Green Nanoparticles for Sustainable Oil Recovery

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ABSTRACT
The current global trend for application of nanotechnology in oil and gas industries has led to the development of novel green nanoparticles (GNPs) which could reduce the overall environmental impact and improve oil and gas production. The use of environmentally benign green products such as plants and microorganism has gain attention among the researchers due to their improved properties and sustainability. The present chapter provides an essential knowledge and understanding to the application of green nanoparticles in EOR with special emphasis on its application in microemulsion.
Keywords— Green nanoparticle, Enhanced oil recovery, Microemulsion, Winsor classification, Interfacial tension

I. INTRODUCTION

Petroleum resources continue to be regarded as the primary source of energy. To meet rising energy demand, improved oil extraction from untapped reservoirs is required, and petroleum is classified as a conventional energy resource. As a result, once these energy sources have been depleted and exploited, they cannot be reused, emphasising the significance of sustainability in energy production. The concept of sustainability is the development and use of products based on need and demand without jeopardising the needs of future generations. Because the environment is a limited resource, it is critical to use it wisely. Therefore, it is necessary to maximize oil recovery. Around 30 % of original oil place can be recovered by using the conventional resources. In order to meet high energy demands, advanced methods have to be developed for enhancing the recoverable volumes from the existing reservoirs [1]. Therefore, for upgrading the recovery, Enhanced oil recovery (EOR) techniques are needed. EOR basically aims at increasing the recovery. EOR techniques involves the infusion of additives, chemicals, gases or thermal energy into a reservoir. The injection of the above mentioned helps in upgrading the existing reservoir properties for displacing the oil to producer well. EOR techniques are divided into Thermal, Chemical, Miscible and Microbial EOR. Thermal Enhanced oil Recovery (TEOR) uses thermal energy to escalate the reservoir temperature. The increase in temperature causes reduction in oil viscosity. When talking about the global energy demand, TEOR is taken as a advanced EOR Strategy which delivers quite a good amount of oil in overall global oil outlook [2].TEOR processes are more convenient to use in sandstone reservoirs and also for high density, thick and viscous oils with API gravity less than 20 [3]. Chemical Enhanced Oil Recovery (CEOR) believed as an inviting EOR approach which stimulates the residual oil by diminishing the capillary forces. Depletion of interfacial tension (IFT) for an increasing capillary number is defined as a systemized and a fiscally viable strategy in EOR. In Miscible Enhanced oil recovery method, low molecular weight hydrocarbons or gasses like carbon dioxide or nitrogen are infused into the reservoir. The infused gas gets miscible with the crude oil in deep reservoirs at moderate temperature conditions which enhances the production [4]. The last but not the least, microbial enhanced oil recovery (MEOR) can be considered as a significant one in considering the sustainability. The significant mechanisms for enhancing the recovery includes Interfacial tension (IFT) reduction, reduction of viscosity, oil swelling as well as alteration of wetting characteristics. But the selection of EOR methods depends on the reservoir characteristics, compatibility of chemicals and the economic feasibility for the particular technique to be implemented [5]. Over the last 60 years, there has been significant progress in chemical flooding, raising the possibility of it becoming the most important EOR method [6]. Many countries, including the United States, China, Germany, France, Austria, and Canada, have reported success with chemical EOR. However, due to the high cost of chemicals, chemical flooding is an expensive recovery method.

As the industry is moving more towards sustainability, it is necessary to incorporate more environmentally friendly solutions for the unsolved problems in the industry. The incorporation of nanoparticles (NPs), ionic liquids, biosurfactants, bio-nanomaterials, deep eutectic solvents (DES) etc. in EOR helps in increasing the microscopic and macroscopic sweep efficiency economically [7]. In case of sustainable biomaterials, GNPs have created more interest among the researchers for its further development and applications.

