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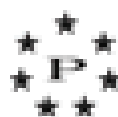


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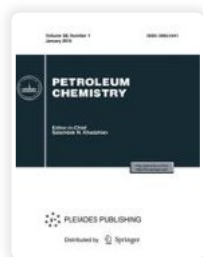
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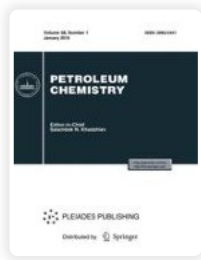
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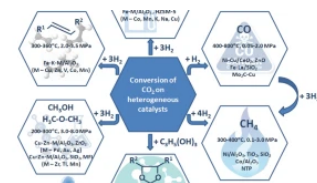
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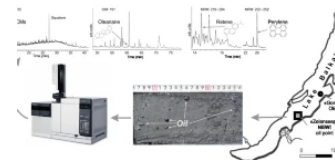


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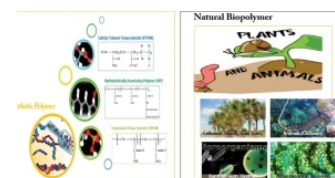
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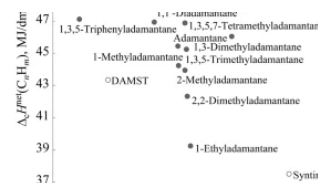
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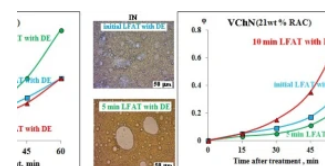


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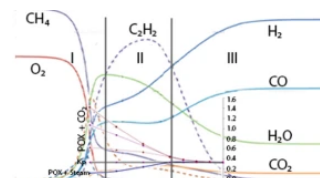


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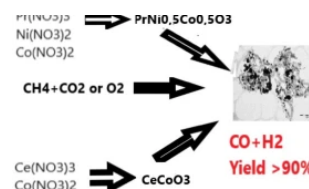


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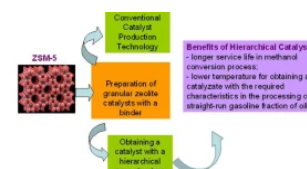
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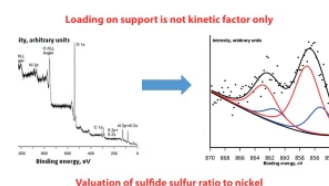
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# A Short Review of Biopolymers for Enhanced of Oil Recovery in Mature Fields

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**Abstract**—The enhanced oil recovery (EOR) techniques are employed to recover more oil from mature reservoirs following the primary and secondary stages of oil production. Polymer flooding is a chemical EOR method that involves the addition of polymer molecules to a treated area to improve its water viscosity and sweep efficiency, and to accelerate oil production. The increased water viscosity will reduce the mobility ratio of the injected fluid to the reservoir fluid. Polymer flooding is most effective when applied in the early stages of a water flood while the mobile oil saturation is still high. The polymers used in polymer flooding are divided into synthetic polymers and biopolymers. Unfortunately, synthetic polymers are susceptible to brine formation and high temperatures. On the other hand, biopolymers have a more rigid chemical chain structure making them more resistant to high temperatures (about 135°C) and salinity (about 220 g/L). This review presents a comprehensive discussion of the use of polymers for EOR applications. First, a summary and explanation are given of the structure and characterization of polymers. Subsequently, the general screening criteria for polymer flooding are highlighted. Then, the properties of synthetic and natural polymers and their application in EOR are presented. Finally, the limitations of this study and suggestion for future work on polymers in EOR are explained.

**Keywords:** biopolymers, polymer flooding, EOR method

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## Table of abbreviations

EOR	Enhanced oil recovery	RIPED	Research Institute of Petroleum Exploration and Development
HPAM	Hydrolyzed polyacrylamide	HAP	Hydrophobically associative polymer
HAPAM	Hydrophobically associating polyacrylamide	C*	Critical concentration
HTHS	High temperature and high salinity	CMC	Carboxymethyl cellulose
Na-CMC	Sodium carboxymethyl cellulose	MCC	Microcrystalline cellulose
HEC	Hydroxyl ethyl cellulose	MC	Methylcellulose
KYPAM	Salinity-tolerant polyacrylamide	HPMC	Hydroxypropyl methylcellulose
PHPA	Partially hydrolyzed polyacrylamide	CMSEC	Carboxymethylsulfoethyl cellulose
HAWP	Hydrophobically associating water-soluble polymer	HPC	Hydroxypropyl cellulose
PV	Pore volume	HEMC	Hydroxyethyl methyl cellulose
NaCl	Sodium chloride	HEMCMC	Hydroxyethylmethylcarboxy cellulose
MWD	Molecular weight distribution	DP	Degree of polymerization

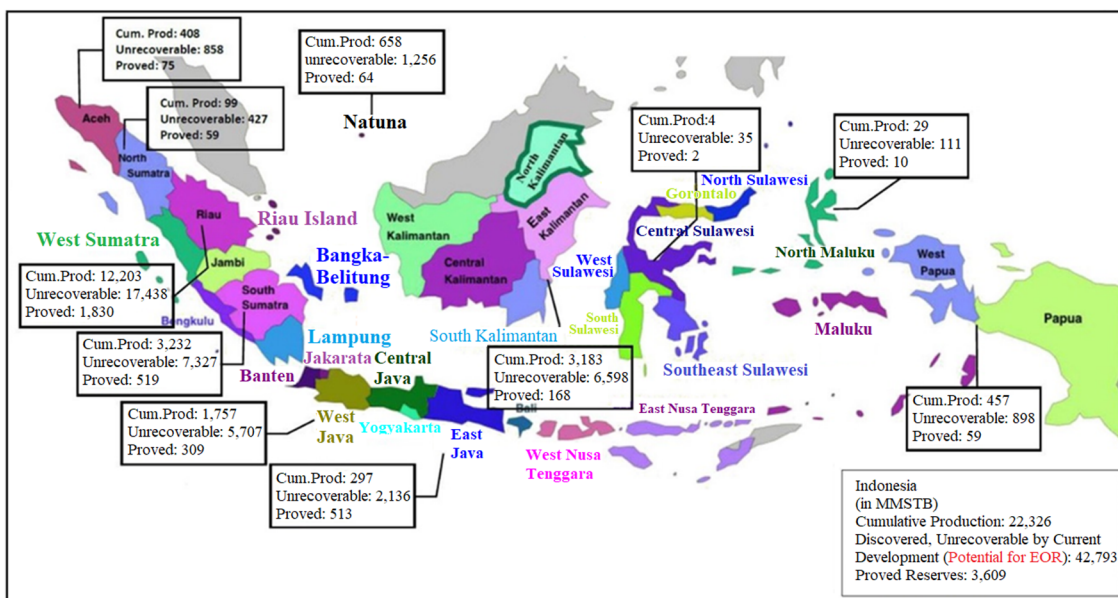


Fig. 1. Status of potential EOR targets in Indonesia as at January 1, 2010 [2].

