Partially hydrolyzed polyacrylamide: enhanced oil recovery applications, oil-field produced water pollution, and possible solutions

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Received: 31 October 2021 / Accepted: 19 March 2022 / Published online: 13 October 2022
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Abstract Polymers, such as partially hydrolyzed polyacrylamide (HPAM), are widely used in oil fields to enhance or improve the recovery of crude oil from the reservoirs. It works by increasing the viscosity of the injected water, thus improving its mobility and oil recovery. However, during such enhanced oil recovery (EOR) operations, it also produces a huge quantity of water alongside oil. Depending on the age and the stage of the oil reserve, the oil field produces ~7–10 times more water than oil. Such water contains various types of toxic components, such as traces of crude oil, heavy metals, and different types of chemicals (used during EOR operations such as HPAM). Thus, a huge quantity of HPAM containing produced water generated worldwide requires proper treatment and usage. The possible toxicity of HPAM is still ambiguous, but its natural decomposition product, acrylamide, threatens humans’ health and ecological environments. Therefore, the main challenge is the removal or degradation of HPAM in an environmentally safe manner from the produced water before proper disposal. Several chemical and thermal techniques are employed for the removal of HPAM, but they are not so environmentally friendly and somewhat expensive. Among different types of treatments, biodegradation with the aid of individual or mixed microbes (as biofilms) is touted to be an efficient and environmentally friendly way to solve the problem without harmful side effects. Many researchers have explored and reported the potential of such bioremediation technology with a variable removal efficiency of HPAM from the oil field produced water, both in lab scale and field scale studies. The current review is in line with United Nations Sustainability Goals, related to water security–UNSDG 6. It highlights the scale of such HPAM-based EOR applications, the challenge of produced water treatment, current possible solutions, and future possibilities to reuse such treated water sources for other applications.

Keywords Partially hydrolyzed polyacrylamide · Enhanced oil recovery · Produced water · Acrylamide · Toxicity · Bioremediation

Introduction

The majority of crude oil-producing countries (mainly Middle Eastern countries) belong to arid regions, having a scarcity of freshwater sources, potable water, and
almost no rain throughout the year. Reverse osmosis of seawater, and other technologies, provides majority of water requirements in such countries, which are either inefficient or highly energy-intensive. Apart from oil-producing countries, access to potable and clean water is one of the major challenges, which has been included in the United Nations Sustainable Goals (UNSDG 6). It states that “Ensure availability and sustainable management of water and sanitation for all” (https://sdgs.un.org/goals/goal6). It may come as a surprise that the petroleum industries are one of the largest consumers of fresh or saline water. Daily basis and enhanced oil recovery operations require substantial volumes of water. Thus, oil industries are suffering from two major issues: the requirement of huge volumes of fresh (or normal) water for EOR operations, and the treatment and safe disposal of huge volumes of oil field produced water. As such, this produced water cannot be used for any other applications (such as irrigation, livestock water supply, wildlife watering), or direct disposal, as it is quite a toxic wastewater source. Besides, this jeopardizes UNSDG 6 as well. The current review paper focuses on one of the possible toxic chemicals, generally used in EOR applications: HPAM and its derivatives, and its possible degradation, for better and safe disposal of produced water.

At present, fossil fuels are the main source of greater than 85% of the energy consumption worldwide, and especially crude oil will still play a major role in transportation and other industrial sectors for years to come (Joshi et al., 2022). The difficulty in discovering new reservoirs, the depletion of crude oil in old reservoirs, and pandemic like COVID-19 has led to fluctuations in crude oil prices (Abidin et al., 2012; Braun et al., 2022; Quadri et al., 2015). Extraction of oil as the upstream process of the petroleum industry occurs in three phases (Fig. 1). At the primary stage, the natural internal pressure of the reservoir from aquifer water and gas helps to extract about 5–10% of total oil reserves. At the second stage, 30–40% of oil is recovered (Al-Sulaimani et al., 2011; Lucas et al., 2009; Geetha et al., 2018; Al-Ghailani et al., 2021; Braun et al., 2022). At this stage, injected water extracts additional oil by increasing reservoir pressure. However, researchers reported that approximately 50% of oil remaining in place needs to be retrieved by enhanced oil recovery (EOR) means (Abidin et al., 2012; Braun et al., 2022; Gao, 2016; Joshi et al., 2022). A tertiary oil recovery includes the injection of different materials normally not present in oil wells. Various EOR methods such as thermal, chemical, microbial, and gas injection methods were reported to extract ~5–30% of the remaining oil (Al-Ghailani et al., 2021; Braun et al., 2022; Geetha et al., 2018; Lehmköster et al., 2014).

Chemical EOR includes flooding of polymers, surfactants, and alkaline to improve the recovery of crude oil (Al-Sulaimani et al., 2011; Al-Ghailani et al., 2021; Joshi et al., 2022; Braun et al., 2022; Xue et al., 2022). Polymers, surfactants, and solvents work via changing reservoir characteristics to improve the recovery process (Bai et al., 2015; Xue et al., 2022). Among them, polymers (synthetic and biopolymers) showed

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**Fig. 1** The three phases of crude oil recovery from oil reservoirs
efficient oil recovery when combined with water flooding, because of their viscosity (Braun et al., 2022; Choi et al., 2014; Elshafie et al., 2017; Quadri et al., 2015). It increases the water viscosity (as viscosifying agent) and decreases the reservoir permeability, which helps to displace the oil uniformly from the porous rocks (Abidin et al., 2012; Braun et al., 2022; Elshafie et al., 2017; Stokka, 2018; Xue et al., 2022). The main types of polymers frequently used for EOR are, microbial xanthan gum and synthetic chemicals — polyacrylamide (PAM) and partially hydrolyzed polyacrylamide (HPAM) (Al-Sabahi et al., 2018; Braun et al., 2022; Martin et al., 1983; Xue et al., 2022). HPAM is a water-soluble, commonly injected polymer into oil reservoirs to improve the water flooding process (Xue et al., 2022; Zhao et al., 2008).