Due to its unique properties such as wettability alteration, improved mobility of trapped oil, enhancing sand consolidation, and reduction of interfacial tension (IFT). Nanotechnology is profoundly appealing but challenging in the petroleum industry to enhance oil recovery [4], [8]. The morphology, size, chemical, mechanical, and physical properties of NPs are used to classify them. As a result, each NP has distinct physicochemical and mechanical properties [9]. It can be free or bound together depending on attractive and repulsive forces. NPs have a higher surface-area-to-volume ratio and are highly reactive to chemicals [10]. These characteristic features make the NPs more applicable in petroleum industry especially in EOR. Among the different mechanism of oil recovery such as IFT reduction, wettability alteration, microemulsion etc. The least discovered area is about microemulsions. The formation of in-situ microemulsions and injection of ex-situ microemulsions formed by surfactants increase the residual oil recovery and overall efficiency. The ex-situ microemulsion formation and stability are two important parameters to be considered for its characterization. The application of green NPs to increase stability and morphology of microemulsion could be a sustainable path to be explored in future for increased oil recovery.

II. NANOPARTICLES

NPs can be referred as particles having size ranging from 1 to 100 nanometers, which are usually made of carbon, metal, metal oxides or organic matters. The NPs possess unique physical, chemical and biological properties due to their distinct properties such as relatively larger surface area to the volume, increased reactivity or stability in a chemical process, enhanced mechanical strength, etc. [11, 12]. These properties causes significant changes in their physical properties when compared to those observed in bulk materials, which has led to their use in a variety of applications. The NPs can be zero dimensional, one dimensional, two dimensional, and three dimensional based on its length, breadth and height. Zero dimensional where length, breadth and height is fixed at a single point and electrons movement is different in all three directions such as Quantum dots. In the case of one dimensional it only possess one parameter and electrons can only move in the X-direction such as graphene. In two dimensional NPs it possess length and breadth and they can only move on the X–Y plane, and in case of three dimensions it possess all the parameters such as length, breadth and height with movement in X, Y, and Z directions, for example gold NPs. NP research is currently an area of intense scientific interest [12].

A. **Classification of NPs**

The NPs utilized for various industrial applications are generally categorized as organic, inorganic and carbon based NPs. The organic NPs composed of synthetic organic molecules which are non-toxic, biodegradable and sensitive to electromagnetic and thermal radiations. Some of the examples could be micelles, dendrimers, liposomes etc. [13]. The next category could be inorganic NPs composed of metal and metal oxides such as iron, aluminum, silver etc. Inorganic NPs have exceptional properties such as pore size, high surface area to volume ratio, spherical and cylindrical shape, crystalline and amorphous structures, surface charge and surface charge density, sensitivity to environmental factors such as air, moisture, heat and sunlight etc. [14]. The carbon based NP are made solely of carbon such as graphene, carbon nano tubes, carbon black, carbon nanofibers etc. [15]. Based on physical and chemical characteristics, some of the well-known classes of NPs for EOR application can be summarized as following:

a) Carbon-based NPs

Fullerenes and carbon nanotubes (CNTs) are the two most common types of carbon-based NPs. Fullerenes contain nanomaterials made of globular hollow cages, such as allotropic carbon forms with superior properties such as higher strength, electrical conductivity, structure, electron affinity, and versatility. These materials contain pentagonal and hexagonal carbon units that have been sp2 hybridized. CNTs have a tubular, elongated shape and a diameter of 1-2 nm [16]. These structurally resemble graphite sheets rolling on top of one another. Carbon-based NPs have numerous application in oil and gas industry and particularly in EOR. The wettability of the sandstone substrates can be altered using different concentration of sulfonated nonporous graphene. It was observed that with increasing NP concentration the water wetness of the surface improves due to the enhanced adsorption of nanomaterials on the surface which in turn increases the disjoining pressure [17]. Another application of different concentration of GO nanosheets on carbonate slices showed similar results of wettability alteration. Kanj et al. investigated the flooding performance in carbonate reservoir by modified carbon NPs and observed an increase in oil recovery factor [18].

