienced injectivity loss; however, none of the wells showed a progressive decline. Typically, injectivity was restored over three to six months when switched to SW, which was associated with thermal changes. The same time scale was reported for the injectivity loss when switched from SW to PW. The quality of the re-injected PW varied from <50 mg/l (occasionally >700 mg/l) of oil and from <10 mg/l (occasionally>45 mg/l) of solids, depending on the performance of the water treatment facility. The authors reported that an increase in solids content had a larger effect on the injectivity impairment at high oil concentrations than at low concentrations.

Hjelmas et al. (1996) summarized the experience of PWRI trial at the Ula field offshore Norway. Seawater was co-injected with PW. Typical oil and solids concentrations at 35% SW and 65% PW mixture were 15 mg/l and 6 mg/l respectively. There was no noticeable injectivity loss, which the authors associated with thermally aided fracturing of the formation. Unlike other authors (Mainguy et al., 2019; Martins et al., 1995), Hjelmas et al. (1996) took into account the pore size distribution of the formation when discussing the injection water specifications.

Table 2
Summary of main points from the presented PWRI cases (M – Matrix; F – fracture).

Author	Field Name	Injection Strategy (M/F)	Progressive degradation of injectivity	Formation Permeability (mD)	Oil content (mg/l)	Solid content (mg/l)	Pore size reported to be considered
Tipura et al. (2013)	Grane (Norway)	M	✓	5000–10000	18–65	2–9	✓
Mainguy et al. (2019)	Block 17 Field A (Angola)	M	✓	Several hundreds to several thousand	18	29	×
Mainguy et al. (2019)	Block 17 Field B (Angola)	M	✓	Several hundreds to several thousand	57	34	×
Martins et al. (1995)	Prudhoe Bay (Alaska)	F	×	100–300	50	10	×
Hjelmas et al. (1996)	Ula (Norway)	F	×	173 (Reed and Johnsen, 1996)	15	6	✓
Andersen et al. (2000)	Brage (Norway)	M[a]	×	1000–2000	40	–	×

[a] Cold aquifer water was occasionally injected which could thermally fracture the formation.

Andersen et al. (2000) reported PWRI under matrix conditions into homogeneous, highly permeable Statfjord formation at the Brage field, the North Sea. The PW was supplemented by aquifer water. The oil content of water was 40 mg/l prior to PWRI. Only slight injectivity impairment was observed during the first year of injection. Injection of 100% of aquifer water improved injectivity, which was associated with thermally induced fracturing.

To summarize, Table 2 presents important points from the cases discussed above. In general, injection under matrix conditions leads to significant injectivity impairment. Trials during the 1990s at the Ula field (Hjelmas et al., 1996) and Prudhoe Bay (Martins et al., 1995) were performed at injection wells previously used for seawater injection; hence, the formation was already thermally fractured. Lately, large scale PWRI projects in the North Sea (Tipura et al., 2013) and offshore Angola (Mainguy et al., 2019) executed matrix injection of PW, but unintentional fracturing of the formation was not eliminated. Mainguy et al. (2019) reported that some wells encountered extreme injectivity reduction, so the decision to inject at high pressures was made. Well stimulations to restore injectivity, either by 100% SW or chemicals, showed mixed results. It was not possible to completely recover the loss, and the injectivity swiftly dropped to pre-stimulation levels once injection commenced.

The presented cases and the analysis by (Rossini et al., 2020) indicates that the attempt to relate injectivity impairment only to the total oil and solids content is deficient. Evans (1994) reported that the total oil and solids content were the main parameters describing the injection water quality. He suggested that the relation of reservoir properties to the size distribution of droplets and particles should be the principle water quality criterion. Since then it was substantiated that considering the reservoir properties is vital (Buret et al., 2010; Khambharatana et al., 1998). The literature showed that still in some cases total oil and solids content is the only deciding parameter considered. On the other hand, some oil producers deploy on site coreflooding rigs to improve data acquisition and obtain measurements representative for the flooded reservoir (Costier et al., 2009; Souza et al., 2005). Formation damage by suspended solids and droplets is a complex phenomenon, which depends on a number of parameters that need to be evaluated (Rossini et al., 2020). Section 4 reviews the literature on the flow of oil-in-water emulsions through porous media and factors affecting droplet retention. The goal is to review existing literature concerning droplet retention in porous media to identify parameters affecting the phenomenon and highlight the gaps in the knowledge.

4. Emulsions in porous media

4.1. Permeability reduction models

4.1.1. Homogeneous model

The homogeneous model considers emulsions as a continious single-phase liquid and does not consider interaction between droplets and pore walls. Alvarado and Marsden (1979) proposed a homogeneous model to describe the flow of stable oil-in-water emulsions through porous media. The fundamental assumption in their concept was that the oil-in-water emulsion behaves as a single-phase fluid, and that the continuum flow model can be applied to it. The homogeneous model does not consider capillary resistance due to pore plugging and, thus, would be applicable for low IFT, high oil concentration systems. Alvarado and Marsden (1979) showed that the emulsions with oil concentration up to 40% showed Newtonian behavior, while the emulsions with concentrations higher than 50% showed non-Newtonian behavior. For the Newtonian emulsions, Darcy's law was suggested to describe the flow. The model estimates the reduction of permeability but does not predict it.

4.1.2. Models of droplet retardation

The droplet retardation theory was initially described by McAuliffe (1973). According to his study, when a droplet enters a pore throat it deforms and becomes squeezed (Fig. 2). Now the droplet has a smaller diameter on the advancing side than on the trailing side. Hence, a higher capillary pressure at the front than at the back hinders the droplet from passing through the throat, which is also known as the Jamin effect. In this case, the severity of the permeability reduction depends on the ability of droplets to pass through pore throats. For droplets larger than pore throats, the Young-Laplace equation, describing the pressure difference across a curved interface, will take the form shown in Equation (1) (Alvarado, 1975).

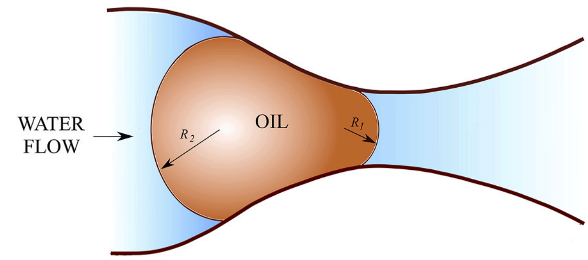

Fig. 2. Oil droplet retention mechanism. The oil droplet is squeezed in the pore throat and retained by capillary forces. Based on McAuliffe (1973).

$$\Delta P = 2\,\gamma\left(\frac{1}{R_1} - \frac{1}{R_2}\right) \tag{1}$$

where ΔP is differential pressure (Pa), γ is interfacial tension (N/m), R_1 and R_2 are the radii of curvature at the front and at the back of the droplet (m).

McAuliffe explained the permeability reduction by the fact that capillary forces caused resistance, which made droplets flow slower than the continuous phase.

The droplet retardation theory was mathematically described by Devereux (1974) for constant pressure flow and by Soo and Radke (1986) for constant velocity flow. The model was capable to describe the permeability change when the emulsion was flowing, but the permeability returned to the initial value when the emulsion flow was stopped and the porous medium flushed with the continuous phase. However, the experimental results showed that the droplets were permanently captured in the porous media, and flushing with the continuous phase did not completely restore the permeability (Buret et al., 2010; Soo and Radke, 1986).