Polyacrylamide and partially hydrolyzed polyacrylamide

Polyacrylamide (PAM) is a synthetic polymer of long-chain acrylamide subunits, high molecular weight (1000 to 20×10^6 g/mol), and a wide range degree of hydrolysis (Abidin et al., 2012; Joshi & Abed, 2017). It is extremely soluble in water, but not soluble in organic solvents, such as methanol and hydrocarbons (Lu & Wu, 2002; Braun et al., 2022). PAM is used to produce partially hydrolyzed polyacrylamide (HPAM), which is linear, anionic, water-soluble, stable, polymeric, and high molecular weight chain of acrylamide (C₃H₅NO) monomers (Bao et al., 2010; Braun et al., 2022; Liu et al., 2016; Xiong et al., 2018). In addition, it is highly prone to degradation in high temperature and saline reservoirs (Al-Moqbali et al., 2018). The general chemical structure of PAM and HPAM is shown in Fig. 2. Although both polymers contain acrylamide, they are chemically and biologically different from that monomer (Caulfield et al., 2002).

In comparison with other polymers, HPAM showed the highest viscosity, highlighting its applications in the EOR sector (Choi et al., 2014; Xue et al., 2022). Its viscosity has direct relation to its chemical structure, which has negative charges on the chains’ backbone causing repulsion between the polymer chains. The chain’s repulsion leads to its extension, making the polymer more viscous yielding the wanted viscosity even with a low concentration (Lee et al., 2009).

Uses of PAM and HPAM

PAM and HPAM performance depends on their molecular weight and degree of hydrolysis (Abidin et al., 2012). They are often used in many industrial applications, mainly as friction reducers and viscosity modifiers for oil extraction, water purification, soil conditioning, erosion control, and mining (Berdugo-Clavijo et al., 2019; Braun et al., 2022; Liu et al., 2016; Nyysölä & Ahlgren, 2019; Xiong et al., 2018; Yu et al., 2015). In the papermaking, PAM works as a strengthener for pulp fiber, so we have to be careful about wastepaper discharges into the environment (Lu & Wu, 2002; Matsuoka et al., 2002). Usually, PAM is used in sewage treatment, where it helps to improve sedimentation and for purification of drinking water (Filippini, 1986; Yu et al., 2015). In addition, it plays a big role in the agricultural field in the case of herbicide application, which helps to increase the viscosity of the sprayed solution. Moreover, it is also used in capillary electrophoresis for DNA sequencing (Caulfield et al., 2002).

On the other hand, the application of PAM in biomedical fields is associated with some concerns, since it contains the neurotoxic acrylamide monomer. The usage reluctance is mainly where the polymer encounters the patient’s blood for filtration or renal dialysis. Similarly, it is not as safe as a component of therapeutic products. The main issue is about the risk of residual acrylamide monomer presence with PAM polymeric material and the dangers of its degradation (Caulfield et al., 2002). According to the Environmental Health Criteria (EHC) number 49 of acrylamide, published

![Figure 2](image-url)
in 1985 by the World Health Organization (WHO), the source of acrylamide in the environment is mainly released from polyacrylamide usage (Filippini, 1986).

Polymers like PAM, HPAM, and xanthan gum work by improving water mobility in the reservoir, aiding oil extraction (Abidin et al., 2012). Among them, HPAM’s long-term stability increases its success in field application and improves oil recovery by up to 45% (Al-Sabahi et al., 2018; Braun et al., 2022; Choi et al., 2014). HPAM flooding as a tertiary EOR method functions to increase the injected water viscosity and reduce its mobility, which accelerates the oil extraction rate (Al-Sabahi et al., 2018; Liu et al., 2016). HPAM viscosity of 150–1000 cP showed improvement in oil recovery, when compared with water injection (Seright, 2010), but its usage has temperature and salinity restrictions. So, reservoir permeability, rock composition, and oil viscosity are important in the selection of the applied polymer in the field (Abidin et al., 2012).

Problems associated with PAM and HPAM

As a byproduct from oilfields using the polymer flooding method, produced water is contaminated with HPAM even after conventional treatment (Al-Sabahi et al., 2018). The potential toxicity of HPAM is neglected, but after natural or chemical degradation, it can be converted into acrylamide (CH$_2$=CHCONH$_2$, Fig. 3). Acrylamide is a toxic chemical that endangers the local ecosystem and human health (Bao et al., 2010; Elshafie et al., 2017; Liu et al., 2016). The International Agency for Research on Cancer has classified acrylamide as a group “group 2A” carcinogenic agent. It is known to be a carcinogenic, neurotoxic, mutagenic, and teratogenic monomer, and could also cause peripheral neuropathy (Al-Azkawi et al., 2013; Liu et al., 2016; Raju et al., 2015). Acrylamide monomer then discharges into terrestrial and aquatic ecosystems, inducing abnormalities in animal and plant cells (Yu et al., 2015). In addition, acrylamide can decompose non-thermally to ammonia, and thermally to carbon monoxide, carbon dioxide, and nitrogen oxides (Charoenpanich, 2013).