b) Metal NPs

Metallic nanoparticles are primarily composed of metal precursors such as silver, gold, copper, palladium, and platinum. Metallic NPs have important properties such as a high surface-area-to-volume ratio, high surface energies, a specific electronic structure provided by their transition between molecular and metallic states, plasmon excitation, and quantum confinement [19]. In the case of EOR, nanofluids containing metallic NPs such as Al_2O_3 and TiO_2 increased 24% more than non-metal oxides, and the contact angle decreased from 54° to 21° (changing the wettability to more water-wet) [20].

c) Polymeric NPs

Polymeric NPs are particles having active compounds entrapped within or surface-adsorbed onto the polymeric core with size ranging from 1 to 1000nm. Due to their increased solubility and stability, increased stabilization of foams and emulsions, and ease of transport through porous media. Polymeric NPs have attracted significant interest as additives and they have recently been investigated for EOR applications as well [21]. They are simple to functionalize and have a wide range of uses in the literature. The morphological structure helps to distinguish between nanospheres and nanocapsules. Polymeric NPs can be nanocapsules or nanospheres, which are distinguished by the morphological structure. Nanocapsules are matrix particles whose overall mass is generally solid and the other molecules are adsorbed at the outer boundary of the spherical surface, while nanospheres are solid mass is encapsulated within the particle completely [22].

III. GREEN NP AS A SUSTAINABLE RESOURCE FOR EOR

In most of the cases, NPs for EOR applications are dispersed in fluids such as oil, deionized water, brine, or gas to form nanofluids. The effects of these nanomaterials on EOR are examined using the mechanisms and phenomena such as wettability alteration by NP adsorption and disjoining pressure, IFT reduction at oil-water interface, viscosity adjustment by means of polymer, surfactants etc., pore channel plugging and prevention of asphaltene precipitation. Depending on the application, surface ligands, stabilizers, and polymer/surfactant used, nanomaterials can have hydrophilic, hydrophobic, or double-faced (Janus) properties [23]. SiO2 and TiO2 NPs have been shown to improve oil recovery in sandstones by altering wettability and decreasing IFT [24]. NP deposition could clog water channels and increase sweep efficiency during flooding. Similarly, SiO2, Al2O3, and TiO2 NPs have been shown to improve oil recovery from carbonates through wettability modification, IFT reduction and viscosity modification [25]. The use of nanofluids could improve the performance of brine injection even further.

The high interfacial activity of NPs and nanofluids results in increase production from oil reservoirs, as NPs have high surface area to volume ratios which adsorb at fluid-fluid interfaces and reduce IFT which is further enhanced in the presence of surfactants as shown in Figure 1. The enhanced emulsification ability is determined by the properties and quantities of NPs. The size of emulsified oil droplets decreases as the NP to oil mass ratio increases [26]. NPs can change the wettability of solid surfaces due to their self-structuring behavior which also raises the structural disjoining pressure in the confined three-phase contact region, particularly near the wedge's tip as shown in Figure 1. It was observed in some outcrops and reservoir rock that NPs can move the oil/water interface forward, alter wettability, and thus detach organic constituents of crude oil from surfaces as structural disjoining pressure increases. The organic constituents of crude oil were further observed to be removed further to be about 70% after NP-stabilized emulsion flooding [27]. When compared to ME alone, the NP stabilized Microemulsion (ME) formulation has improved residual oil recovery to 19% with increased oil displacement efficiency. The NP-stabilized ME showed stable adsorption at the oil/water interface, synergistic stabilization effects between the NP and surfactant, and increased brine phase viscosity. While NP-stabilized MEs frequently outperform MEs alone, little is known about their displacement mechanism in porous media and the types of fluid-fluid and fluid-rock interactions that occur. The generation of NPs are generally done by three routes i.e. physical, chemical and biological. However, the production of NPs via physical and chemical processes causes toxicity and environmental challenges. The physical process is time-consuming, takes up a lot of space, and produces a lot of heat, which raises the temperature in the area around the source material. The toxic nature of the solvents and chemicals could cause harm to already degrading environment [28]. Thus, the dire need for developing alternative method for greener development has led to the concept of green nanotechnology. Green nanotechnology in petroleum industry and EOR majorly uses plants, fungi or bacteria as its source for NP synthesis which are eco-friendly. The utilization of plants for NPs production has drawn attention in recent years because its rapid, eco-friendly, non-pathogenic, and economic protocol provides a single-step technique for the biosynthetic process. The major advantages of using plant extracts for NP synthesis is that they are easily available, safe, nontoxic in most cases, have a broad variety of metabolites that can aid in the reduction of silver ions, and are quicker in synthesis than microbes [29]. In our research, a similar technique was used to synthesize Si-NPs from bamboo leaves. Bamboo Leaf Ash was produced by calcining the leaves in a muffle furnace. The plant ash was mixed with NaOH solution and heated. The obtained intermediate product was filtered and HCL was added to decrease the pH and left stagnant. The obtained silica gel was later heated to get silica NPs.