4.1.3. Filtration models

Radke and co-workers performed major work on the flow of emulsions in porous media. Their mechanism of droplet capture for dilute, stable emulsions relies on the principles of deep bed filtration process (Soo et al., 1986; Soo and Radke, 1984a, 1984b, 1985, 1986). Following Herzig et al. (1970), a number of retention sites are differentiated in deep bed filtration (Fig. 3). When the particle size is comparable to the pore size, the particles lodge in constriction sites/pore throats and, as a result, clog them. This mechanism of retention is known as *straining capture*. On the other hand, particles smaller than pore throats can be wedged in crevice sites or in caverns and captured on surface sites due to surface forces, which is referred to as *interception capture*. Straining and interception capture, can influence each other. If the effective pore diameter is reduced due to the interception of particles, it might trigger the straining of other particles, or droplets in case of emulsion flow (Soo and Radke, 1986; Yu et al., 2018a).

The efficiency of the surface capture of particles is then dependent on the surface forces. The surface forces include repulsive electric and/or steric forces, depending on the properties of dispersion, and attractive van der Waals forces. When it comes to wedged particles, the friction forces keep them in place, while in case of straining, the axial pressure of

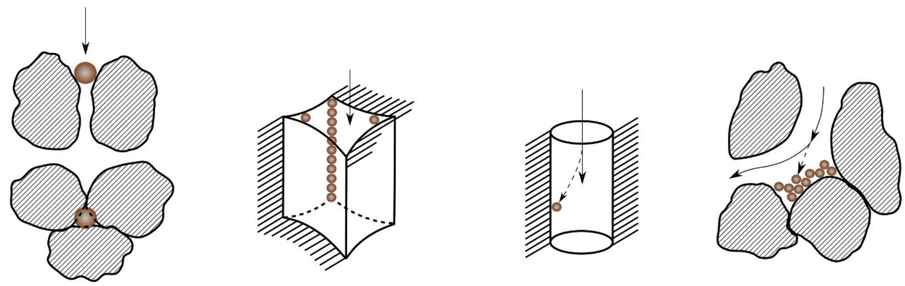

Fig. 3. Retention sites: particles are captured by surface forces at surface sites; particles are trapped in crevice sites due to friction forces; pressure hold the particles in the constriction sites; particles deposit in cavern sites due to flow recirculation. Based on Herzig et al. (1970).

the fluid holds the particles in the constrictions. The transport of particles to a pore wall can occur due to sedimentation or creaming because of the density difference between dispersed and continuous phases. When flow recirculation occurs, as in the case of cavern sites in Fig. 3, the particles deviate from the streamline and become trapped. Furthermore, hydrodynamic effects and Brownian motion influence the transport to the surface.

Although the droplet retention mechanism suggested by Radke and co-workers is similar to the traditional deep bed filtration, it differs in some aspects as it takes into account differences between physical properties of particles and droplets. Firstly, the particle-to-pore size ratio in the traditional filtration is significantly smaller in comparison with the droplet size, which can be similar to pore size. Therefore, the interception mechanism dominates in the case of particles, while for droplets, both straining and interception are present. It is believed that straining is likely to be dominating for droplets. Secondly, unlike particles, droplets are deformable and can re-enter the flow if squeezed through constrictions when the pressure difference exceeds the capillary resistance.

The model of Radke and co-workers characterize the flow of dilute, stable emulsions using three empirical parameters: the filter coefficient describes the sharpness of the emulsion front, the flow redistribution parameter defines the flow redistribution and the time needed to obtain steady-state retention, and the flow restriction parameter estimates the effectiveness of the permeability reduction. Soo et al. (1986) described the procedure for the estimation of the filtration parameters showed that the model is capable to adequately describe the flow of emulsions with oil concentration up to 1% and for oil viscosities in the range from 1.5 mPa s to 23 mPa s. The filtration theory by Radke and co-workers is widely accepted in the literature and has been the basis for most of experimental investigations (Buret et al., 2008; Coulibaly and Borden, 2004; Demikhova et al., 2016; Ding et al., 2020; Khambharatana et al., 1998).

The group of Dong published a series of papers dedicated to the modelling of pore plugging by droplets in porous media (Ding et al., 2020; Ding and Dong, 2019; Yu et al, 2018a, 2019). They introduced a filtration model which incorporates geometry of a porous medium by presenting it as a network of non-uniform capillaries, where the narrow part of the capillary represents a pore throat and the wide part represents a pore body (Fig. 4). The total resistance force arising from pore clogging is estimated as the accumulation of capillary resistance in every plugged capillary. The plugging ability of droplets is defined by drop-to-pore size ratio. The model considers droplet size, droplet number, and droplet-pore wall interactions and incorporates the oil concentration, IFT, oil viscosity, and sandpack permeability and length. They also took into account the injection rates and the volumes of injected emulsion. While the model of Radke and co-workers is accurate for very dilute emulsions (1% oil concentration), the model of Dong and his group gives a good match with experimental data at oil concentration as high as 20%.

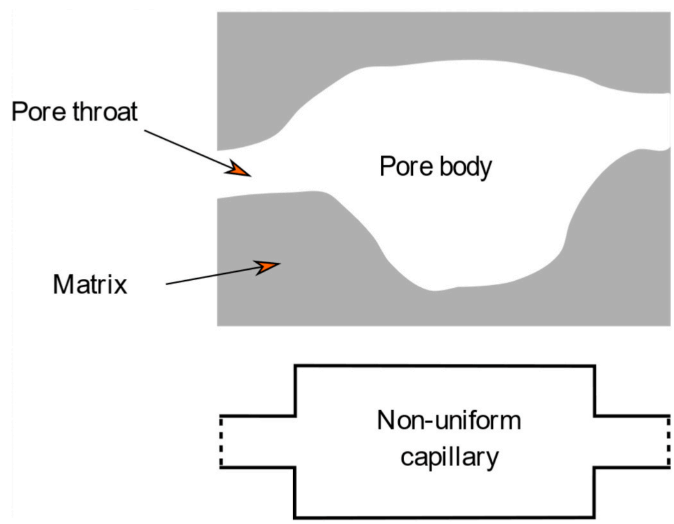

Fig. 4. A pore body represented by a non-uniform capillary. Top: schematic of a pore. Bottom: representation of the pore by non-uniform capillary. Based on Yu et al. (2018a).

4.1.4. Capillary network models

Several network models were proposed in the literature to describe the permeability reduction. Rege and Fogler (1988) proposed a network model for deep bed filtration of droplets and particles that showed good agreement with the experimental work of Soo and Radke (1984a). The model requires only one parameter that takes into account hydrodynamic, gravitational, surface forces and the likes on deposition to characterize the capture probability in a bond. The parameter must be estimated from experimental data; however, the found parameter is still adequate if pore size or particle size distribution changes. Romero et al. (2011) presented a network model for steady state flow of monodispersed oil-in-water emulsion. The model is based on the experimental data from single constricted capillary flow results of Cobos et al. (2009) and utilizes flow rate-pressure drop relationship in each bond of the network to describe the flow of emulsion. The flow rate-pressure drop relation of emulsion flow is presented as a mobility reduction factor (discussed in Section 4.2.2). The model is able to predict the experimental data qualitatively, but lacks quantitative accuracy. Nogueira et al. (2013) extended the work of Romero et al. (2011) by introducing dynamic considerations into the model. The proposed model takes into account dispersed phase volume fraction in the flow through each bond and allows to study transient flow. The model of Nogueira et al. (2013) showed that injection of pure continious phase restores permeability of the network, fully or partially depending on the flow rate.