## INTRODUCTION

Polymers have been widely applied in many kinds of industries in the world, including the chemical, textile, construction, agricultural, transportation, as well as the oil and gas industry [1]. In the petroleum industry, polymers are used for enhanced oil recovery (EOR), where they act as a thickening agent in a method known as polymer flooding. Polymer flooding is a chemical injection method that is used to improve oil recovery, and has reportedly been successfully applied in some oilfields since the 1950s [1].

Figure 1 shows the potential EOR targets in Indonesia based in discovery of oil accumulations. However, these targets have yet to be reached with the technology that is currently available. According to the description of the EOR potential in Indonesia, there are large amounts of residual oil remaining after the primary and secondary oil recovery processes. Hence, the application of EOR methods can efficiently increase the oil recovery [2]. Polymer flooding has been widely used as an EOR technique because the application is relatively simple and effective for mobility control [3, 4]. The largest polymer flooding in the world was implemented in the Daqing oilfield in 1996 [5], and by 2007, about 22.3% of the total production from the oilfield was attributed to polymer flooding [6]. Generally, the injection process involves adding polymers into injected water to increase the water

viscosity and improve oil recovery. Subsequently, the mobility ratio of the injected water/oil and the relative permeability of the water in the reservoir will decrease [7, 8], hence enhancing the sweep efficiency [9].

Polymers are macromolecules that consist of a large number of repeating structural units (also known as monomers) that are linked together by well-defined chemical bonds. When the number of monomers is very large, the compound is referred to as a high molecular weight polymer. Meanwhile, lower molecular weight polymers are sometimes called dimers, trimers, tetramers, pentamers, etc. The properties of polymers are strongly related to their molecular weight [10]. Besides that, polymers are classified into two types, i.e., synthetic (man-made) polymers and naturally-occurring polymers (biopolymers). Both synthetic and natural polymers are being used in EOR, with HPAM (synthetic-based) and xanthan gum (bio-based) being the most commonly used polymers in EOR projects [11, 12]. This is because HPAM is inexpensive and is known for its good viscosifying and established physicochemical characteristics, while xanthan gum is popular for its outstanding performance in high salinity brine and relative compatibility with most of the surfactants and other injection fluid additives utilized in oil recovery [3]. The characteristics and screening criteria of the polymers commonly used in EOR are presented in Tables 1 and 2.

**Table 1.** Structure and characterizations of polymers [11]

Structure	Characteristic	Polymers sample
Carbon chain in the backbone	Has excellent thermal stability, no severe degradation at <math><110^{\circ}\text{C}</math>	HPAM, polyacrylamide, sodium polyacrylate, polyvinyl
–O– in the backbone	Low thermal stability, thermal degradation at high temperatures, only suitable at <math><80^{\circ}\text{C}</math>	Polyoxyethylene, sodium alginate, Na-CMC, HEC, xanthan gum
–COO– in hydrophilic group	Good viscosifier, less adsorption on sandstones to the repulsion between chain links, but precipitation with $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$ , less chemical stability	Sodium alginate, Na-CMC HPAM, xanthan gum
–OH– or –CONH <sub>2</sub> in hydrophilic group	No precipitation with $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$ , excellent chemical stability, but there is no repulsion between chain links, thus less viscosifying power, high adsorption due to hydrogen bonds formed on sandstone rocks	HEC, polyacrylamide, HPAM

**Table 2.** General screening criteria for polymer flooding [13]

Crude oil parameter	Recommended value	Reservoir parameter	Recommended value
Viscosity, mPa s	<math><150 (>1200 \text{ in low-thickness fields})</math>	Type of formation	Sandstones preferred (carbonates also possible)
Gravity, °API		Depth, m	<math><2.800</math> (due to thermal restrictions)
Composition fluid		Temperature, °C	<math><95</math>
		Initial reservoir pressure, MPa	Not critical
		Net thickness, m	Not critical
		Porosity, %	Not Critical
		Permeability, $10^{-9} \text{ m}^2$	>20 (preferred >50)
		Initial oil saturation, % PV	>50
		Total dynamic solute, mg/L	<math><20.000</math>
		Total divalent cations, mg/L	<math><500</math>
		Special remarks	Low Ca clay content

On the other hand, polymer flooding may impact the environment, where it has been associated with (1) erosion, sedimentation, and loss of biota, (2) water quality degradation, (3) aquifer contamination in the groundwater and air pollution [14]. Hence, an understanding of the properties and rheological behaviour of the polymers used for EOR processes is vital for minimizing the environmental pollution.

#### POLYMER SELECTION

**Synthetic polymers** are man-made polymers with backbones that consist of relatively stable carbon-

carbon bonds. Examples of synthetic polymers that are generally used in EOR are polyacrylamides and their derivatives, including hydrolysed polyacrylamide (HPAM), hydrophobically associating polyacrylamide (HAPAM), and copolymers of acrylamide [15]. Synthetic polymers are inexpensive, with good viscosity in fresh water and acceptable adsorption on rock surfaces [16]. However, synthetic polymers exhibit poor thermal stability and would be hydrolyzed at high temperatures, thereby creating a higher charge density of anionic functionalities along the polymer backbone [17]. This phenomenon tends to subsequently reduce the viscosity