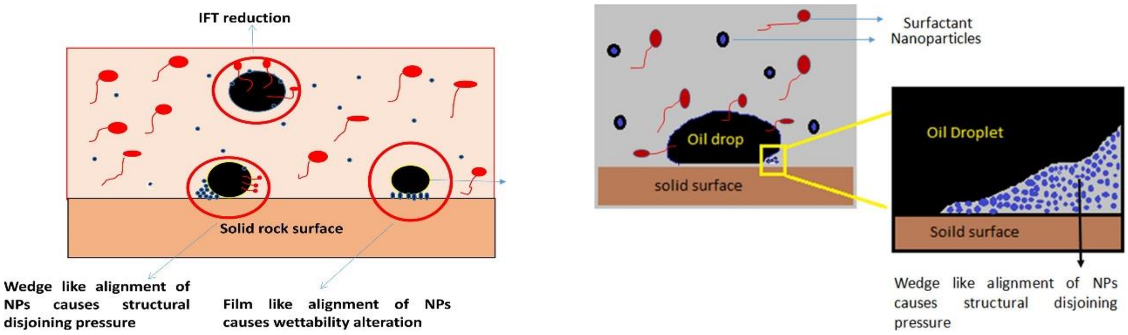

Figure 1. Shows surfactant-NP arrangement for wettability alteration and IFT reduction [4]

IV. MICROEMULSION AND ITS CHARACTERIZATION

Emulsions are formed when surfactants and co-surfactants are combined with oil and saline water. Microemulsions (MEs) are a thermodynamically stable, lucent, and clear analogous combination of petroleum and water with surface-active agents. Because of its exceptional ability to reduce IFT at the oil-water interface and change the degree of wetting of rocks, it can be a wise choice or defined as an effective framework for EOR process implementation. Microemulsion flooding is feasible in a wide range of subsurface settings. The use of microemulsion in support of oil revival is not a modern advancement in petroleum expertise. The desirable solubility proportion and similarly low IFT of microemulsion make it more proficient and sensible for the EOR execution most assuredly. Winsor, 1954 characterized microemulsions into three types like Type I, Type II, and Type III. Type I can be made sense of as an O/W microemulsion where oil is solubilized by the additional surfactant as shown in Figure 2. Simultaneously, Type II can be characterized as a W/O microemulsion in which water present is solubilized by the surfactant. Furthermore, in Type III of the microemulsion, both the oil portion and water fragment are solubilized by a surfactant and it very well may be potentially alluded to as in balance with overabundance oil and water stage. As made sense of by Winsor, Type III is more thought of and ideally picked as a replaceable liquid for EOR [30], [4]. The most important point that needs to keep in about the microemulsion study is the phase behaviour. The phase characteristics might direct us as a mark of ultra-low IFT [31]. The phase behavioural studies of microemulsions supports the quick evaluation of the most ideal and positive surfactant plans. The highlights of microemulsions have exhaustively been broke down by a few exploration laborers (For example, it very well may be made sense of that the saltiness addition might cause the change of microemulsion through middle person stages). That is Type I to Type II through Type III, and that implies that the design and properties of the microemulsion can be changed or altered by the saltiness varieties.

