GREEN NANOPARTICLES FOR SUSTAINABLE OIL RECOVERY

Abstract

The current global trend for the 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 microorganisms has gained attention among researchers due to their improved properties and sustainability. The present chapter provides essential knowledge and understanding of the application of green nanoparticles in Enhanced Oil recovery (EOR), with particular emphasis on its application in microemulsion.

Keywords: Green nanoparticle, Enhanced oil recovery, Microemulsion (ME), Winsor classification, Interfacial tension

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I. INTRODUCTION

In global platform petroleum resources still continues to be the primary source of energy. To meet rising energy demand, improved oil extraction from untapped reservoirs is essential. As a result, once these energy sources have been depleted and exploited, they cannot be reused, emphasizing the significance of sustainability in energy production. The concept of sustainability is the development and use of products based on need and demand without jeopardizing the needs of future generations. As the environment is a limited resource, it is critical to use it wisely, thus it is necessary to maximize oil recovery. Around 30 % of the original oil in place can be recovered by using conventional resources. To meet high energy demands, advanced methods must be developed to enhance the recoverable volumes from the existing reservoirs [1]. Therefore, Enhanced Oil Recovery (EOR) techniques are needed to improve residual oil recovery. The EOR aims to increase recovery by infusing additives such as chemicals, gases, or thermal energy into a reservoir. The abovementioned injection helps upgrade the existing reservoir properties for displacing the oil to the producer well. The 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 an advanced EOR Strategy which delivers quite a good amount of oil in the overall global oil outlook [2].TEOR processes are more convenient for sandstone reservoirs and for high-density, thick and viscous oils with API gravity of less than 20. Chemical Enhanced Oil Recovery (CEOR) is a critical EOR approach that 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 fiscally viable strategy in EOR. In Miscible Enhanced oil recovery method, gases like carbon dioxide or nitrogen low molecular weight hydrocarbons are infused into the reservoir. The injected gas gets miscible with the crude oil in deep reservoirs at moderate temperature conditions, enhancing production [3].

Last but not least, Microbial Enhanced Oil Recovery (MEOR) can be considered as a significant one in considering sustainability. The effective mechanisms for improving the recovery include Interfacial tension (IFT) reduction, reduction of viscosity, oil swelling, and 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, chemical flooding has made significant progress, raising the possibility of it becoming the most critical EOR method [6]. Many countries, including the United States, Germany, China, Austria, Austria, and Canada, have reported success with chemical EOR. However, chemical flooding is an expensive recovery method due to the high cost of chemicals. As the industry is moving more towards sustainability, it is essential 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 the case of sustainable biomaterials, GNPs have created more interest among researchers for their further development and applications.

Nanotechnology is profoundly appealing but stimulating in the petroleum industry to enhance oil recovery [3] due to its unique properties such as wettability alteration, improved mobility of trapped oil, enhancing sand consolidation, and reduction of interfacial tension (IFT) [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 repulsive and attractive 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 the petroleum industry, especially in EOR. Amongst the various mechanism of oil recovery, for example, IFT reduction, wettability alteration, Micro Emulsion (ME), etc., the least discovered area is MEs. The formation of in-situ MEs and injection of ex-situ MEs formed by surfactants increase the residual oil recovery and overall efficiency. The ex-situ ME formation and stability are two crucial parameters to be considered for its characterization. The application of green NPs to increase the stability and morphology of ME could be a sustainable path to be explored in the future for improved oil recovery.

II. NANOPARTICLES

NPs can be referred to as particles ranging from 1 to 100 nanometers, usually made of carbon, metal, metal oxides, or organic matter. The NPs possess unique physical, chemical, and biological properties due to their distinct properties, such as the larger surface area to the volume, increased reactivity or stability in a chemical process, enhanced mechanical strength, etc. [11, 12]. These properties cause significant changes in their physical properties compared to those observed in bulk materials, which has led to their use in various applications. The NPs can be zero-dimensional, one-dimensional, two-dimensional, and three-dimensional based on their length, breadth, and height. Zero-dimensional, like quantum dots, are those in which the dimensions of length, breadth, and height are all fixed at a single location. One-dimensional materials, like graphene, only have one parameter and let electron motion solely in the X direction. Two-dimensional NPs have length and breadth, but they can only move in the X-Y plane. Three-dimensional NPs, like gold NPs, have all the parameters, including height, length, and breadth, and they can travel in all three directions NP research is currently an area of intense scientific interest [12].