**Table 3.** Synthetic polymers application in EOR

No.	Field	Polymer product	Characteristic	Reservoir temperature, °C	References
1	Daqing Oilfield, China (Onshore)	HPAM	Polymer solution viscosity, mPa s: 35–40 Polymer solution concentration, mg/L: 1000 Sandstone reservoir	45	[5]
2	Shengli Oilfield, China (Onshore)	HPAM	Formation water salinity, mg/L: 10000 Polymer solution concentration, mg/L: 5000 Apparent of polymer viscosity, mPa s: $\geq 11.5$ Sandstone reservoir	70	[19]
3	Yanganmu Oilfield, China (Onshore)	3730S Hengju (KYPAM-2) TS-65 (Zhangjiakou)	Polymer solution concentration, mg/L: 1000 Polymer solution viscosity 3730S : 32 mPa s (63°C) Polymer solution viscosity Hengju (KYPAM-2): 55 mPa s (63°C) Polymer solution viscosity TS-65 (Zhangjiakou): 850 mPa s (63°C) Sandstone reservoir	62	[20]
4	Yariguí-Cantagallo Field (Pilot project)	HPAM	Polymer solution concentration, mg/L: 600 Polymer injection viscosity, mPa s: 22 Polymer pore volume, % PV: 0.1–0.5 Sandstone reservoir	59	[21]
5	Sanand Field, India (Onshore)	PHPA	Polymer solution concentration, mg/L: 800 Formation water salinity, mg/L: 9880 Sandstone reservoir	81	[22]
6	Dalia Offshore Oilfield in Angola (Offshore)	HPAM	Formation water salinity. g/L: 110–120 Polymer solution concentration, mg/L: 900 Formation water salinity, mg/L: 20000 Polymer solution viscosity, mPa s: 7.5 Sandstone reservoir	50	[23]
7	El Corcobo Norte field, Argentina (Onshore)	HPAM	Polymer solution concentration, mg/L: 1400 Formation water salinity, mg/L: 46000 Sandstone reservoir	38	[24]
8	SZ36-1 Oilfield of Bohai Bay, China (Offshore)	HAWP	Formation water salinity, mg/L: 9048 Polymer solution concentration, mg/L: 1500 Sandstone reservoir	65	[25]
9	Grimbeek II Field, South of Argentina (Pilot project)	HPAM	Polymer solution concentration, mg/L: 2500/3000 Carbonate reservoir	60	[26]
10	East Bodo oilfield, Alberta Canada (Onshore)	HPAM	Polymer solution concentration, mg/L: 500–1500 Polymer solution viscosity, mPa s: 25 Sandstone reservoir	23	[27]

of the polymer solution; thereby rendering entire polymer injection process inefficient [18]. Moreover, this group of polymers is highly sensitive to salinity, flow rate, shear forces, pH, and hardness [15, 16, 18]. The disadvantages of synthetic polymers have restricted their applications

in high temperature and high salinity (HTHS) reservoirs. The applications of synthetic polymers in EOR are presented in Table 3.

**Hydrolysed polyacrylamide (HPAM).** The synthetic water-soluble polymer, HPAM, has been extensively



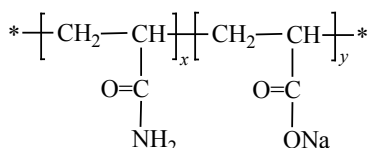


Fig. 2. Chemical structure of HPAM [11].

used for EOR activities in oil industries throughout the world HPAM can be obtained by the partial hydrolysis of polyacrylamide or by the copolymerization of acrylamide and acrylic acid [11, 28, 29]. Figure 2 shows the chemical structure of HPAM [11]. In polymer flooding, HPAM solution, at a concentration ranging from several hundreds to thousands of ppm, is injected with the formation water, thereby causing the oil to move into the production well. The slug used in polymer flooding ranges between 50 to 100% pore volume (% PV) [2]. Sheng [11] and Seright et al. [9] explained that when the degree of hydrolysis is more than 40% at temperatures above 60°C, the acrylamide group in the HPAM will undergo hydrolysis to form acrylate groups. At higher levels of hydrolysis (greater than 40%), the flexible chains of HPAM are severely compressed and distorted, resulting in a decrease in viscosity [30]. Moreover, HPAM can form flocs with  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  cations in hard water solution [30]. However, the stability of the polymer needs to be taken into consideration due to the relatively long EOR process. Generally, after three months, the degree of hydrolysis must be less than 40% [30].

Moreover, the hydrolysis of HPAM occurs very quickly under both acidic and basic conditions, as well as at high temperatures above 60°C [9, 11, 30]. Seright et al. [9] carried out detailed experiments using different temperatures to evaluate the temperature limit for the stable application of HPAM in EOR. According to their findings, in the absence of dissolved oxygen and divalent cations, the HPAM backbone can remain stable for more than 8 years at 100°C and for 2 years at 120°C, while maintaining at least half of its original viscosity. Besides that, the viscosity of HPAM can decrease significantly because it is designed to be more sensitive to high salinity and the hardness of the formation water in offshore oilfields [11]. According to Seright et al. [9], the viscosity of the HPAM solution increased by about 12.4 mPa s (i.e. from 31.6 to 44 mPa s) after 3 days at 120°C in low-salinity brine (0.3% NaCl). Meanwhile, in high-salinity brine (3% NaCl), the increase in the

viscosity of the HPAM-solution was insignificant (only 0.1 mPa s, i.e. from 39.3 to 39.4 mPa s) on exposure to the same conditions. HPAM creates a negative charge on the carbon polymer chains, which has a significant effect on the rheology of the polymer solution. At low to moderate levels of salinity, the negative charges on the carbon polymer chains will repel each other, hence contributing to the extension of the chains and the resultant high viscosity [30]. One of the limitations of low salinity is the decline in injectivity due to fluid incompatibility, excessive polymer retention and accumulation of poorly dissolved polymer molecules, impurities or additives at the pore throats [31]. It is known that a severe decline in injectivity can cause a major downtime for cleaning jobs, and potentially lead to the economic failure of a project. Another limitation of low salinity is the loss in viscosity because of the cation exchange and/or mixing with the formation water [31].

Polyacrylamide, which is used in EOR, has an average molecular weight ranging from  $2-10 \times 10^6$  g/mol, while HPAM, which is used in polymer flooding, has a molecular weight of  $9 \times 10^6$  g/mol [32]. HPAM is usually used in EOR projects due to its abundant availability and low cost [33]. It has excellent viscoelasticity compared to xanthan gum as the polyacrylamide strongly absorbs minerals [11]. It also has potential as a mobility control agent to improve waterflooding operations [32] and has been reported to be successful in low-salinity formations [34]. Unfortunately, this particular polymer is not resistant to salinity formations comprised of high levels of brine, divalent ions, and at high reservoir temperatures [34,35].

**Salinity-tolerant polyacrylamide (KYPAM).** KYPAM is a new product from China. This polymer is a commercial product that is used in polymer flooding projects such as in the Daqing and Shengli oilfields [33]. In May 2001, KYPAM solution was injected for EOR in the Lamadian Field, Daqing oilfield [33]. With the use of KYPAM for EOR, the average water cut was successfully decreased by 15% in comparison to the use of conventional HPAM [36]. The chemical structure of this polymer is shown in Fig. 3.

There are several types of KYPAM products, including RSP1, which is used for treating drilling fluids; RSP2, which is used in EOR; and RSP3, which is used for water shut-off or profile control. RSP2 is the most commonly used KYPAM product commercially [11]. Based on Fig. 3,  $R_1, R_2, R_3,$  and  $R_4$  can be alkyl-H or  $C_1-C_{12}$ , whereas A in the structure represents ionic functional groups that



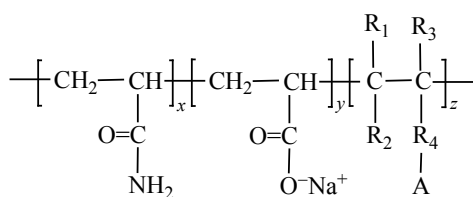


Fig. 3. Chemical structure of KYPAM [11].