4.1.5. Langmuir isotherm-based model

Jin and Wojtanowicz (2014) proposed an analytical model that

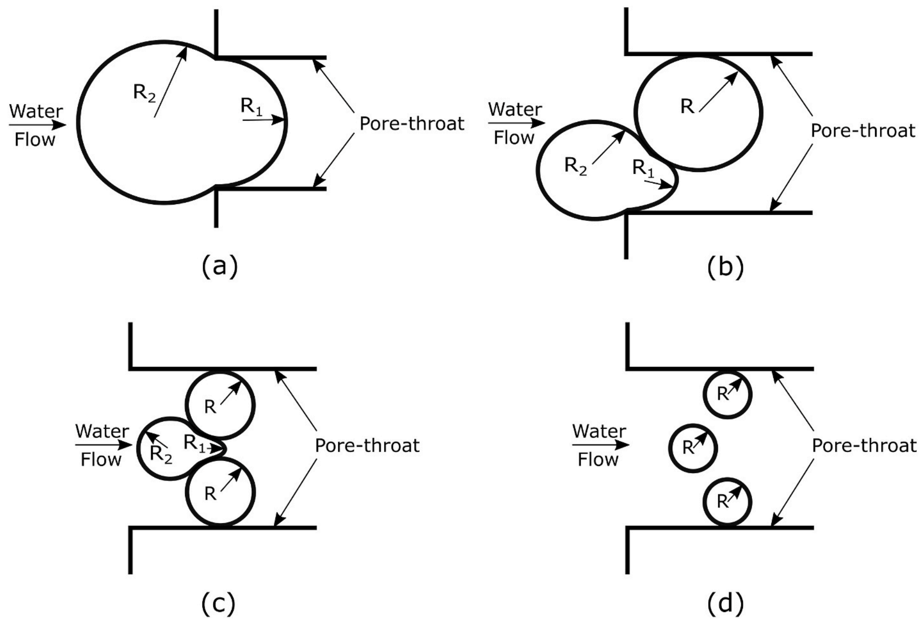

Fig. 5. Pore clogging mechanisms by droplets of various sizes: a) the capture of the droplet of the size larger than the pore throat; b) the capture of the droplets that are slightly larger than the radius of the pore throat; the capture of the droplets that are slightly smaller than the radius of the pore throat; d) the capture of the droplets that are significantly smaller than the radius of the pore throat. Adapted from Yu et al. (2018a).

describes permeability decline in terms of relative permeability change for water when captured droplets increase the oil saturation. The deposition process is described as an adsorption process using a Langmuir isotherm. The model was calibrated using experimental data of Soo and Radke (1984a) and Buret et al. (2010) and showed a good match. The influence of rock wettability and injection rate on injectivity damage is included in the model. This model shows simple analytical solution, but it might require experimental data from bump rate and relative permeability tests for accurate predictions. Jin and Wojtanowicz (2017) extended the model by proposing analytical solution for radial flow. The model predicts oil saturation, relative permeability and pressures around a well bore over time.

4.2. Factors influencing droplet retention

As discussed in Section 3.2, retention of droplets in porous media is a complex process that depends on variety of factors. The literature show that these factors can be divided into two groups: physical and physicochemical properties. These include drop-to-pore size ratio, flow velocity, dispersed phase concentration, interfacial tension (IFT), viscosity, permeability, wettability of medium, stability of emulsions, and interactions between droplets and pore walls. In the following subsections, the factors are discussed in details and the gaps are pointed out.

4.2.1. Reservoir properties

A number of studies investigated the effect of drop-to-pore size ratio on the permeability reduction (McAuliffe, 1973; Soo and Radke, 1984a; Khambharatana et al., 1998; Błaszczyk et al., 2016; Yu et al., 2017; Ding et al., 2020). All studies agree on the fact that larger drop-to-pore size ratio have a more pronounced effect on the permeability reduction because droplets that are larger than pore throat are more likely to plug pore constrictions than smaller droplets. The minimum droplet size in treated PW is limited by the technology used for the treatment (Table 1); therefore, capabilities of treatment facilities and pore size distribution of the formation of interest must be evaluated together when planning re-injection.

The straining capture mechanism is governed by capillary-induced lodging of the droplets in pore throats as shown in Fig. 5a. The droplet passes through the constriction when pressure difference across the pore exceeds the pressure predicted by Equation (1); otherwise, the droplet remains captured. When droplets are slightly larger than half of the pore size as illustrated in Fig. 5b, two droplets are required to clog the pore. The first droplet is captured on the surface due to surface forces, reducing the effective diameter of the pore/pore throat, then the droplet flowing through the restricted pore throat becomes trapped between the already captured droplet and the pore surface. When the droplets are slightly larger than half of the pore size, as illustrated in Fig. 5c, a similar situation occurs, however in this case, the droplet becomes blocked by two droplets already captured on the surface, also referred to as bridging (Moradi et al., 2014). For the cases when droplets are smaller than a third of pore throat size as depicted in Fig. 5d, the retention happens by interception capture.

Another important reservoir property to consider is rock permeability. Permeability of formation can vary from tight rocks to good quality rock of high permeability. In general, experimental results in the literature shows that samples of lower permeability are more prone to permeability decline (Chen et al., 2018; Soo and Radke, 1984a; Yu et al, 2018b, 2019). This suggests that formations with higher permeability are more suitable for re-injection.

Sandstone reservoirs are usually water-wet/intermediate-wet, while carbonates are preferentially oil-wet (Schön, 2015). The wettability of rock decides the flow, morphology of fluids, secondary and tertiary recovery, and might be extremely important for the flow of oil-in-water emulsions. Simulation studies show that wettability influences the droplet passing through constrictions (Wei et al., 2020). To the best of our knowledge, there is no experimental data in the literature on the influence of wettability on the permeability decline in packed beds or core samples. On the other hand, the effect of wettability on oil-in-water emulsion separation in fibrous filters has been widely studied (Agarwal et al., 2013; Bansal et al., 2011; Magiera and Blass, 1997). Extrapolation of these results on reservoir rock can be reasonable as the wettability is independent of media morphology. Bansal et al. (2011) showed that droplet capture was more efficient when the porous media was more oil-wet. Adherence of oil droplets on surfaces with various wettability in a laminar flow substantiate this suggestion (Han et al., 2020). As the drop-to-pore size ratio approaches one, the droplet capture becomes more independent of wettability and is governed by flow velocity since

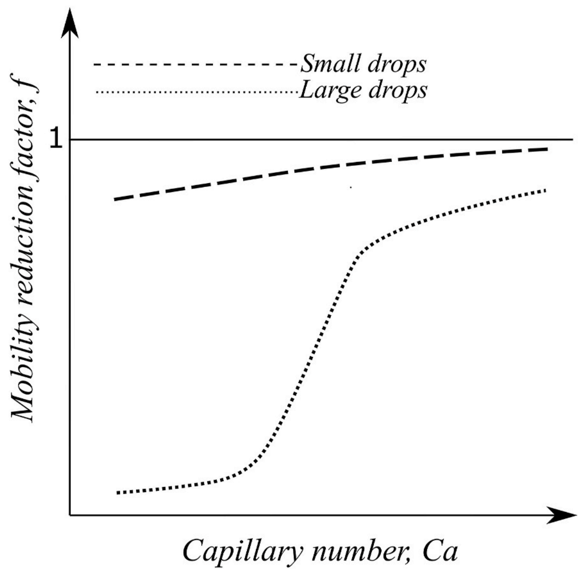

Fig. 6. Schematic visualization of the mobility reduction factor as a function of capillary number.

at low velocity droplets would be retained regardless of surface energy and at high velocities hydrodynamic forces would push droplets through constrictions (Bansal et al., 2011). Relative permeability studies suggest that water relative permeability is higher for oil-wet rocks than in water-wet rocks (Anderson, 1987). Therefore, although oil-wet rock might have higher droplet capture efficiency, injectivity decline can be slower in oil-wet formations. This hypothesis is substantiated by the injectivity decline model of Jin and Wojtanowicz (2014).