Another challenge is the difficulty of reinjecting HPAM-containing produced water because of its viscosity. Likewise, it is hard to separate the extracted oil from the produced water and consequential drop in oil yield (Al-Sabahi et al., 2018). Increasing the HPAM concentration in the produced water makes the separation and treatment process quite challenging. This is due to the anionic charge of HPAM, which increases the viscosity of oily produced water and decreases the rising tendency (Zhao et al., 2008). Subsequently, oil seeps into the sewage, which means that wastewater will exceed the local discharge limit (Bao et al., 2010). Besides, the contaminated produced water can leak into groundwater aquifers, which can endanger plants, animals, and human health (Sang et al., 2015). Thus, there is a need to treat produced water from acrylamide, but water treatment and disposal are costly (Bai et al., 2015; Warmt-Murray, 2015).

One of the main obstacles in the oil and gas industry is microbial-induced corrosion (MIC) and reservoir souring because of microbial growth in the pipeline system (Hasrizal & Yaakob, 2019). The injection of HPAM polymer promotes this effect since the oilfield biofilm consortium (mainly sulfate-reducing bacteria) use it as a secondary nutrient (Jia et al., 2018). Sulfate-reducing bacteria (SRB) work by utilizing sulfate as an electron acceptor to produce hydrogen sulfide (H$_2$S). This product reacts with the metal (Fe), corroding it by forming iron sulfide (FeS) and hydrogen (Fig. 4) (Chen et al., 2013; Makhlouf et al., 2018).

Oilfield produced water

Throughout the oil production phase, water produced along with oil (produced water) is the largest liquid waste (Ebrahimi et al., 2010; Li et al., 2010; Tellez et al., 2002). During the oil extraction process, the injected water flows through the organic phases of the reservoir because of rock permeability, developed pressure, and temperature changes. In addition, water is an
excellent extraction solvent, which means to dissolve a variety of chemical compounds used to enhance the recovery of oil. All these aspects make the composition of produced water complex and difficult to manage (Klemz et al., 2021). The volumes of produced water in different countries depend on the fields’ location and the methods of oil extraction. Table 1 highlights produced water volumes reported from oil and gas fields of different parts of the world (Al-Ghouti et al., 2019; Alsarayreh et al., 2022; Echchelh et al., 2018; Liu et al., 2021). Around 250 million barrels of produced water are generated globally, along with 80 million barrels per day of crude oil (Fakhru’l-Razi et al., 2009). It is the main source of oily water, as it reached 90% in the final stage of oil production in many Chinese oilfields (Lu et al., 2009). Thus, the water-to-oil ratio is around 3:1, and the ratio is increasing as the oil wells age (Al-Ghouti et al., 2019; Hirayama et al., 2002; Liu et al., 2021). Nowadays, there is a big effort worldwide to utilize and treat a significant quantity of produced water properly.

Different countries have many regulatory standards and restrictive regulations for oil and gas discharges. The United States Environmental Protection Agency (USEPA) allows a daily maximum limit of 42 mg/L, and a monthly limit of 29 mg/L of treated produced water discharge, while the People’s Republic of China (PRC) established a monthly limit of less than 10 mg/L, ~30 mg/L daily discharge in Australia, and a monthly limit of 30 mg/L in Canada. In the UK, following the Oil Pollution Prevention Control (OPPC) of the Energy and Climate Change Departments (DECC), the limit is set at 30 mg/L monthly. Moreover, the annually produced water discharge into the sea is 40 mg/L, according to (OSPAR Convention) the Convention of the Protection of the Marine Environment of the North-East Atlantic. Also, the National Environmental Council (CONAMA) in Brazil stated a monthly

![Fig. 4 Microbial induced corrosion (MIC) in the oil and gas pipeline system](Modified from Chen et al., 2013)

### Table 1 Volumes of oil and gas fields produced water in different countries (Al-Ghouti et al., 2019; Alsarayreh et al., 2022; Echchelh et al., 2018; Liu et al., 2021)

| Country    | Produced water (bbl/year) |
|------------|--------------------------|
| USA        | 21,000,000,000           |
| Australia  | 207,570,000              |
| China      | 45,917,000               |
| Colorado   | 92,274,300               |
| Iraq       | 105,853,190              |
| Oman       | 900,000*                 |
| Qatar      | 50,508,816               |
| Australia  | 3.02 × 10¹⁰              |
| New Mexico | 1.25 × 10⁹ μm³/day       |

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limit of 29 mg/L, and daily values of 42 mg/L (Fakhru’l-Razi et al., 2009; Klemz et al., 2021; Tellez et al., 2002). The European Union Water Framework Directive (WFD) in 2000 adopted a decision of “zero discharge” of the harmful produced water after 20 years (by 2020) to protect the marine ecosystem from sequel pollution. Currently, the majority of oil and gas companies in different countries are trying to fulfill this agreement (Klemz et al., 2021).

**Components of produced water**

A large quantity of produced water with distinctive characteristics is generated from oil fields, but its volume and characteristics are mainly dependent on the lifetime of a reservoir and the method of recovery (Ebrahimi et al., 2010; Li et al., 2021; Tellez et al., 2002). Produced water contains different combinations of organic and inorganic materials (Klemz et al., 2021) (Table 2). Its physical and chemical properties are mainly affected by the field’s geological location, depth and age of the well, and chemical composition of the reservoir (Al-Ghouti et al., 2019). Oilfield produced water contains soluble and insoluble petroleum fractions with different concentrations. The major hydrocarbons are alkanes, alkenes, alkynes, and aromatics. Produced water pH, salinity, temperature, and dissolved chemicals affect the amount of oil present in it. In addition, dissolved salt concentration as inorganic compounds exceed the seawater levels. Sodium chloride salt is the main salinity contributor (between 1000 and 40,000 mg/L), but phosphate is present as the lowest concentration, while water-soluble organic compounds involve different types of organic acids (such as formic, acetic, and propionic acids), where the pH, temperature, and pressure influence the solubility of such organic acids, but not salinity. Regarding the ionic composition, cations include calcium (Ca²⁺), barium (Ba²⁺), potassium (K⁺), sodium (Na⁺), iron (Fe²⁺), magnesium (Mg²⁺), and strontium (Sr²⁺), while anions include sulfate (SO₄²⁻), carbonate (CO₃²⁻), bicarbonate (HCO₃⁻), and chloride (Cl⁻). These ions can react to form CaCO₂, CaSO₄, and FeS₂, which cause corrosion in the pipeline.