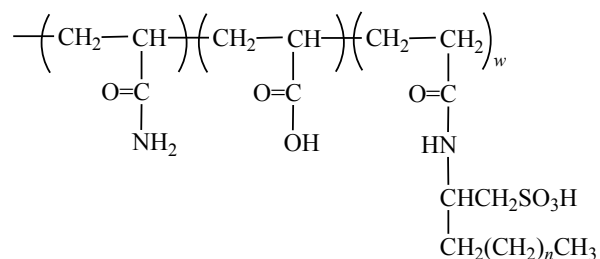


Fig. 4. Chemical structure of HAP [11].

are tolerant toward  $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$  [11]. The elasticity of KYPAM is mainly affected by R3. As the carbon number increases, the elasticity of the polymer increases. R affects the tolerance of the polymer to salinity, where it was found that with increasing carbon, the tolerance to salinity increases [11]. KYPAM was developed by the Research Institute of Petroleum Exploration and Development (RIPED), and the physiochemical properties of KYPAM are shown in Table 4 [37]. Additionally, Table 5 shows the comparison between the physiochemical properties of HPAM and KYPAM [11].

**Hydrophobically associative polymers (HAP).** HAP are water soluble polymers that consist of a small fraction of hydrophobic functionality (<1%) in the polymer backbone [25, 33]. The chemical structure of HAP is shown in Fig. 4 [11]. In recent years, these polymers have developed very quickly, one such example being as the HAP A-DH, which was developed by South West Petroleum University, the characteristics of which are shown in Table 6 [37].

HAP has a small number of hydrophobic groups that are attached directly to the polymer backbone [25].

Table 4. Physiochemical properties of KYPAM [37]

Parameters	KYPAM
Appearance	White powder
Molecular weight, $\times 10^6$ g/mol	15–25
Intrinsic viscosity, mPa s	2530
Hydrolysing degree, %	24.6
Solid content, wt %	886
Filtrating factor	1.39
Insoluble, wt %	0.17
Dissolution time, h	< 2

Table 5. Physiochemical properties of KYPAM and HPAM polymers [11]

Parameters	KYPAM	HPAM
Appearance	White powder	White powder
Solid content, wt %	90.0	90.2
Molecular weight, $\times 10^6$ g/mol	25.14	17
Hydrolysis, %	26.4	26.8
Dissolution time, h	$\leq 2$	$\leq 2$
Insoluble, wt %	0.115	0.19
Residual monomers, %	0.0096	0.021
Filtration index	1.12	1.22
Screen coefficient	102.6	41.3

**Table 6.** Physiochemical properties of the HAP A-DH [37]

Parameters	A-DH
Appearance	White powder
Molecular weight, $\times 10^6$ g/mol	8–12
Intrinsic viscosity, mPa s	1386
Hydrolysing degree, %	23
Solid content, wt %	90
Filtrating factor	1.29
Insoluble, wt %	0.08
Dissolution time, h	< 2

HAP has a rather low molecular weight [37]. The critical concentration ( $C^*$ ) can be used to determine the viscosity of HAP. If the concentration of the polymer is less than the value of  $C^*$ , it will cause the interaction of polymer molecules to be more dominant and the polymer chain to be circular. If the concentration is close to or higher than  $C^*$ , then the intermolecular interaction will be stronger, and this will result in a sharp increase in polymer viscosity [25]. The advantage of HAP is that it has strong salt tolerance where it can withstand high salinity water, while the disadvantage is that it has weak solubility and injection ability, which can result in a high injection pressure and steep pressure gradient [37].

HAP was successfully applied in a single injector pilot test in Liaodong, Bohai Bay (SZ36-1 oilfield), China [25]. Table 7 presents the reservoir characteristics of the SZ36-1 oilfield. The corresponding well obtained incremental oil of about 25 000 m<sup>3</sup> and the water cut was reduced from 95% to 54% [25].

**Natural polymers** are also known as biopolymers. Both synthetic and natural polymers consist of repeating monomeric units that are bonded into larger formations [38]. The advantage of a natural polymer is that its viscosity is suitable for high levels of salinity, but is unsuitable when mixed with freshwater or low-salinity water [39]. Natural polymers are easily available as they are present in plants and animals [39, 40]. The use of natural polymers for oil and gas applications is very attractive as they are economically effective and their production cost is inexpensive compared to when using synthetic materials [41]. Moreover, natural polymers are known to be non-toxic, safer to use, and biodegradable [38].

The applications of biopolymers in EOR are presented in Table 8.

**Table 7.** Reservoir characteristics of SZ36-1 oilfield [25]

Depth, m	1300–1600
Average pay thickness, m	61.5
Porosity, %	28–35
Average permeability, $10^{-9}$ m <sup>2</sup>	2600
Original reservoir pressure, MPa	14.28
Reservoir temperature, °C	65

The disadvantage of natural polymers is that they are susceptible to contamination during production where they might be exposed to microbial contamination as well as heavy metal contamination due to exposure to the external environment [42]. Additionally, the percentage of chemical constituents present in different natural materials may vary according to species, time, region, and climate conditions. Synthetic polymers are made using controlled procedures and fixed quantities of ingredients while the production of natural polymers is dependent on the environment and various physical factors, which means a much slower rate of production [41].

Natural polymers can be classified according to their origin, whether they are plant based or animal based. Examples of plant-based natural polymers are cellulose, hemicellulose, glucomannan, pectin, inulin, starch, agar, guar gum, locust bean gum, acacia gum, karaya gum, tragacanth gum, and aloe vera gel. Meanwhile chitin, xanthan gum, alginates, carrageenans, and psyllium are examples of animal-based natural polymers [38, 46]. However, the focus of the current research on the topic of biopolymers is to explore the advantages of cellulose, xanthan gum, and scleroglucan since they have been tested for use in the oil and gas industry. Table 9 summarizes the typical natural polymers used in EOR [49].

**Cellulose.** Cellulose is an organic polysaccharide that was discovered in 1883 by the French chemist, Anselme Payen. Its formula is  $(C_6H_{10}O_5)_n$  and it consists of a linear chain of several hundred to over ten thousand  $\beta$ -(1 $\rightarrow$ 4) linked D-glucose units.

Figure 5 shows the molecular structure of cellulose. The polysaccharides of the plant cell wall consist mainly of cellulose, hemicelluloses, and pectin [38].