4.2.2. Capillary effects

The ability of large droplets to overcome the capillary forces and pass through a constriction can be described by the capillary number (Equation (2)). For flow in porous media, the capillary number represents the balance between viscous forces and capillary forces, and it is routinely used to investigate mobilization of residual oil.

$$Ca = \frac{\mu_c v}{\gamma} \quad (2)$$

where γ is the interfacial tension (N/m), μ_c is the dynamic viscosity of the continuous phase (Pa*s), and v is the Darcy velocity (m/s).

Cobos et al. (2009) characterized the flow of droplets through a constricted capillary using flow rate-pressure drop relationship. It can be presented as a mobility reduction factor, *f*, which is the ratio of the pressure drop of the continuous phase flow to that of the emulsion flow at the same flow rate. The mobility reduction factor can be explained as a scale factor that describes the liquid mobility when it flows through throats. The authors found that when droplets are significantly smaller than the pore throat, *f* is independent of the capillary number and is about 1. For droplets that are larger or close to the size of a throat, there is a critical capillary number, Ca_c, at which mobility reduction factor falls abruptly and become significantly smaller than one (Fig. 6). When capillary number is larger than the critical value, *f* approaches 1. Guillen et al. (2012) experimentally found that for a sandstone sample $Ca_c \approx 10^{-4}$. Considering a typical injection conditions, the capillary number near well-bore area is about than 10^{-5}, while in the reservoir the number is about 10^{-7} (Jin and Wojtanowicz, 2014; Mendez, 1999). The mobility reduction factor has been successfully implemented in the modeling of permeability decline to describe flow rate-pressure drop relation of emulsion flow (Nogueira et al., 2013; Romero et al., 2011). In general, when it comes to the flow rate, there is agreement in the literature – high flow rates cause higher droplet mobility (Hofman and Stein, 1991; Moradi et al., 2014; Yu et al., 2017). Moreover, the experimental data shows that larger droplets require higher flow rate to be squeezed through a constriction (He et al., 2019).

Ding et al. (2020) varied flow rate with respect to various IFT, emulsion quality (dispersed phase concentration), droplet size, sand pack length, and sand pack permeability. For all sets of experiments, they found that there are two flow regimes distinguished by a critical yield pressure – the total capillary resistance force that needs to be overcome to start the flow of emulsion. Below the critical yield point, only a few droplets were able to pass through pore throats, and most of them remained trapped since the displacing pressure was not high enough to overcome the capillary forces. Above the critical yield point, the droplets were deformed and started to flow through the pore throats. The findings of Ding et al. (2020) are well in line with the discussed mobility reduction factor.

Another parameter to consider when discussing capillary effects is interfacial tension. It is known that the interfacial tension influences the depth of emulsion penetration into a porous medium (Ding and Dong, 2019; Yu et al., 2018c) and the effectivity of droplets to plug the pores (Yu et al., 2018c). Yu et al. (2017) showed that higher IFT leads to increased permeability reduction. Moreover, by the analysis of effluent droplet size distributions, they found that as the IFT increased the median diameter of droplets in the effluent decreased. They explained this by the fact that droplets with low interfacial tension are more deformable and more mobile than droplets with high IFT. Consequently, for a fixed constriction diameter at the same pressure drop, droplets with low interfacial tension and a slightly larger diameter than the constriction were able to pass through the pore throat, while at high IFT the droplets were less likely to squeeze through the constriction.

4.2.3. Oil viscosity

In addition to capillary forces, the pressure difference across the pore is a function of a frictional force (Chen et al., 2018). The frictional force arises from the viscosity of the dispersed and continuous phase. The frictional resistance in a pore throat depends on the viscosity of oil, flow velocity, droplet size and the throat geometry. When a droplet passes through a constriction, the frictional resistance occurs at the contact between the droplet and the wall. As the oil viscosity increases it becomes more difficult for a droplet to pass through the pore throat. The frictional resistance in a pore body, however, depends on emulsion viscosity.

Chen et al. (2018) showed that the oil viscosity affects the effective viscosity of emulsion during flow through a permeable medium. The effective viscosity of the emulsion increases with the oil viscosity. They observed that at high oil viscosities, the increase levels off, which was explained by wall slipping. In the case of the water-wet matrix, the oil droplet and the matrix is separated by a thin water layer (Churaev, 1993), which leads to wall slipping. Wall slipping happens because of weaker molecular attraction between oil and solid molecules than between oil molecules. Fluids with low viscosity do not exhibit slipping; however, the effect is more pronounced as the viscosity increases (Bonaccurso et al., 2003; Craig et al., 2001).

On the other hand, Chen et al. (2018) argue that the flow path of the emulsion is another possible explanation for the non-proportional increase of frictional resistance (Fig. 7). It is difficult for oil droplets with higher viscosity to pass through narrow pores; thus, the emulsion flow is redirected into larger pores where they have less contact with the matrix. For droplets with low viscosity, it is easier to flow through narrow constrictions, and they maintain a contact with pore walls while moving, which results in higher frictional resistance. Chen et al. (2018) reported that effect of viscosity significantly affects permeability decline and becomes more pronounced in samples of lower permeability. For a sample of 3 Darcy permeability, when the oil viscosity increased from 9.4 mPa*s to 496 mPa*s, the pressure drop across the sample increased 3.8 times. Heavy crude oils (1200 mPa*s) can lead to up to 99% permeability reduction (Yu et al., 2018b). The oil concentration in the

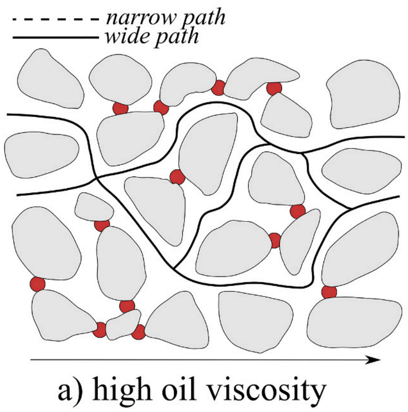

Fig. 7. Schematic representation of flow paths for different oil viscosity emulsions at steady state (arrows indicate the flow direction): a) flow of droplets of high viscosity – narrow paths are blocked and droplets flow through wide paths; b) flow of droplets of low viscosity - droplets of low oil viscosity flow through narrow paths causing additional friction when squeezing through pore throats.

study of Chen et al. (2018) is 10 wt%, while the lowest concentration in the study of Yu et al. (2018b) is 2.5 wt%. This is significantly higher than the oil content in produced water after treatment (Table 1). Therefore, the effect of oil viscosity on the permeability decline for PW might be less significant than in the mentioned studies or become more substantial as permeability decreases. The flow of 0.5 vol% emulsions with oil viscosities of 1.5 mPa*s and 23 mPa*s did not show a significant difference between two systems (Soo and Radke, 1984b). However, it is difficult to draw conclusions as the tested viscosity range is small. Therefore, an investigation at low oil concentration and broad viscosity range is required to supplement the above-mentioned studies.