**Table 2 General produced water parameters from different oil fields in the world (Klemz et al., 2021; Rahman et al, 2020; and some of our results from oilfields in Oman)**

| Oilfield produced water parameters | Typical value | Metal concentration | Value (mg/L) |
|-----------------------------------|--------------|---------------------|--------------|
| Density (kg/m³)                   | 1000–1140    | Calcium             | 13–30800     |
| Conductivity (μS/cm)              | <1000–600.000+ | Sodium             | 50–297000+   |
| Surface tension (mN/m)*           | 38–78        | Potassium           | 24–5500      |
| Turbidity (NTU)                   | >100         | Magnesium           | <8–10000+    |
| Total organic carbon (TOC) (mg/L) | 0–1500       | Iron                | <0.1–100     |
| Chemical oxygen demand (COD) (mg/L)| 177–1300    | Aluminum            | 310–410      |
| Total suspended solids (TSS) (mg/L)| 1–1000       | Boron               | <5.95        |
| pH                                | 4.0–10       | Barium              | <1.3–650     |
| Total Dissolved Solids (TDS) (mg/L)| 2650–138,500 | Nickel              | 0.06–0.2     |
| Total oil (IR, mg/L)              | ~0.0–565     | Cadmium             | 0–0.2        |
| Volatile (BTEX; mg/L)             | <0.20–55     | Chromium            | 0–10         |
| Total petroleum hydrocarbon (TPH) | <0.5–350     | Copper              | 0–50         |
| Oil and grease (mg/L)             | ~0–540       | Lithium             | 0.1–500      |
| Chloride (mg/L)                   | 80–300,000+  | Manganese           | <0.004–175   |
| Bicarbonate (mg/L)                | 71–3990      | Lead                | 0–8.8        |
| Sulfate (mg/L)                    | <2–3000      | Strontium           | 0.02–1000    |
| Ammoniacal nitrogen (mg/L)        | 10–300       | Titanium            | <0.01–0.7    |
| Sulfite (mg/L)                    | 0–50         | Zinc                | 0–55         |
| Phenols (mg/L)                    | 0–23         | Arsenic             | 0–0.3        |
| Volatile fatty acids (VFAs) (mg/L)| ~0–6000      | Mercury             | 0–10         |

*Depending on the type of surfactants used for EOR operations*
Another type of heavy metals could also be present, but their concentration depends on the wells’ age and geology. Heavy metals in produced water are usually in trace quantities, such as nickel (Ni), chromium (Cr), lead (Pb), copper (Cu), cadmium (Cd), zinc (Zn), mercury (Hg), and silver (Ag). Besides that, the inorganic salts and organic compounds in produced water create an enriched environment for microbial growth, such as algae, bacteria, and fungi. The bacteria are usually saprophytic bacteria and sulfate-reducing bacteria, which enhance the pipelines’ corrosion issue (Al-Ghouti et al., 2019; Fakhru’l-Razi et al., 2009; Li et al., 2021; Tellez et al., 2002).

Characteristics of produced water

Produced water is a mixture of organic and inorganic chemicals varying from simple to complex nature, making the produced water different from other type of liquid wastes. Some contain high polymer content, such as PAM and HPAM, which ranges between 200 and 500 mg/L. The presence of a huge quantity of polymers raises the water viscosity. In addition, the oil in the produced water is with a smaller diameter droplet (approximately 3–5 μm) than the conventional size (about 35 μm) that makes it difficult to remove. The residual oil-in-water forms a stable emulsion, which increases the complexity of the produced water. As well, the existence of different organic compounds (such as benzene, toluene, and xylene) raises the biological toxicity of produced water. Their concentration is significantly elevated, exceeding the allowed standards. For example, benzene concentration in produced water is >35,000 μg/L, which is considered 7000 times higher than the limit for drinking water in the USA (Al-Ghouti et al., 2019; Li et al., 2021).

Environmental risk of produced water

Recently, the influence of untreated produced water discharge has become a significant environmental issue (Lu et al., 2009; Tellez et al., 2002). In the past, the produced water from oil fields was disposed of in large evaporation ponds. This leads to the emission of volatile organic compounds (VOCs) in the atmosphere, which contains ozone precursor compounds. Increasing ozone to levels above the air quality standards is associated with many respiratory problems in humans (Wang et al., 2012).

Various organic and inorganic components of produced water, including salts, are dangerous for surface and underground water (tend to precipitate and accumulate in aquatic sediments), plants, atmosphere, soil, and human health. Irrigation of the soil with produced water with high salts and oil content affects the soil negatively. It blocks the soil pores and weakens the soil ecosystem. In addition, the presence of hydrocarbons in the produced water discharged in the streams has a great effect on the species diversity of the marine communities (Fakhru’l-Razi et al., 2009; Li et al., 2021; Tellez et al., 2002). Researchers have reported that some petroleum hydrocarbons are carcinogenic, and can cause embryo distortion, organ (heart, kidney, and brain) damage, and gene mutations in organisms (Lahiri et al., 2021). In addition, produced water discharges in surface water tend to form an oil layer at the top that reduces the oxygen transmission into the water ecosystem (Ossai et al., 2020).