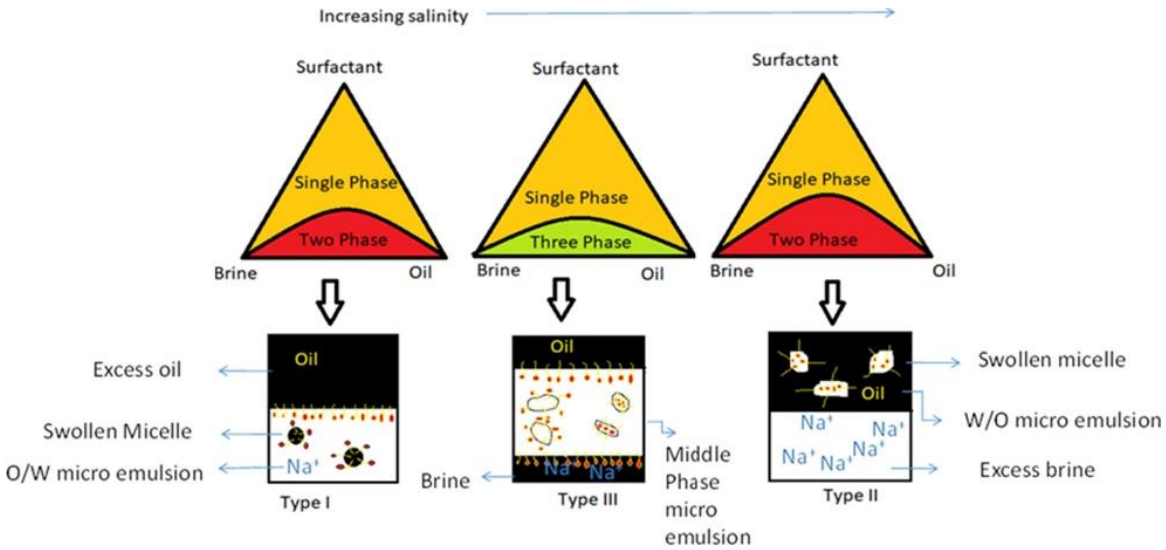

Figure 2. Shows Phase transition of ME under varying salinities [4]

For the creation of a successful microemulsion, proper characterisation is the first and most important stage. Analyzing a microemulsion's characteristics makes it easier to determine its chemical make-up and molecular structure. It is necessary to characterise the phase behavioural behaviour of the microemulsion in order to better determine the optimal formulation for efficient EOR execution and EOR. A microemulsion formulation is characterised by a few key factors, including IFT, wetting degree, viscosity, O/W solubilization ratio, coalescence time, and adsorption behaviour as depicted in Figure 3. This characterisation aids in comprehending the unique characteristics of the microemulsion for improved core flooding performance [4]. All of these characterization studies revealed a more effective technique to formulate a formulation for a reservoir that will successfully stimulate the residual oil. For the intended formulation, the right amount of surfactant, co-surfactant, oil, and water is needed. Use of the optimal amounts of the required could result in a formulation that is both economical and effective. An optimised formulation might be anticipated to have a high level of oil extraction efficiency [32], [33]. This contributes to reducing the overall cost of the EOR process.