4.2.4. Oil content

In injection studies, oil concentration in an emulsion is commonly referred to as emulsion quality, while the volume of injected emulsion is regarded as slug size. The effect of slug size can be studied either by injecting a fixed volume of emulsion into sandpacks/core samples of various length (Błaszczyk et al., 2016; Ding et al., 2020) or by injecting different volumes (Yu et al., 2018c) into sandpacks of the same length. When it comes to oil concentration, it is found that higher oil concentration understandably leads to more pronounced permeability decline as there are more droplets available to be trapped (Ding et al., 2020; Ding and Dong, 2019). The studies of slug size by varying packed bed length show inconsistent results. Some studies demonstrated that permeability reduction increases with the increase of the slug size (Ding et al., 2020; Ding and Dong, 2019). While Błaszczyk et al. (2016) showed that although the overall permeability decreased, it did not depend on the slug size and the decrease was almost the same for all runs. The discrepancy could be associated with type of plugging: rapid external cake formation or more uniform deposition along the sample.

4.2.5. Surface forces

The surface forces play an important role in the interception of droplets during flow in porous media. The efficiency of surface capture of droplets is significantly influenced by the flow velocity (Soo and Radke, 1984b; Rousseau et al., 2007) and the ionic strength (Buret et al, 2008, 2010). Following Soo and Radke (1984b), the interaction energy curve between a droplet and a sand grain as a function of the separation distance is a convenient approach to describe the potential role of the velocity on surface capture (Fig. 8). A weak capture of droplets happened at low velocities in the secondary minimum. When the velocity slightly increased but was still not high enough to make hydrodynamic forces overcome the repulsive energy barrier, the capture rate decreased because droplets were dragged out of secondary minimum. Once the velocity allowed the hydrodynamic forces to overpower the energy barrier, strong capture occurred in the primary minimum.

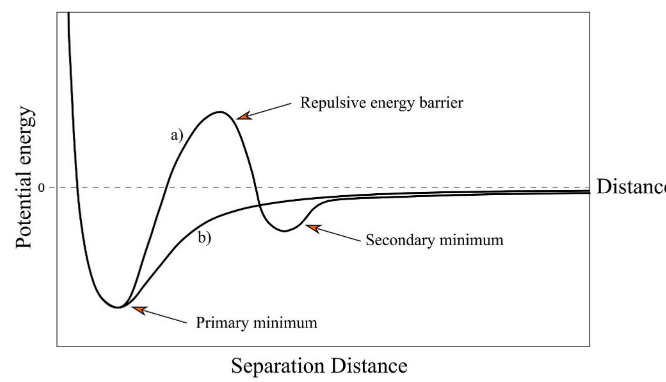

Fig. 8. Interaction energy curves between a droplet and a sand grain: a) an interaction curve for low salinity system; b) an interaction curve for high salinity system such as produced water.

Naturally, in a system with high ionic strength, like produced water, there is no secondary minimum or repulsive energy barrier according to the classical DLVO theory; however, the repulsive barrier can be present due to steric interactions induced by the indigenous surface-active components present in crude oil.

Rousseau et al. (2007) described surface capture in terms of dimensionless Peclet number, *Pe*, which is the ratio of convective to diffusive effects Equation (3).

$$Pe = \frac{Ua_g}{D} \tag{3}$$

where U is the interstitial velocity of colloidal particles (m/s), e.g., interstitial velocity is the Darcy velocity divided by the effective porosity, a_g is the radius of the collector grain (m), and D is the particle Brownian diffusion coefficient (m^2/s).

They argued that, depending on *Pe,* three surface capture regimes can exist: convection-diffusion, hydrodynamic, and interception. Noteably, Soo and Radke (1986) define retention of droplets smaller than pores as interception capture in all the cases. In the convection-diffusion regime, as *Pe* increased, the capture efficiency decreased until a critical Peclet number (Pe_c^g) was reached. Once *Pe* became larger than Pe_c^g, the hydrodynamic regime came into action and the capture efficiency increased with *Pe* since the hydrodynamic forces overpower the energy barrier and "push" the droplets into the primary minimum as discussed previously. At high Peclet number (high velocity), the hydrodynamic forces are high enough that the deposition probability was equal to one.

In the follow-up work, Buret et al. (2008) investigated the effect of

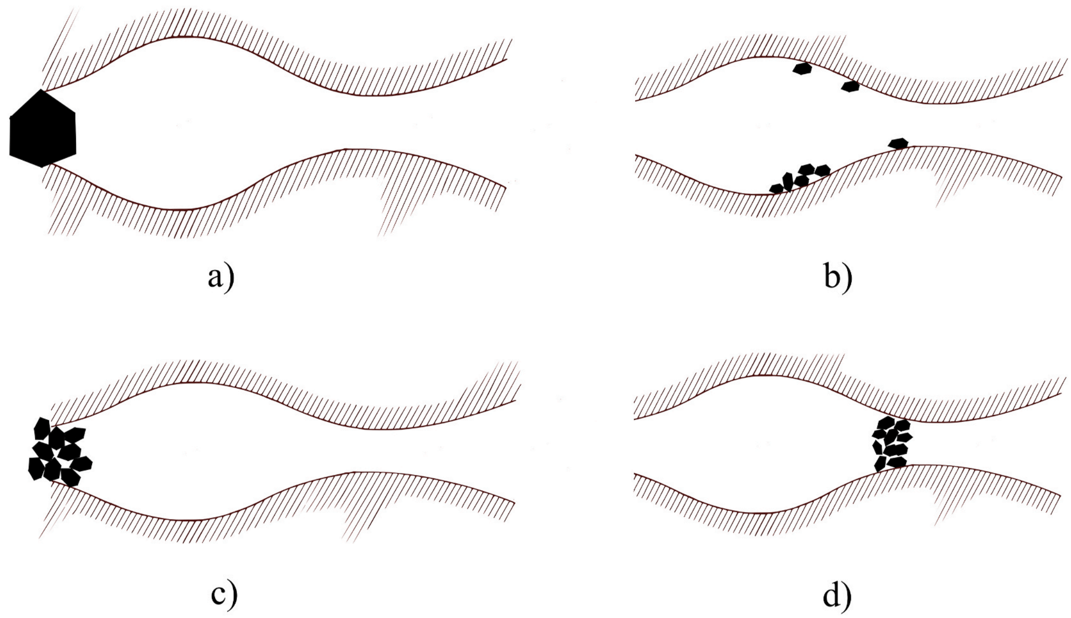

Fig. 9. Pore plugging mechanisms by solid particles: a) external cake formation by a particle larger than the pore constriction; b) particles smaller than 1/3 of the pore throat deposited because of adsorption or sedimentation c) external cake formation by particles larger than 1/3 of the constriction form a bridge; d) particles smaller than 1/3 of the pore throat bridging deeper in the reservoir.