Management of produced water

To mitigate and control pollution, it requires proper produced water management techniques. Several factors determine the selection of produced water management options, such as the county laws and regulations, cost, technical potentials, and geographical situation with the available infrastructure. The first step in that direction is to minimize its production from the oil fields by different technologies, which showed not-so convincing results.Produced water reuse in oil and gas operation is a second possibility, after the suitable accurate treatment, such as utilizing to prepare a polymer solution or in other beneficial uses such as livestock consumption, irrigation, fire control, industrial power generation, and drinking water. The most common reuse of produced water is re-injection into oil reservoirs for EOR. The final management option is disposal, but after extensive treatment because of the complex nature of produced water components. Produced water discharged should meet the national standards which differ between different countries and even between regions of the same country (Al-Ghouti et al., 2019; Arthur et al., 2005; Fakhru’l-Razi et al., 2009; Li et al., 2021). The most effective handling option for the produced water is by
applying a suitable method of treatment to convert it to a harmless product. There are many treatment techniques but all focus on de-oiling, disinfection, desalination, and suspended particle removal (Ebrahimi et al., 2010; Echchelh et al., 2018; Fakhru’l-Razi et al., 2009). The degree of treatment depends mainly on the produced water application afterward. Minimal treatment is required to use the treated water in industrial applications, but a high level of treatment is needed to use the treated water for drinking or agriculture (Al-Ghouti et al., 2019; Echchelh et al., 2018).

### Produced water treatment methods

The produced water as a byproduct after EOR operations requires special-effective treatment to reduce the viscosity, the content of free oil, and break the emulsion without any environmental issues (Li et al., 2021; Nicolaisen, 2003). Such treatment is a challenging task since it is produced in large amounts (~90% of the total extracted fluids) with complex compositions and different contaminants (Al-Ghouti et al., 2019; Li et al., 2021). Thus, the easily available handling options are either disposal after following environmental regulations, or re-injection after skillful planning to avoid damage (Ebrahimi et al., 2010; Lu et al., 2009).

For produced water treatment, different types of physical, chemical, and biological treatment methods are available. In the case of physical methods, the pollutants are not destroyed, but are destroyed with chemical and biological methods. Physical treatments include the adsorption on activated carbon (achieving better quality, low contamination water), sand filtration, gravity separation, and evaporation. In the case of chemical treatment, it involves chemical precipitation (through coagulation and flocculation), chemical oxidation, demulsifier (oil–water separation), electrochemical, and photocatalytic processes, while biological treatment (via aerobic and anaerobic microorganisms) is more effective that supports the economic feasibility with lower cost. The biological treatment includes using activated sludge (the usual biological method), sequencing batch reactors (SBRs), and biological aerated filters (BAF). This was accomplished after understanding the mechanisms of the used microbes (Adewumi et al., 1992; Al-Ghouti et al., 2019; Fakhru’l-Razi et al., 2009; Li et al., 2010, 2021; Lu et al., 2009; Wang et al., 2012). In the past 20 years, many researchers have performed projects aiming to study the oilfield-produced water and find alternative treatment methods. These researches involved the produced water characterization, compositional analysis, environmental impacts, treatment technologies, and cost. Klemz et al. (2021) reported that around 2000 publications were published between 1999 and 2019, related to this field.

### The degradation of PAM and HPAM

Polymers are susceptible to various degradation processes, depending on polymers’ nature and conditions. Most degradation processes are irreversible reactions that change the polymers’ physical and chemical characteristics (Caulfield et al., 2002). HPAM persists for a long time in the water with a slow degradation rate contaminating ground and surface water affecting the environment, animal, plant, and leading to serious public health problems if not treated efficiently (Al-Moqbali et al., 2018; Al-Sabahi et al., 2018). The degraded hydrophilic PAM increases the mobility of environmental molecules because of its carboxylic group content, after hydrolysis by many environmental factors (Xiong et al., 2018). In nature, HPAM and PAM degradation may occur thermally, mechanically, chemically, or photolytically. Thermal degradation depends on polymer molecular weight, impurities, and the mode of synthesis, while mechanical degradation occurs because of polymer plumbing with a high flow rate and high shear, elongational flow, or elevated agitation, depending on the application of the polymer. In the EOR process, the polymer is injected with high viscosity through the porous reservoir rock, which results in the polymer’s exposure to high mechanical stress. This subsequently affects the polymer’s properties and causes a reduction in its viscosity and performance. The chemical degradation of the polymer chain is related to the amide group reactivity, which results in the release of free hydroxyl radicals. Similarly, oxygen-free radicals are released photolytically by sunlight, which is extremely unstable reactive species. In both cases, oxygen radicals will have negative effects on the environment and public health. Al-Sabahi et al. (2018) reported photocatalytic degradation of ~74% of HPAM in produced water using supported zinc oxide nanorods. The photo-degradation reaction is a photon’s energy absorption mechanism, which results in backbone cleavage of the polymer and free radical formation. (Caulfield et al., 2002; Reddy et al., 1994; Xiong et al., 2018). Khan et al. (2019)
reported innovative combination of ultrasonic waves and chemical agents for HPAM degradation and deplugging of the core sample. It has been reported that acrylamide is formed upon polyacrylamide exposure to UV radiation (Al-Moqbali et al., 2018; Smith et al., 1997). Thus, the byproducts of these reactions are important since they can raise the toxicity of the degradation processes, especially radical formation. Feng et al. (2022) reported ~97.63% HPAM removal (as measured by viscosity reduction), by an electrochemical method using a novel La-In co-doped PbO2 electrode. Although various kinds of chemical agents are reported handle PAM/HPAM degradation, it still needs a huge financial investment, and also suffers from environmental and other associated issues such as corrosion, which motivates researchers to look for other efficient and environmental friendly techniques (Khan et al., 2019).