Indonesia, which is a tropical country with a population of 220 million, has a substantial need for cellulose. In 1978, Indonesia was only able to produce

**Table 8.** Biopolymer applications in EOR<sup>a</sup>

No.	Field	Status	Biopolymer type	Characteristic	References
1	Bockstedt Oilfield in Northern Germany (Onshore)	Pilot project	SPG	Salinity concentration (NaCl and CaCl <sub>2</sub> ), g/L: 130 and 50 Temperature SPG up to 135°C Biopolymer concentration, mPa s: 300 Viscosity of biopolymer, mPa s: 25 Sandstone reservoir	[12], [27], [43], [44]
2	The Sabriya-Maudud (Sama) Reservoir, in Kuwait (Onshore)	A methodology for the preparation and characterization	SPG	An optimal viscosity of 35 mPa s at 22°C Acceptable salinity for use, g/L: 180 Carbonate reservoir	[45]
3	SZ36-1 Oilfield in Bohai Oilfield, China (Offshore)	Pilot and further application	Xanthan	Calcium and magnesium components, mg/L: 568; 228 Sandstone reservoir	[46]
4	Safaniya Oilfield, Saudi Arabia (Onshore)	Laboratory models	Xanthan	Molecular weight, g/mol: $9.6 \times 10^6$ Polymer concentration, %: 0.15, 0.20 Formation water salinity, %: 3.5 Sandstone reservoir	[47]
5	Eddesse-Nord Sandstone Reservoir, Germany (Onshore)	Pilot project	Xanthan	Concentration xanthan solution, mPa s: 800 Salinity of reservoir brine, g/L: 120 Salinity of injection water, g/L: 50 Viscosity of biopolymer, mPa s: 12 Sandstone reservoir	[48]

<sup>a</sup> SPG—Schizophyllan; CaCl<sub>2</sub>—calcium chloride.

**Table 9.** Characteristics of natural polymers used in EOR [49]

Natural polymer	Source	Molecular weight, g/mol	Properties
Xanthan gum	Fermentation product of <i>Xanthomonas campestris</i>	$2 \times 10^6$ to $2 \times 10^7$	Thickening Crosslinking
Scleroglucan Guar gum	Fermentation product of <i>Sclerotium rolfsii</i> Endosperm component of <i>Cyamopsis tetragonolobus</i>	$1.3 \times 10^5$ to $6 \times 10^6$ $10^6$ to $2 \times 10^6$	Thickening Thickening Crosslinking
Cellulose	Lignocellulose of plants Fermentation product of <i>Acetobacter Xylinam</i>	$2 \times 10^6$	Thickening Filtration Adsorption
Chitin/Chitosan	Shells of crustaceans, exoskeletons of insects, and cell walls of fungi	$2 \times 10^3$ to $10^6$	Adsorption

455 tons, the cellulose out of its total requirement of 91 300 tons. In 1988, the production was 5 000 tons, whereas the demand was for 200 000 tons. In 1995, Indonesia became the second-largest importer of cellulose in the world [50].

Hence this natural polymer is becoming increasingly important. Beside, being environmentally friendly, the other advantages of cellulose are that is low cost, non-

toxic, non-melting, and dimensionally [51]. Cellulose, which has a substantial molecular weight, is found in the form of microfibrils with diameters ranging between 2–20 nm and lengths ranging between 100–40000 nm [52].

Cellulose has no colour, taste nor smell, is insoluble in water, relatively stable at high temperatures, resistant to hydrolysis, stable with regard to oxidation, does not melt when heated and decomposes at 260–270°C. However,

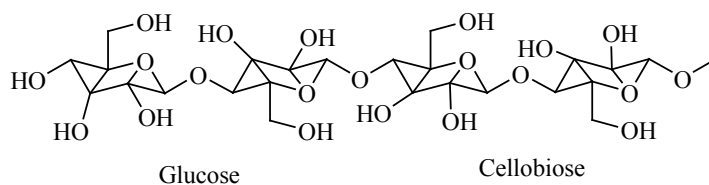


Fig. 5. Molecular chain structure of cellulose [58].

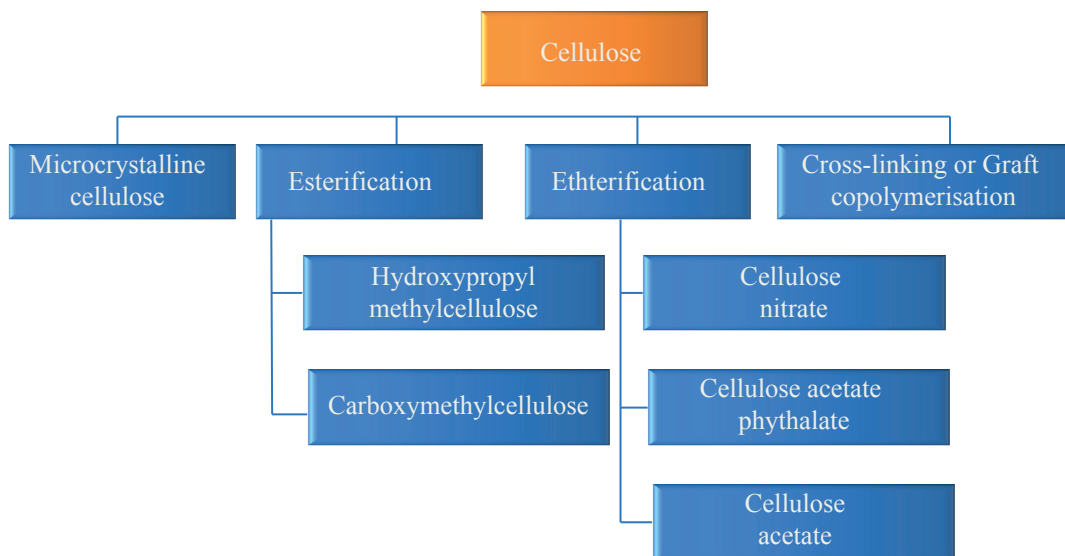


Fig. 6. Cellulose derivatives [38].

cellulose will dissolve in high concentrations of mineral acids through hydrolysis. For instance, Chen [53] found that cellulose can be completely dissolved through the use of 72% sulphuric acid or 42% hydrochloric acid or 77–83% phosphoric acid at 100°C. Hence, if the hydrolysis is incomplete, cellulose can be re-precipitated to form fragments of polymer solids by diluting the solution in a strong acid [53]. Cellulose exists in abundance on the earth as it can be found in every part of a plant i.e. its roots, stems, and twigs, making it a virtually inexhaustible source of raw material. Cellulose has been successfully utilized as wetting, binding, thickening, packaging, film-forming, gel-forming, and emulsifying agents. These are good reasons to use cellulose on a large scale in industries as construction materials, industrial paints, industrial papers, textile materials, detergent raw materials, cosmetics, foods, as well as in the oil and gas industry [54].