salinity on the surface capture in the convection-diffusion regime. They demonstrated that the deposition of droplets was uniform along the length of the sand pack, and an increase in salinity led to higher permeability reduction. Moreover, it was found that at high salinities, the deposition occurred by two different mechanisms. In the beginning of injection, there was a rapid deposition of droplets and delay in the droplet breakthrough. When the effluent concentration curve reached a plateau, it was still lower than the initially injected concentration; therefore, there was a slow, constant deposition of droplets happening. They argued that the first mechanism is consistent with a low energy barrier, while the second mechanism came from the presence of already deposited droplets, which caused steric hindrance. However, they speculated that the ability of the droplets to deform allowed them to pack better at the surface, which explained the constant deposition. Buret et al. (2010) showed that at very low salinities (less than 0.6 g/l NaCl) there was no permeability reduction for their system (negatively charged droplets and surface at low salinity), while there was significant permeability reduction at higher salinities. They found their retention results to be consistent with the estimated energy barriers at all the tested salinities. In addition, Buret et al. (2010) studied the effect of drop-to-pore size ratio on permeability reduction. For the drop-to-pore size ratio of 0.1 (largest tested), a permeability reduction of around 57% was obtained. It was shown that the permeability reduction was uniform along the core and increased with an increasing drop-to-pore size ratio.

4.2.6. Droplet stability

Most of the conclusions regarding droplet capture are based on the experiments conducted using model oil emulsions that are stabilized by surfactants (Yu et al., 2017), and only a few papers reported results with crude oil (McAuliffe, 1973). It is common practice in the literature to stabilize emulsions by high concentrations of surfactants (Cobos et al., 2009; Romero, 2009; Ding et al., 2020). This prevents the coalescence of droplets flowing in porous media and gives better control over experiments. However, the stability aspect of crude oil emulsion could play an important role in the prediction of injectivity decline. Indigenous surface-active components that are present in crude oil form interfacial layers promoting the stability of droplets against coalescence (Goodarzi and Zendehboudi, 2019). At high salinities, typical for PW, electrostatic interactions are suppressed, and steric interactions play the main role in droplet-droplet and droplet-pore wall interactions. Surface active components can provide steric hindrance when droplets approach each other and impede coalescence. Additionally, steric effects can influence the attachment of droplets to pore walls. The amount of these surface-active components is highly oil-dependent and consequently stability of droplets changes from one oil field to another (Dudek et al., 2017). The coalescence of droplets suggests that drop-to-pore size ratio would dynamically change during the re-injection. Increase in the drop-to-pore size ratio would decrease mobility of droplets (He et al., 2019) and causes more pronounced injectivity decline. To the best of our knowledge, the effect of coalescence on permeability decline has not been systematically studied in the literature and only a few studies investigated the phenomena, exclusively with model oils.

Fundamental work on the transport of unstable emulsions through porous media was performed by Spielman and Su (1977) and Spielman and Goren (1970). Initially, it was suggested that there are two regimes of coalescing droplets within the pore space: 1) droplets suspended in water; 2) coalesced droplets which travel through the medium in the form of ganglia (Spielman and Goren, 1970). Later, the theory was reformed by the addition of an intermediary regime when discrete globules are immobile until they grow enough, due to coalescence with other droplets, to merge with each other and form ganglia.

Soma and Papadopoulos (1995) studied the effect of pH and salinity on the flow of unstable oil-in-water emulsions through packed beds. They showed that pH had a significant influence on the permeability reduction as the surface charge of oil droplets changed from positive to negative in the range of tested pH values (the matrix was always negatively charged). The general trend showed that the permeability reduction increased with decreasing pH as the oil droplets passed the isoelectric point (IEP) and became positively charged, which caused strong attraction of the droplets to the matrix. Moreover, they demonstrated that at IEP, the flowing droplets experienced the easiest coalescence with already deposited droplets and subsequent re-entrainment. When it comes to salinity, Soma and Papadopoulos (1995) demonstrated that in the absence of any surfactant the retention of droplets increased as salinity increased (Hofman and Stein, 1991). showed that less stable emulsions are more effective in clogging porous media.

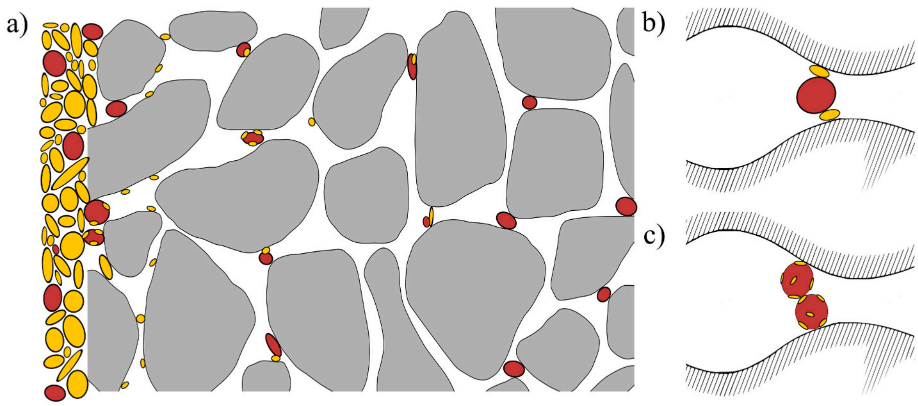

Fig. 10. Conceptualized formation damage mechanism during co-injection of droplets (red) and particles(yellow): a) particles deposit mainly at the face of the formation, while droplets deposit deeper in the reservoir carrying some solids with them; b) a droplet smaller than the pore constriction is captured because of the deposited particles; c) two droplets stabilized by particles clog a constriction bridging. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

4.2.7. Synergy of solid particles and droplets

As discussed in Section 2.2, produced water contains both dispersed oil and solids. Formation damage by solid particles has been extensively studied in the literature; however, the topic is outside of the scope of this paper and will be discussed briefly here. The reader is referred elsewhere for an extensive overview of common formation damage theories and models (Civan, 2007). On the other hand, there are not many studies dedicated to the permeability decline during the injection of droplets and solids together (Al-Riyamy and Sharma, 2004; Ochi and Oughanem, 2018; van den Broek et al., 1999; Zhang et al., 1993). First, it is useful to briefly describe injectivity impairment by particles only. As shown in Fig. 1, solid particles plug the formation either by creating an internal or external filter cake. During the initial time of injection, suspended particles form internal filter cake reducing the permeability. After some time, only a few particles can enter the internal filtration zone, and an external filter cake starts to build on the surface of the well (Pang and Sharma, 1997). External filter cake build up is a rapid process that significantly reduces the injectivity (Ershaghi et al., 1986). The extent of the plugging is governed by particle-to-pore size ratio, surface charge and injection rates (Bennion et al., 2001). Particles that are larger than 1/3 of a pore throat form external filter cake either via bridging or plugging pore throats by itself (Abrams, 1977), while particles smaller than 1/3 and larger than 1/14 of pore size travel deeper into formation and deposit in pores either by bridging, sedimentation or adsorption (van Oort et al., 1993). Some studies show that droplets that are even smaller than 1/14 can build internal filter cake at low injection rate (Bennion et al., 2001). The mechanisms of pore plugging by particles are illustrated in Fig. 9.