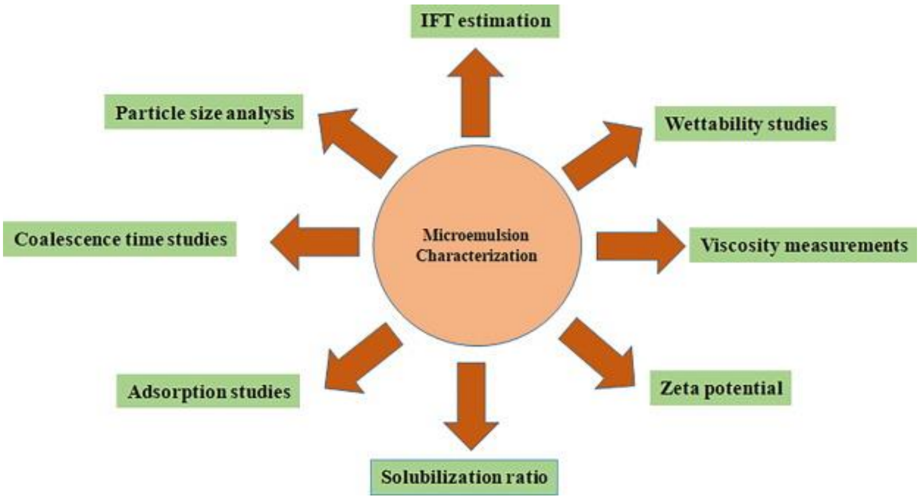

Figure 3. Shows various ME characterisation techniques [5]

V. APPLICATION OF GNPS AS STABILITY ENHANCER FOR ME

Micro emulsions can sometimes be unstable in the reservoir environment, hence adding Nano particles to these micro emulsions can enhance its stability. The use of NPs to stabilize ME has a number of benefits, such as increased conformance control, reduced surfactant consumption, and high tolerance to temperature and salinity in reservoirs. NP surface wettability can be tuned to generate ME droplets in desired shapes and sizes. The NP can also serve as a sensor and have other functionalities that interact with changes in temperature, pressure, and particular chemicals, among other things. Despite the fact that silica is the particle that is most frequently employed to stabilize ME, only a small number of alternative NPs, such as hydrophilic silica NPs, partially hydrophobic modified SiO2 NPs, and partially hydrophobic clay particles.

A. **Visual Stability Analysis:**

The MEs formulated by synthesized NPs were initially screened by means of visual inspection and classified into different groups according to Winsor classification. In present application of GNPs, initially MEs formulations were prepared in 1:1 ratio by varying the concentration of synthesized GNPs 0, 0.025, 0.05, 0.075 and 0.1 % and visual stability analysis was performed as shown in Figure 4. In the figure it can be observed that 0.05% and 0.075% concentration of silica NPs resulted in better visual stability and better Winsor Type III Microemulsion formation.

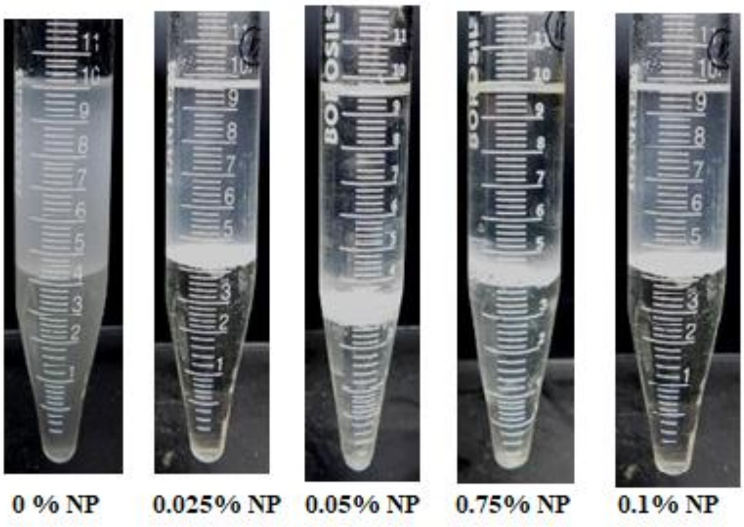

Figure 4. Decane oil/ surfactant microemulsion with varying synthesised NP concentration