Cellulose can be chemically modified to improve its processability and to produce cellulose derivatives (cellulosic), as shown in Fig. 6 [38], which can

be adapted for specific industrial applications. Carboxymethylcellulose (CMC) is a type of polymer derivative that has high solubility in water and good thermal stability (up to 149°C) [55, 56]. In general, CMC is a biodegradable, non-toxic, and odourless powder with a creamy white colour. Moreover, it is widely used in the industry due to its excellent rheological properties in aqueous solutions [55].

One study indicated that a cellulose-based polymer system, namely CMC graft polymer, was suggested for use in polymer flooding at an offshore reservoir in Norway; however, it is still not optimal for use in high salinities and temperatures (harsh conditions) [57]. According to Jouenne [42], the development of a polymer that can withstand harsh conditions (salinity up to 280 g/L and temperatures higher than 100°C) would unlock a great number of reserves. Therefore, further research is still needed to optimize the cellulosic polymer so that it can be used as an effective material for polymer flooding in oilfields. Figure 5 shows the molecular chain structure of cellulose [58].

**Table 10.** Applications of cellulose and its derivatives [54]

Application	Function and product application	Cellulose derivative
Drilling oil and gas industry, mining	Water retention, flow characteristics, surface activity	CMC, CMSEC, HEC, HPC, HPMC
Polymerization	A protective colloid, surface activity	HEC, HPC, HPMC
Detergents	Anti-redeposition power, wetting ability, suspending and emulsifying agents	CMC, HEMC, HPMC
Pharmacy	Gelling agent, suspending agent, stabilizer, coating agent, water-absorbing agent, disintegrant in tablets and medicine capsules Examples of products are ointments, jellies, and syrups	CMC, MC, HEC, HEMC, HPMC
Foodstuffs	Food additives which function as flavour enhancers, emulsifiers, water binding, ice crystal growth control, thickening, control of water content in food. Examples of products are sauces, frozen food, pet food, protein food, bakery products	CMC, HPMC, MC
Cosmetics	Friction reduction, thickener, stabilizer, water retention, skin protector. Examples of products are toothpaste, soap and shampoo, body lotion cream	CMC, MC, HEC, HEMC, HPMC
Textile industry (sizes, textile printing dyes)	Adhesive and film-forming properties, thickening, soil release, colour booster, thickener and water binder. Examples of products are fabrics, clothing, and laundry	CMC, MC, HPMC, CMSEC
Paper manufacture	Internal additive, pigment coating, accelerates drying of paper, agent for binding and suspending, sizing aids and stabilizers. Examples of products are pulp and paper	CMC, HEC, HEMC, HPMC
Paints, Photography	Stability of suspension, thickening, film formation, wetting, dyes, water inks, colour boosters, and water binders. Examples of products are binders, photos	CMC, HEC, HEMC, HPMC, HEMCMC

Cellulose makes up the structure of plant cell walls, and as it cannot be digested by humans, it is a potential source of fiber that helps smoothen defecation. For broader applications, several products can be derived from cellulose such as microcrystalline cellulose (MCC), CMC, methylcellulose (MC), and hydroxypropyl methylcellulose (HPMC). These products are used, among others, as emulsifiers, stabilizers, thickeners, and dispersing and gelling agents. The applications of cellulose and its derivative products are presented in Table 10 [54].

**Xanthan Gum** is produced by a bacterium and was one of the first commercially successful bacterial polysaccharides to be produced by fermentation. It is a cream-coloured powder that quickly dissolves in hot or cold water to form a viscous, non-thixotropic solution. Xanthan gum produces a thick solution when used in low concentrations and shows slight changes in thickness with the changes in the temperature [38]. Additionally, it is stable in a broad range of pH values as well as when

frozen [38]. Traditionally, gums have been derived from a wide variety of plants. Chemically, gums are carbohydrate polymers or polysaccharides (however, gelatine is a protein). Microbial polysaccharides are composed of regularly repeating units of simple sugars such as glucose, mannose, fructose, etc. The primary structure of xanthan gum consists of a backbone of glucose monomers (cellulose-like chain) and trisaccharide side chains. The backbone consists of glucose residues connected by  $\beta$ -(1 $\rightarrow$ 4) glycosidic linkages. A side chain that contains the sequence mannose/glucuronic acid/mannose is attached to every other glucose unit. In each side chain, an O-acetyl group is usually attached to the mannose closest to the polymer backbone [29]. The structural unit of xanthan gum is displayed in Fig. 7 [59].

Alquraishi et al. [60] investigated the influence of salt concentrations on the rheology of xanthan and guar solutions, as well as their combined mixtures. They observed that xanthan was more viscous than guar due to the nature of its chemical structure i.e. its higher

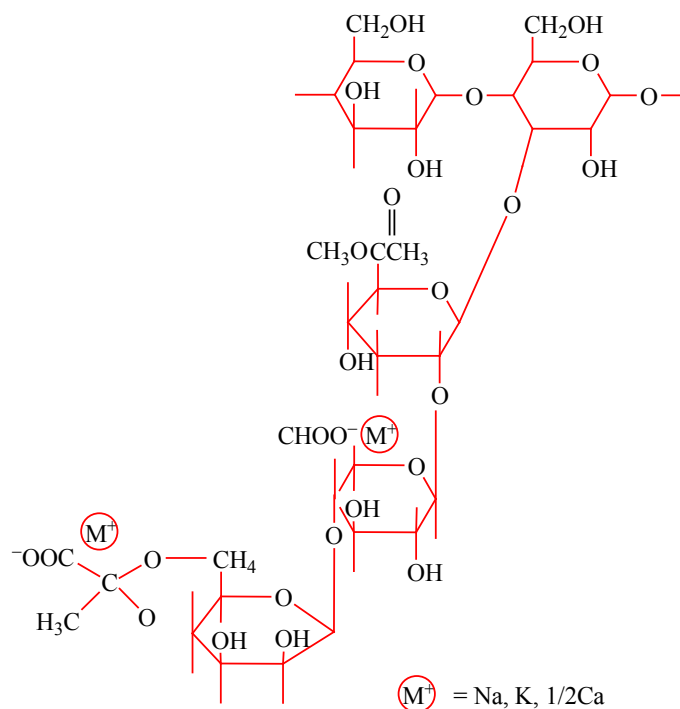


Fig. 7. Structural unit of xanthan gum [59].

molecular weight. Despite this fact, the guar and xanthan mixtures exhibited a synergism, where the viscosity of the mixtures was found to be higher than the viscosity of the neat gums. They suggested a combination of xanthan and guar solution as is an effective strategy for controlling the rheology and morphology of the solutions used in the field of EOR.