It was shown that emulsions containing solids did not behave according to the filtration model (Al-Riyamy and Sharma, 2004) and the permeability damage caused by the droplets and solids together was more severe than by each component alone (Ali et al., 2011; Ochi and Oughanem, 2018; van den Broek et al., 1999; Zhang et al., 1993). Ochi and Oughanem (2018) injected only particles, only droplets, and particles and droplets together into unconsolidated sand pack. The results showed that particles preferentially deposited at the entrance of the sand pack, while droplets deposited along the whole length (Fig. 10a). They speculated that more severe permeability reduction during co-injection of particles and droplets might be explained by the fact that in the presence of oil droplets the particles were carried deeper into the sand pack. Moreover, Ochi and Oughanem (2018) reported that the higher the solid concentration, the more oil was trapped at the entrance of the sand pack. Zhang et al. (1993) suggested that while for oil droplets, it was more difficult to plug the pore throats of comparable size, solid particles are capable to plug/bridge those pore throats (Fig. 10b). This provided favorable configuration for oil droplets to plug these pore throats, which contributed to severe permeability decline. Furthermore, solids that are present in produced water, play an important role in the droplet stabilization and pore clogging. The solids can also be coated with resins and asphaltenes, which could likely influence the attachment to the pore walls or promote adherence to each other to create "schmoo"-like substance (Gaweł et al., 2016). Additionally, solid particles adsorbed on the interface contribute to higher stability of droplets impeding the droplet coalescence (Pickering emulsions) and possibly to pore clogging (Fig. 10c). The origin of the suspended solids might also be a factor to consider as their wetting properties play an important role in interaction with crude oil droplets (Gaweł et al., 2016). To date, co-injection of solids and droplets together has not been sufficiently studied to obtain good understanding of the plugging mechanisms.

5. Outlook

Produced water is a complex fluid consisting of many components that can cause formation damage through various mechanisms. Dispersed oil and suspended particles that are present in PW are believed to be the main cause of significant loss of injectivity. In general, the oil and solids content are the main parameter deciding the quality of produced water for injection, although reservoir properties are considered in some reports (Section 3.2). However, the literature review shows that planning the injection solely based on the content is inadequate as the droplet- or particle-to-pore size ratio plays a significant role in permeability reduction. Moreover, unlike solid particles, oil droplets can re-enter the flow by being squeezed through pores and penetrate deeper into the formation potentially damaging a larger area. The literature suggests that there is a number of parameters that needs to be considered during the transport of droplets through porous media in addition to concentrations. Table 3 summarizes these parameters and highlights, accordingly, the gaps in the literature that needs to be addressed to improve the understanding of droplet retention.

The topic of droplet retention is of importance both for the field of emulsions enhanced oil recovery studies and produced water re-injection. However, most of the studies aimed at the fundamental understanding of the droplet retention phenomenon are performed by the former, while the latter mainly reports data from case studies where the focus is typically on the overall change of the injectivity. The effect of flow rate, drop-to-pore size ratio, permeability, interfacial tension, oil concentration and oil viscosity on droplet retention are of importance for emulsion injection as EOR. Table 3 shows that these parameters apart from viscosity were studied extensively in the literature and are well understood. On the other hand, the parameters that could potentially affect the efficiency of PWRI (droplet stability, wettability, and the coupled effect of particles and droplets) lack understanding and pose multiple gaps. This observation substantiates the lack of systematic fundamental studies of droplet retention in the field of produced water re-injection. The case-to-case approach that is typically pursued in re-injection studies allows to obtain some insights characteristic of a

Table 3
Summary of the main results on droplet capture from the literature and suggestion for further studies.

Parameter	Well understood?	The current state of research/Suggestion for the future research
Drop-to-pore size ratio	✓	- Drop-to-pore size is the governing parameter for droplet retention. - Large droplets are more likely to be trapped by straining capture and cause more prominent and rapid permeability decline.
Permeability	✓	- Formations of higher permeability are more suitable for re-injection.
Wettability	×	- Lack of studies in oil-wet media. - The comparison of droplet retention in water-wet and oil-wet conditions is difficult to make. - Experimental work focused on the effect of wettability conditions on droplet retention is needed.
Flow velocity	✓	- Flow velocity is significant for the straining of large and interception of small droplets. - At high velocities large droplets are less likely to be captured, while small droplets are more likely to overcome repulsive barrier and adsorb on a surface.
IFT	✓	- Droplets with large IFT are more likely to be captured by straining mechanism.
Oil viscosity	×	- Oil viscosity plays a significant role at high oil concentrations. More experimental work needs to be done to substantiate this observation at low concentrations typical for PW.
Oil concentration	✓	- Oil concentration is one of the main parameters for permeability reduction. - Larger oil concentration cause more prominent permeability reduction because there are more droplets to be trapped.
Surface forces	✓	- Surface forces play important role in droplet capture. - Flow velocity and pH control the interception of droplets. - Effect of steric forces on interception needs to be investigated.
Droplet stability	×	- Coalescence of droplets in porous media was reported to cause more severe permeability reduction. - Most of the studies in the literature consider only surfactant stabilized model oil droplets. - Gap in the knowledge on the influence of crude oil composition on droplet retention.
Coupled effect of solids and droplets	×	- Solid particles influence retention of droplets and it becomes more severe. The mechanisms are not completely understood. - Microfluidic studies are suggested to examine this issue.

discrete systems but misses out on building the knowledge methodically. As the result, key elements of the retention phenomenon remain practically unexplored.

Although the emulsion EOR studies utilize surfactant-stabilized emulsions, the knowledge regarding the examined parameters is relevant to PWRI. These parameters are mainly physical properties that are independent of surface forces. When it comes to IFT, it is usually analyzed as part of dimensionless capillary number, which makes it possible to establish a correlation (Cunha et al., 2018). However, it is important to highlight that the emulsions investigated in EOR studies have an IFT value one-two orders of magnitude lower than typical values for crude oil-brine systems. These might cause some difference when it comes to breakage of droplets in porous media; however, all in all, the trends established in the literature give a very good insight into the influence of interfacial tension. Presumably, the principal gap in the literature is the lack of systematic research on the effect of droplet stability. The dynamic change in the drop-to-pore size ratio through coalescence can significantly change the behavior of droplets and potentially result in a larger permeability reduction (Section 4.2.6). Considering high salinity of produced water, repulsive energy barrier, which could hinder coalescence of droplets, could be governed by steric forces or Gibbs-Marangoni effect caused by indigenous surface-active components present in crude oil (Dudek et al., 2020). Buret and co-workers performed a substantial work on the influence of surface forces on stabilized droplets capture and suggested that steric forces need to be investigated in the future studies (section 4.2.5). A systematic study examining model oil emulsions stabilized by a wide range of surfactant concentrations from low to high could provide a good foundation for a more complex study using crude oils with varying compositions. Keeping droplet stability in mind, the coupled effect of solids and droplets on permeability reduction is another large gap (Section 4.2.7). On the one hand particles can stabilize droplets controlling drop-to-pore size ratio. On the other hand particles alone can plug the pores preventing droplets passing through them. Experiments considering particles added only to the oil phase and only to the water phase, and then combined could shed light on plugging mechanisms. Hypothetically, the ability of particles to rapidly build a filter cake capturing droplets at the face of formation or being carried deeper into the formation with oil and forming schmoo-like substance could depend on the phase that the particles are suspended in. Moreover, as mentioned in Section 4.2.7, the origin of particles governs their wetting properties and interaction with crude oil suggesting that experiments considering particles of various origin are needed. However, this kind of studies should initially be done with model oils to isolate physicochemical interactions that could arise from surface active components of crude oil to understand the mechanics of droplet and particle transport. Afterwards, experiments using various crude oils could be performed to examine the contribution of crude oil components on the interactions between dispersed components within porous medium. Wettability of rock is known to change the behavior of fluids flowing through permeable materials (Anderson, 1987); however, its effect remains unexplored and it its impractical to draw comparison between droplet transport in oil-wet and water-wet system. The difficulty to investigate the retention mechanisms for different wettability systems and, in general, for abovementioned gaps arises from the experimental methods that are typically utilized in the literature.