Biodegradation of PAM and HPAM

Although environments like oil fields, produced water, and soils surrounding oil fields are considered as quite harsh, microbes beneficial for biodegradation of such pollutants are reported (Abed et al., 2021; Al-Sayegh et al., 2021; Wei et al., 2022; Zhang et al., 2021). Among different types of toxic and recalcitrant chemicals present in produced water, polymers such as PAM and HPAM are somewhat difficult for biodegradation, as both PAM and HPAM cannot pass through most bacterial membranes due to their higher molecular weight (Berdugo-Clavijo et al., 2019; Joshi & Abed, 2017; Ma et al., 2021). PAM and HPAM biodegradation occur when microbes utilize the amide group of the polymer as a nitrogen source and/or the polymer carbon backbone as a carbon source. Microbial biodegradation is favored since it yields higher removal efficiency without additional environmental pollution (Liu et al., 2016; Nyysölä & Ahlgren, 2019). Bao et al. (2010) reported that some microorganisms from produced water could utilize HPAM either as carbon or nitrogen sources. Caulfield et al. (2002) explained that bacteria can grow in the presence of PAM, by utilizing it as a nitrogen source by hydrolyzing the amide to acid and NH3 by extracellular amidase. The resulting byproducts were of a lower molecular weight than the polymer, with several COO− groups and without acrylamide formation (Fig. 5). Many types of amidase differ in their activity depending on the bacterial genera it is isolated.

To degrade the main carbon backbone of PAM and to utilize it as a carbon source for bacterial growth, other enzymes are required. Several reports showed potential role of mixed microbial biofilms in the treatment of PAM or HPAM containing wastewater (Li et al., 2016; Song et al., 2019a, b; Zhang et al., 2020, 2021). Zhang et al. (2019) reported efficient degradation of PAM and acrylamide in a multistage contact reactor, using anamox and ammonia-oxidizing bacteria enriched biosilms. Song et al. (2021) reported ~85% HPAM removal using mixed microbial-ozonic-anaerobic–aerobic biofilm multistage bioreactor treatment process.

Two polyacrylamide-degrading bacterial strains were isolated from soil, Bacillus sphaericus and Acinetobacter sp., utilizing 16–19% polyacrylamide after 14 days of cultivation (Matsuoka et al., 2002). Al-Moqbali et al. (2018), on the other hand, isolated Bacillus sp. and Chelatococcus sp., from polymer flooded produced water, which degraded HPAM efficiently after 9 days of treatment. Azomonas macrocytogenes and Enterobacter agglomerans, PAM-degrading bacteria, were isolated from the soil, which consumed PAM as the sole source of carbon and nitrogen (Nakamiya & Kinoshita, 1995). Pseudomonas putida showed 45% PAM degradation efficiency in 7 days as its sole nutrient source (Yu et al., 2015). Several reports highlight bacterial degradation of PAM and HPAM polymers, using different isolates from diverse sources. However, it varies in its removal efficiency under anaerobic or aerobic conditions (Table 3).
| Microorganism                                      | Source of isolation                                                                 | Conditions                                      | HPAM/PAM removal % | Reference                   |
|--------------------------------------------------|-------------------------------------------------------------------------------------|------------------------------------------------|--------------------|-----------------------------|
| Mixed microbes                                   | Wastewater from “alkaline-surfactant-polymer” flooding water treatment station       | Aerobic–anoxic–anaerobic/20–33 °C              | 11–16.79%          | Wei et al. (2022)           |
| Mixed microbial mats                             | Constructed wetland treating oilfield produced water                                 | Aerobic–anoxic /30 °C                          | 7–34%              | Abed et al. (2021)          |
| Mixed microbes/microalgae                       | Biological contact oxidation reactor used for the treatment of PAM containing wastewater |                                               |                    |                             |
| Mixed microbes                                   | Simulated synthetic wastewater and activated wastewater sludge                       | Ozone-an aerobic–anaerobic/25–35 °C            | 85.06%             | Song et al. (2021)          |
| *Klebsiella* sp. PCX-biochar composite and other mixed microbes | The laboratory of Southwest Petroleum University (China)                           | Aerobic/38 °C                                  | 69.1%              | Ma et al. (2021)            |
| Mixed microbes                                   | Domestic sewage treatment plant                                                     | Aerobic–anaerobic/30 °C                        | 43.6%              | Zhang et al. (2020)         |
| *Acinetobacter* sp. and *Pseudomonas* sp.       | Wastewater samples from coking plant and shale gas field                            | Aerobic/35 °C                                  | 45.82%             | Dong et al. (2020)          |
| Mixed microbes                                   | Sludge from wastewater treatment plant                                              | Aerobic/25 °C                                  | ~75% (as viscosity reduction) | Zhang et al. (2019)         |
| Mixed microbes                                   | Oilfield produced water and activated wastewater sludge                             | Aerobic/50 °C                                  | Partial            | Berdugo-Clavijo et al. (2019) |
| Mixed microbes                                   | Activated sludge from a wastewater treatment plant                                  | Anaerobic/33 °C                                | 40.5%              | Zhao et al. (2019a,b)       |
| Mixed microbes                                   | HPAM containing samples from Wastewater treatment stations                           | Aerobic and anoxic/35 °C                       | 38–65%             | Zhao et al. (2019a,b)       |
| Mixed microbes                                   | Oilfield wastewater treatment station and aerobic activated sludge from a domestic wastewater treatment plant | Aerobic-Ozone–anaerobic/25–35 °C              | 90.79%             | Song et al. (2019a)         |
| *Bacillus megaterium* SZK-5                      | Oilfield soil contaminated with produced water                                      | Aerobic/24 °C                                  | 55.93%             | Song et al. (2019b)         |
| Mixed microbes                                   | Production water samples                                                           | Anaerobic/36–37 °C                              | Partial/not specified | Hu et al. (2018)            |
| Mixed microbes                                   | Wastewater from “alkaline-surfactant-polymer” flooded oilfield                     | Aerobic–anaerobic–anaerobic/30 °C             | 7%                 | Liu et al. (2018)           |
Bacterial degradation metabolic pathway