Xanthan gum is suitable for the petroleum industry because of its unique rheological properties, namely, its excellent compatibility with salt formations and resistance to thermal degradation. Hence, xanthan gum is applied as an additive in oil drilling fluids [61]. Additionally, xanthan gum is applied as a thickener in tertiary oil recovery operations [62]. The xanthan gum-thickened brine is used to drive the slug of the surfactant through porous reservoir rocks to mobilise the residual oil, thereby preventing the drive water from bypassing through the surfactant band and ensuring good sweeping of the area [63]. Another work by Jang et al. [64], examined the characteristics of xanthan gum and HPAM for polymer flooding to explore the possibility of improving heavy oil recovery. A detailed schematic diagram of the experimental set-up is shown in Fig. 8 [65]. To perform the polymer flooding tests, the core holder was filled with glass beads, and compacted tightly for more than 1 h. The core holder was packed with

glass beads, and placed vertically, after which, the brine was injected from its inlet. Heavy oil was also injected in the same direction until the oil reached the outlet. The effect of a salinity of 3 wt % on the polymer solutions at a concentration of 1500 mPa s in a local reservoir was investigated. The final recovery of the original oil was higher (61.9%) with the use of the xanthan gum solution at a salinity of 3 wt % than with the use of the HPAM solution (61.9%). The main purpose of using polymers in the EOR process is to increase the viscosity of the solution and reduce the rock permeability, both of which will help increase the efficiency by reducing the mobility of the displacing fluid [66]. Table 11 presents the utilization of xanthan gum in various applications [67].

**Starch** is a polysaccharide compound consisting of monosaccharides that are bonded by oxygen bonds (Fig. 9) [38]. Starch can be derived from grains such as corn, wheat, rice, sorghum and nuts, and can also be produced from tubers such as cassava or potatoes [38]. Apart from these two sources, starch can also be produced from plant stems, such as sago starch, and young fruit flesh like bananas. Tuber starch has a gelatinization temperature ranging between 70–80°C, is elastic, easily damaged, and has a translucent appearance when cold. Grain starch has a higher gelatinization temperature of

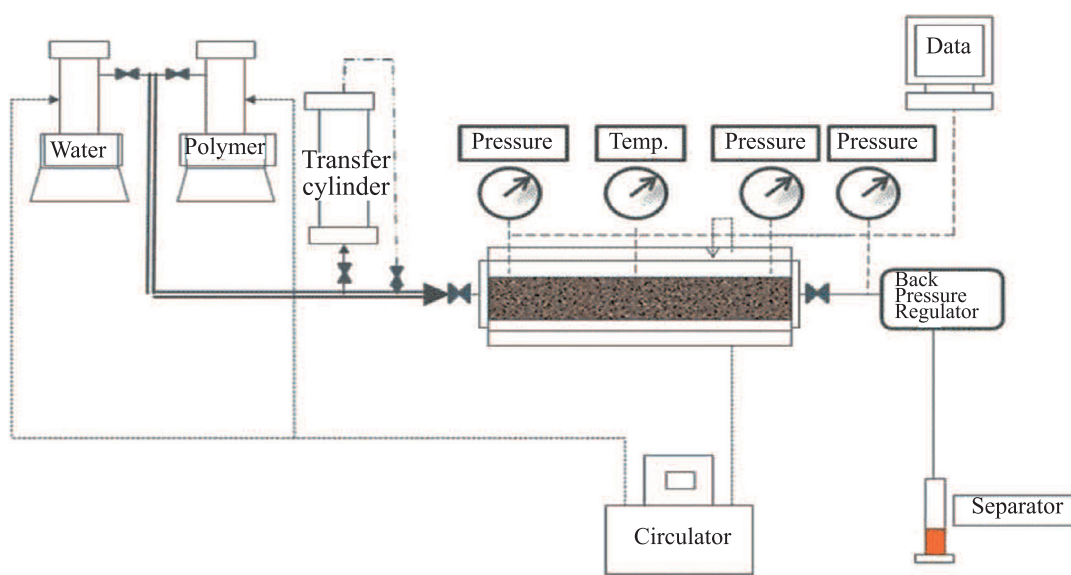


**Table 11.** Utilization of xanthan gum in various applications [67]

Application	Concentration, % w/w	Functionality
Petroleum production	0.1–0.4	Lubricant or to reduce friction in the borehole
EOR	0.05–0.2	Reduces water mobility ratio by increasing the viscosity and decreasing the fluid permeability
Slurry explosives	0.3–1.0	Thickens formulation to improve heat stability (in combination with guar gum)
Ceramic glazes	0.3–0.5	Prevents agglomeration during grinding of ceramics
Textile printing and dyeing	0.2–0.5	Control of rheological properties of paste, preventing dye migration
Pharmaceuticals (creams and suspensions)	0.1–1.0	Emulsion stabilizer, uniformity in dosage formulations
Cosmetics (denture cleaners, shampoos, lotions)	0.2–1.0	Thickener and stabilizer
Agriculture (additive in animal feed and pesticide formulations)	0.03–0.3	Suspension stabilizer, improves spray ability, reduces drift, increases cling and permanence
Salad dressings	0.1–0.5	Emulsion stabilizer, suspending agent, dispersant
Dry mixes	0.05–0.2	Facilitates dispersion in hot or cold water
Syrups, toppings, relishes, sauces	0.05–0.2	Thickener, provides heat stability and uniform viscosity
Beverages (fruit and non-fat dry milk)	0.05–0.2	Stabilizer
Frozen foods	0.05–0.2	Improves freeze-thaw stability
Dairy products	0.5–0.2	Stabilizer, controls viscosity of the mix

95°C, is in the form of a gel, and has an opaque appearance when cold. In general, grains contain 75% carbohydrates in the form of starch, cellulose, hemicellulose, and pectin, 10–14% protein, 1–2% fat, 10% water, and 12% ash (mineral).