B. **DLS and Zeta Potential for droplet stability analysis:**

Dynamic light scattering (DLS) is a technique for quickly and accurately determining the size or size distribution of a distinct species dispersed in a continuous phase as well as the translational particle diffusion coefficient. The Stokes-Einstein relation can be used to determine the hydrodynamic size of the dispersed species if the diffusing species are spherical, non-interacting bodies [34, 35]. The DLS of the synthesized MEs were investigated with varying time and concentration to determine the droplet size and stability to understand its coalescence behavior as well. The droplet size with varying concentration which shows that the hydrodynamic radius was lowest in the case of 0.75% in comparison to other concentrations. It was observed that it decreased with increasing concentration till 0.75% and after that it increased which may be due to increased coalescence rate. The zeta potential is a physical property that any particle in suspension, macromolecule, or material surface exhibits. It can be used to optimize suspension, emulsion, and protein solution formulations, predict interactions with surfaces, and optimize film and coating formation. Knowing the zeta potential can help you save time when creating trial formulations [36]. It can also be used to forecast long-term stability. For a microemulsion to be stable, zeta potential value should be +/-35mV. Since the prepared microemulsion has values corresponding to -35 mV, it could be concluded that the prepared microemulsion was stable as shown in Figure 5.

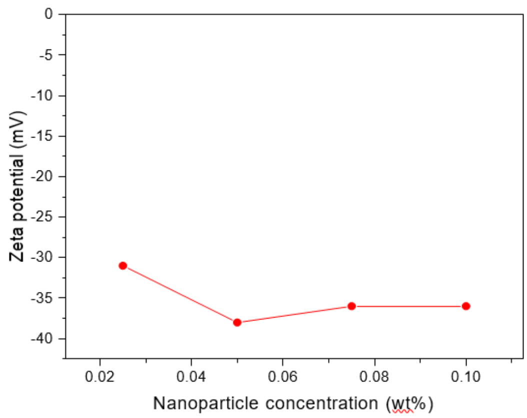

Figure 5. Shows variation of Zeta Potential with respect to change in NP concentration

VI. SHIFT TOWARDS SUSTAINABILITY

The sustainability challenges currently faced in global platform are complex and nanotechnology has emerged as a technical solution to them. Nanomaterials have unique physicochemical properties that make them particularly appealing as functional materials for green technologies. To increase their affinity for a specific compound, such as dissolved solutes or gases, nanomaterials can be functionalized with different chemical groups. Nanomaterials are also enabling the development of functional materials with superior properties that can be applied to harsh reservoir conditions. The use of green NPs in the petroleum industry could reduce the existing potential risk to the environment as well as the risk of potentially negative effects in natural systems.

The nature is well suited to producing a repertoire of nanomaterials through processes such as combustion, abrasion, precipitation, and oxidation, as well as bio-reduction and related processes. Thus this has fueled research into these native materials and processes, as well as strategies for producing biological and biologically active materials using similar materials and methods. It is now time to take stock and look forward to some of the most exciting developments in the coming years.

VII. SUMMARY

In summary, we have discussed various biological or eco-friendly green nanomaterial synthesis and its petroleum applications. Though physical and chemical methods for producing nanomaterials are available, biological methods are preferred because they are less hazardous than chemical methods. Plant extracts, naturally derived polysaccharides, and microbes are the materials of choice for satisfying the desire for suitable methods for biological production of NPs. The synthesis of green NP and its application in EOR has been elaborated. The sustainable development of ME formulations using synthesised NP and its stability was verified by DLS and zeta potential studies. The application of ME for recovery of residual oil from reservoir could be applied in later stages to analyse its potential. However, some numerous concepts, such as methods for large-scale production at a lower cost, must be investigated further for its industrial application.

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