The complete metabolic pathway of PAM and HPAM degradation is still not clear. The enzyme amidase is known to initiate the biodegradation of PAM and HPAM under aerobic or anaerobic conditions, followed by different enzymes involved yielding partial or complete degradation (Joshi & Abed, 2017). Extracellular amidases work on PAM amide group hydrolyzation, as reported by *Pseudomonas putida* after incubation at 39 °C for 7 days (Yu et al., 2015). The Fourier transform infrared (FTIR) spectrograms of *Bacillus cereus* and *B. flexus* cultures after PAM degradation showed the removal of the amide group from the polymer (Wen et al., 2010). While catalyzing PAM’s carbon backbone via monooxygenases oxidation into smaller molecules, such as acrylamide and/or acrylic acid, which could be further degraded (Joshi & Abed, 2017). In general, PAM degradation by bacteria cause changes in the polymer structure (Caulfield et al., 2002). The degradation pathway of PAM and HPAM to a smaller molecular weight product like acrylamide has been proposed (Fig. 6) (Bao et al., 2010; Joshi & Abed, 2017). The amidase action on the amide group of PAM and HPAM results in the release of NH₃ and COOH addition. Subsequent monooxygenase oxidation leads to cleaving the main carbon backbone of PAM and HPAM. After many cycles, smaller organic molecules are formed including acrylamide (Eubeler et al., 2009, 2010).

### Table 3 (continued)

| Microorganism | Source of isolation | Conditions | HPAM/PAM removal % | Reference |
|---------------|---------------------|------------|---------------------|-----------|
| *Bacillus sp.* | Polymer flooded produced water | Aerobic/40 °C | 59.9% 58.4% | Al-Moqbali et al. (2018) |
| *Chelatococcus sp.* | | | | |
| *Paenibacillus sp.* | Activated sludge | Anaerobic/36 °C | 68.27% | Li (2017) |
| Mixed microbes | Water injection pipeline from oilfield | Not specified | Partial/not specified | Li et al. (2016) |
| *Pseudomonas aeruginosa* | HPAM-containing wastewater | Aerobic/35 °C | 41.6% | Liu et al. (2016) |
| *Bacillus cereus, Bacillus sp., Rhodococcus sp.* | Polymer flooded produced water | Anaerobic and aerobic/37 °C | Partial/not specified | Zhao et al. (2016) |
| *Bacillus cereus* and *Rhodococcus sp.* | Polymer flooded produced water | Anaerobic and aerobic | 75.8% | Sang et al. (2015) |
| *Pseudomonas putida* | Dewatered sludge | Aerobic/39 °C | 45% | Yu et al. (2015) |
| Mixed bacteria | HPAM-containing sewage | Aerobic/35 °C | 62.1% | Liu et al. (2013) |
| *Bacillus cereus* and *Bacillus sp.* | Polymer flooded produced water | Aerobic/40 °C | 42.1% | Bao et al. (2010) |
| *Bacillus cereus* and *Bacillus flexus* | Activated sludge and oil-contaminated soil | Aerobic/30 °C | 70% | Wen et al. (2010) |
| *Clostridium bifermentans H1* | Curing pot in the HPAM distribution station | Anaerobic | 30.8% | Ma et al. (2008) |
| *Anaerofilum pentosovorans A9* | Oilfield | Anaerobic | 61.2% | Li and Fang (2007) |
| *Enterobacter sp.* | Curing pot in the HPAM distribution station | Anaerobic/38 °C | 63.17% | Wei et al. (2007) |
| *Bacillus sphaericus* | Soil | Aerobic/30 °C | 16% 19% | Matsuoka et al. (2002) |
| *Acinetobacter sp.* | | | | |
| *Enterobacter agglomerans Azomonas macrocytogenes* | Soil | Aerobic/30 °C | 20% 15% | Nakamiya and Kinoshita (1995) |