Almost all studies investigating the flow of oil-in-water emulsion through porous media use core plugs or packed beds, either sand or glass beads, for the experiments (Ali et al., 2011; Ding et al., 2020; Ochi and Oughanem, 2018; Soma and Papadopoulos, 1995; Yu et al., 2017). Since there is no possibility to observe the flow of the emulsion inside the pores in a core plug or packed bed flooding without expensive and complicated methods such as micro-computed tomography (Mikolajczyk et al., 2018), most of the conclusions regarding the plugging mechanisms and efficiency are based on pressure readings (Vaz et al., 2017) and comparison of injected emulsion with effluent droplet size distribution and concentration. On the other hand, visualization of the droplet transport might provide a new insight into the pore plugging mechanisms. Cobos et al. (2009) performed visualization of the emulsion flow by the use of a constricted glass capillary that mimicked a pore body and a throat. This allowed them to decsribe pressure behaviour when a droplet is passing through the constriction. While constricted capillaries allow to study single trapping events in 1D flow, microfluidic chips (referred to as micromodels later in the text) open an opportunity to observe multiple events in 2D and complement conventional measurements with image analysis.

There were only a few studies on the flow of oil-in-water emulsions using micromodels available in the literature. Soo and Radke (1984a) built a micromodel from two glass plates, and sand squeezed between them. The micromodel allowed to observe emulsion flow under a microscope visually; however, only one set of experiments aimed to justify the filtration viewpoint of permeability reduction was published. Xu et al. (2017) fabricated glass micromodels with varying depth mimicking pore throats. They injected model oil emulsions of droplet

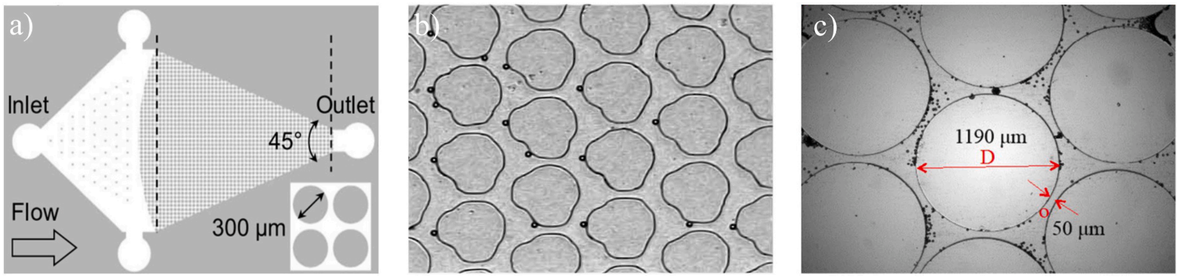

Fig. 11. Particle retention studies using micromodels: a) design of a microfluidic chip for the study of particle retention around production well (Liu et al., 2019); b) pore-scale visualization of latex particle attachment by interception mechanism (Auset and Keller, 2006); c) pore clogging by latex particles in a 50 μm pore throats (Jung et al., 2018). Reprinted by permission from Springer Nature Customer Service Centre GmbH, Springer Nature, Microsystems Technologies, A microfluidic pore model to study the migration of fine particles in single-phase and multi-phase flows in porous media, Jongwon Jung et al, © 2018.

size significantly higher than pore size to study capillary snap off phenomenon; however, no emphasis on droplet retention was done. Moses and Ng (1985) used glass micromodels to study emulsion breakdown by a granular porous coalescer.

Nevertheless, micromodels have been widely used to investigate the retention of particles in porous media (Auset and Keller, 2006; Jung et al., 2018; Van de Laar et al., 2016; Liu et al., 2019; Wang et al., 2011; Wyss et al., 2006), and the demonstrated capabilities substantiate the use of micromodels for the study of droplet retention (Fig. 11). Great advantage of micromodels over packed beds/core plugs is the ability to obtain experimental data on the pore-clogging at a single- and multiple-pore scale. The ability to visualize the flow at the pore scale allows to apply image analysis to track clogging events (Auset and Keller, 2006; Liu et al., 2019; Wyss et al., 2006), consequent flow redistribution (Liu et al., 2019), and utilize additional tools to examine the influence of pore geometry on the retention, e.g., particle imaging velocimetry (PIV) (Van de Laar et al., 2016). Additionally, micromodels provide very good control over the experimental conditions and allow to test various pore geometries. A study to highlight is by Liu et al. (2019), where authors simulated the convergent radial flow to investigate particle retention phenomenon near the production well Fig. 11a. The use of micromodels enables the possibilty to compare the retention of droplets in identical pore networks at different wettability conditions (Grate et al., 2013). Moreover, micromodels can be modified to create surface interactions representatives of real reservoirs, e.g., clay particles can be deposited in micromodels to simulate sandstone reservoirs (Song and Kovscek, 2015). Additionally, the use of micromodels might help to achieve a breakthourgh in the study of co-injection of particles and droplets. All in all, the "behavioral" interpretation of droplet retention complemented by micromodel studies would give a new mechanistic understanding at the capillary level.

6. Conclusions

The mechanisms of droplet capture, physical parameters and surface forces affecting the retention rate in porous media were reviewed. Several models are available to describe the flow of oil-in-water emulsions and predict permeability reduction. The efficiency of droplet capture and pore clogging is defined by drop-to-pore size ratio in most of the models and all the models require some parameters for calibration that can be obtained experimentally. Although a number of meaningful correlations regarding the various parameters were established, several gaps in the knowledge were identified, which can make it difficult to project existing information to produced water re-injection. This is mainly associated with the lack of understanding of the coalescence of droplets during the transport through porous media. Additionally, the uncertainty regarding the coupled effect of droplets and particles on pore clogging, as well as wettability effects, exacerbates the gap. Novel approaches that allow visualization of the retention phenomenon at pore scale are expected to be beneficial for further advancements. Microfluidics was suggested as a promising technique in this respect and could complement conventional core flooding experiments and provide a new insight into the retention studies.

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.

Acknowledgements

This work was carried out as a part of SUBPRO, a Research-based Innovation Centre within Subsea Production and Processing. The authors gratefully acknowledge the financial support from SUBPRO, which is financed by the Research Council of Norway, major industry partners and NTNU. Additionally, the authors thank Jamie Stuart Andrews from Equinor and Sébastien Simon from NTNU for valuable comments during the preparation of the manuscript.

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