Generally, starch is an important biopolymer that consists mainly of two polymers, namely, amylose (20–30%) and amylopectin (70–80%) [68]. Amylose has a straight structure with  $\alpha$ -(1→4)-D-glycosidic bonds and is water soluble [69]. Furthermore, it is also known to

**Fig. 8.** Schematic diagram of experimental apparatus for polymer flooding [66].

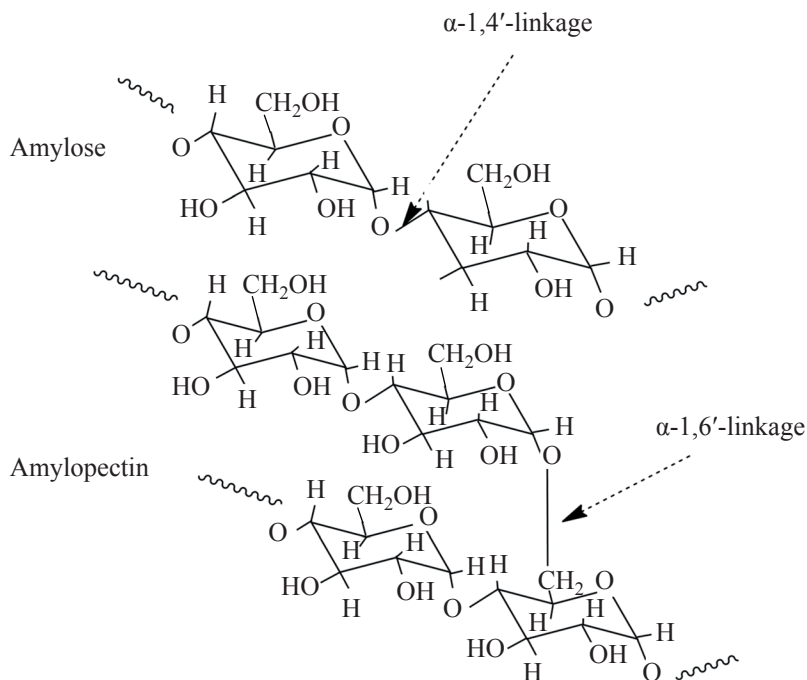


Fig. 9. Molecular structure of starch [38].

form complexes when interacts with iodine, fatty acids and organic alcohols. The formed complexes are generally referred to as helical inclusion complexes or amylose-lipid complexes [70]. The degree of polymerization (DP) from amylose ranges between 500–6000 glucose units. Amylopectin, which has  $\alpha$ -(1 $\rightarrow$ 4) and  $\alpha$ -(1 $\rightarrow$ 6) bonds with branched structures, easily expands and forms colloids in water. The DP from amylopectin ranges between 105 to  $3 \times 10^6$  of glucose units. The DP of amylose and amylopectin is affected by the type of starch. Amylose and amylopectin are the major components of starch, with the others being lipids (about 1%), protein, phosphorus, and minerals. Some lipids bind to amylose, and some are free. The shape and size of the starch granule vary depending on the source of the plant. Rice starch granules are small (between 3–8  $\mu\text{m}$ ), polygonal in shape, and tend to occur as aggregates or lumps.

Corn starch granules are somewhat more massive (about 15  $\mu\text{m}$ ) and have a rounded polygonal shape. Tapioca granules are larger (about 20  $\mu\text{m}$ ), and are commonly round in shape, but conical at one end. Wheat starch granules tend to come in various sizes; with the standard size being 18  $\mu\text{m}$ , the larger granules on average being 24  $\mu\text{m}$ , and the smaller granules being between 7–8  $\mu\text{m}$ . Wheat starch granules are round to oval. In

shape, while potato starch granules are oval and very large, measuring on average between 30–50  $\mu\text{m}$  [38, 71].

Different kind of starches have been used in the enhanced oil recovery applications. For instance, the starch biopolymer from potato peels was used as chemical agent for polymer flooding by Ayoola et al. [72]. They observed that the starch biopolymer shows a good pore-plugging abilities, relative thermal stability, less sensitive to the presence of NaCl, and good rheological properties up shear rates of to 300 rpm. Hence, they suggested that the starch biopolymer can be successfully applied for EOR applications. In another work by Agi et al. [73], the rheological performance of crystalline starch nanoparticles (CSNPs) extracted from plant and fruit was compared with native cassava starch (CS) and xanthan polymer, in extracting oil. They observed that with increase of NPs surface area and temperature, the viscosity of CSNPs and CS solutions increased. On the contrary, the viscosity of the xanthan polymer solution decreases with increasing temperature. Hence, they proposed that CSNP could be a potential material for practical industrial applications including drilling fluids and EOR.

**Limitations and future work.** Polymer flooding is mostly applied to sandstone rock because anionic



polymers such as HPAM are significantly adsorbed onto carbonate rock surfaces. Several factors, including temperature, salinity, and biological activity tend to degrade the polymer used, consequently leading to the loss viscosity and adsorption of the polymer [18]. From an environmental point of view, the use of synthetic polymers in EOR is undesirable. For instance, after injection, HPAM either remains in the reservoir or follows the flow of the dispersed oil/water out of the reservoir via a production well. Subsequently, this will increase the cost, as an additional separation process will be required for recovery. In optimal cases, most of the injected HPAM is retained in the reservoir. Due to tight environmental regulations, industrial processes are mandated to trim down the utilization and spillage of non-biodegradable polymers, that are unintentionally released into the environment. Consequently, biopolymer-based EOR formulations will become more attractive in the future. Biopolymers, including nanocelluloses, polysaccharides, xanthan gum, and schizophyllan, are environmentally friendly candidates which can possibly be fully or partially replace HPAM in future chemical EOR operations [74].

### CONCLUSIONS

Recently, research on biopolymers has been sought after by the industry due to several reasons, including the fact that natural polymers such as cellulose starch, chitosan, lignin, etc. are sufficiently available from a variety of resources and can be processed into biopolymers. Advances in science and technology have enabled the creation and development of biopolymers of various shapes, sizes, structures, properties, and functions like never before. Biopolymers can be easily modified to the required specifications of the polymers used in the oil and gas industry, due to their flexible molecular structure and active group. The application of biopolymers for example, xanthan gum and schizophyllan can be utilized at high salinity and high temperature reservoirs. In addition, biopolymers are well-known as environmentally friendly candidates that can considerably reduce the environment impacts. With this review, it is expected that in the future, the potential of biopolymers from natural resources, especially cellulose, can be utilized as EOR injection chemicals to help improve oil recovery. It is hoped that the application of natural polymers can reduce the use of synthetic polymers, which have many harmful effects on the environment and human health.

### AUTHOR CONTRIBUTIONS

Formal analysis: Muslim Darbi Abdurrahman, Shania Regina, and Muhammad Irtin Syariefudin); Investigation: Muslim Darbi Abdurrahman, Ully Zakyatul Husna, Pang Ai Ling and Mohsen Ahmadipour; Methodology: Muslim Darbi Abdurrahman, Ully Zakyatul Husna, Pang Ai Ling and Mohsen Ahmadipour; Resources: Muslim Darbi Abdurrahman and Agus Arsad; Validation: Radzuan Junin and Agus Arsad; Writing original draft: Ully Zakyatul Husna; Review and editing: Agus Arsad, Pang Ai Ling and Mohsen Ahmadipour.

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### CONFLICT OF INTEREST

The authors declare no conflict of interest requiring disclosure in this article.

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