NA not available
Characterization of biodegraded PAM and HPAM

After the biodegradation process of PAM and HPAM, it is necessary to measure its concentration and determine its fate, via various methods, depending on its sensitivity and detection limits. The initial method used was a photometric titration procedure which involved the reaction of crystal violet (CV) dye with PAM in the sample and was followed by an absorbance reading at 590 nm (Masadome, 2003). The C18 and ion-exchange columns were developed in high-performance liquid chromatography (HPLC) to measure acrylamide in polymers (Vers & Leonard, 1999). This method works successfully for all types of polymers and matrices with a parts-per-billion (ppb) ranges of detection. Yu et al. (2015) reported change in PAM molecular weight and functional groups after bacterial biodegradation by gel transmission chromatography (GPC) and Fourier transform infrared (FTIR) methods. The concentration of HPAM can also be measured by the starch-cadmium iodide method (Yongrui et al., 2015). After the measurement of HPAM at 585 nm, the difference between control and biodegraded sample readings are used for HPAM removal ratio calculation. Using this method, the quantity of PAM is presented as a concentration by spectrophotometry after a series of reactions with amide groups in PAM (Lu & Wu, 2002). In addition to HPAM concentration measurements, the viscosity pre- and post-biodegradation can be determined by a rheometer to acquire the viscosity reduction rate (VRR) which is the difference between control and biodegraded samples’ shear viscosity (Zhao et al., 2016). HPAM removal efficiency can be determined based on total organic carbon (TOC) that helps in evaluating PAM and HPAM biodegradation using a TOC analyzer (Sang et al., 2015).
Nuclear magnetic resonance (NMR) studies confirmed PAM degradation by the action of microbes (Wampler & Ensign, 2005). Other methods include NH$_3$ selective electrode (Zi-jun & Lang, 2008), a sodium hypochlorite oxidation method (Neng et al., 2005), film drying method (You et al., 2007), quantitative turbidimetry (Qing-Xiang et al., 2009), biochemical methane potential (BMP) test (Dai et al., 2014), and size exclusion chromatography (SEC) (Asano et al., 1982). Furthermore, liquid chromatography-mass spectrometry (LC–MS) can verify the biodegradation of HPAM to acrylamide and measure its concentration in the samples (Al-Moqbali et al., 2018).

**Biopolymers as an alternative for PAM and HPAM**

One of the important ways to recover more oil from exhausted oil reservoirs after water flooding is polymer flooding (Pu et al., 2018). PAM and HPAM are the most extensively used polymers in enhancing oil recovery due to their customized properties and wide availability. Although the usage of PAM and HPAM as an EOR method resulted in increasing the production of oil considerably to overcome the current energy crisis, it is adverse environmental and health issues suggested searching for other eco-friendly alternative polymers. Extracellular polysaccharides formed by different bacterial species give light to work as environmental-friendly, chemical stability, and biodegradable polymers can be used in EOR applications. Xanthan gum produced by Xanthomonas campestris (Abidin et al., 2012), pullulan produced by Aureobasidium pullulans (Al-Araimi et al., 2021; Elshafie et al., 2017), and schizophyllan produced by Schizophyllum commune (Joshi et al., 2016), are well-known polysaccharide biopolymers showed an outstanding performance in tertiary oil recovery (Fig. 7). Such microbial metabolites helps in the oil recovery process by reducing mobility and modifying permeability during water flooding (Shabani Afrapoli et al., 2012). Another reported advantage for the biopolymers are stability at high salinity and temperature, unlike chemical polymers (Gao, 2016; Quadri et al., 2015). This is due to its helical structure (triple or double), rigidity, and charge-free chains which increase its stability and thickening potential in harsh elevated temperature and saline oil reservoirs (Pu et al., 2018). However, its drawback is related to its poor filterability, which can lead to

**Fig. 7**  The chemical structure of polysaccharide biopolymers (A pullulan, B schizophyllan, C Xanthan gum) (Ferreira et al., 2015; Lochhead, 2017; Sutivisedsak et al., 2013)
cell debris plugging during the injection which requires a pretreatment to eliminate it (Kulawardana et al. 2012; Rivenq et al., 1992). Additionally, biopolymer’s properties have to be tested before using it in polymer flooding for EOR application. The properties include molecular and chemical stability, temperature and salt tolerance, thickening capability, rheology, and adsorption (Pu et al., 2018).

**Conclusion and future prospects**

Renewed interests and improved economy of the non-fossil-based energy sources, awareness about a role in environmental pollution, international policies governing the supply, and demand of crude oil (and other types of fossil-based energy sources), are some challenges faced by petroleum industries. Apart from those current challenges, the petroleum industry was already facing two major issues: the requirement of water for enhancing/increasing oil recovery, and the treatment and disposal of such a huge quantity of oilfield produced water. To make matters worse, the majority of oil-producing countries are arid regions, having almost little or no rain or freshwater resources, making the water even a precious commodity, as it is needed for day-to-day activities. Produced water is one of the most challenging types of wastewater for treatment that too produced in huge quantities. It contains heavy metals, high salt concentrations, and toxic compounds (PAHs, traces of crude oil, chemicals such as HPAM, surfactants, etc.), normally occurring radioactive materials, and more. The treatment becomes even more difficult when specific EOR chemicals like HPAM are used, which leads to stable emulsions, difficult oil–water separation (for further treatment), the possibility of generation of neurotoxins (such as acrylamide). Thus, degradation and/or removal of HPAM/acrylamide are some of the sought after issues in petroleum industries. Currently, it is performed by different chemicals and thermal technologies, such as electrochemical precipitation, use of oxidizing chemicals, photocatalytic degradation in presence of nanoparticles, etc., but some of these techniques are either immature, expensive, or not environmental friendly. One of the promising environmentally friendly techniques are biobased degradation. Several lab scale and field scale reports showed quite promising results for PAM/HPAM biodegradation, where it also showed possible removal of highly toxic acrylamide monomer. Although several laboratories based and pilot-scale biodegradation studies have been reported, it still needs additional-extensive studies to better understand the exact mechanisms and optimized-economic scaled-up processes for field-scale applications, and to make it economically attractive option. Advances in “omics” and synthetic biology-based technologies could play an important role in the future applications of biodegradation of HPAM from produced water, and a safe disposal or other applications using such treated water, to support UNSDG 6.

**Acknowledgements** SK and SJ would like to kindly acknowledge research fellowship and research funding, respectively, by The Research Council (TRC) Oman, and Sultan Qaboos University (RC/RG-DVC/OGRC/18/01). Authors would also like to acknowledge kind support provided by local oil companies (for providing produced water samples, polymers, and analysis data), and Central Analytical and Applied Research Unit, Sultan Qaboos University, Oman.

**Declarations**

**Conflict of interest** The authors declare no competing interests.

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