1. **Classification of NPs:** The NPs utilized for various industrial applications are generally categorized as organic, inorganic and carbon-based NPs. The organic NPs are synthetic organic molecules that are non-toxic, biodegradable and sensitive to electromagnetic and thermal radiation. Some 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, and 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 nanotubes, 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 follows:

   - **Carbon-Based NPs:** The two most common types of carbon-based NPs are Fullerenes and Carbon Nanotubes (CNTs). Fullerenes contain nanomaterials made of globular

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hollow cages, such as allotropic carbon forms with superior properties such as higher strength, electrical conductivity, electron affinity, structure, 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 applications in the oil and gas industry and particularly in EOR. The wettability of the sandstone substrates can be altered using different concentrations 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 increases the disjoining pressure [17]. Another application of different concentrations of GO nanosheets on carbonate slices showed similar results of wettability alteration. Kanj et al. examined the flooding performance in carbonate reservoirs using modified carbon NPs and found a rise in oil recovery factors [18].

- **Metal NPs:** Metallic NPs primarily comprise metal precursors such as gold, silver, copper, palladium, and platinum. Metallic NPs have essential 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 $Al2O_3$ 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].

- **Polymeric NPs:** Polymeric NPs are particles having active compounds entrapped within or surface-adsorbed onto the polymeric core with sizes 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 have recently been investigated for EOR applications [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 characterized by their morphological structure. Nanocapsules are primarily deduced as matrix particles whose overall mass is primarily solid, and the other molecules are adsorbed at the outer boundary of the spherical surface, while nanospheres are predominantly those particles whose solid mass is encapsulated within the particle entirely [22].

2. **Green NP as a Sustainable Resource for EOR:** In most cases, NPs for EOR applications are dispersed in fluids such as oil, deionized water, brine, or gas to form nanofluids. Various mechanisms include wettability alteration by NP adsorption and disjoining pressure, IFT reduction at the oil-water interface, viscosity adjustment by polymer, surfactants etc., pore channel plugging and prevention of asphaltene precipitation effects are examined to investigate the effects of nanomaterials on EOR. Depending on the application, surface ligands, stabilizers, and polymer/surfactant, nanomaterials can have hydrophilic, hydrophobic, or double-faced (Janus) properties [23]. SiO_2 and TiO_2 NPs have been shown to improve sandstone oil recovery by altering wettability and decreasing IFT [24]. NP deposition could clog water channels and

increase sweep efficiency during flooding. Similarly, SiO_2, Al_2O_3, and TiO_2 NPs have been shown to ameliorate oil recovery from carbonates through wettability modification, IFT reduction, and viscosity modification [25]. The use of nanofluids could improve the performance of brine injectioneven further.

The high interfacial activity of NPs and nanofluids results in upgrading production from oil reservoirs, as NPs havehigh surface area to volume ratios which adsorb at fluid-fluid interfaces and reduce IFT, which is further enhancedin the presence of surfactants, as shown in Figure 1. The properties and quantities of NPs determine the enhanced emulsification ability. It can be delineated that as the oil mass ratio of NPs increases, the size of emulsified oil droplets decreases[26]. NPs can alter the solid surface wettability due to their self-structuring behavior, which raises the structural disjoining pressure in the constricted three-phase contact region, explicitly 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 removed to be about 70% afterNP-stabilized emulsion flooding [27]. The NP stabilised ME formulation has improved residual oil recovery to 19% with higher oil displacement efficiency as compared to ME alone. 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. Even while NP-stabilized MEs typically perform better than MEs alone, little is understood about their displacement process in porous media and the kinds of fluid-fluid and fluid-rock interactions. The synthesis of NPs is 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, raising the temperature around the source material. The toxic nature of the solvents and chemicals could cause harm to an already degrading environment [28]. Thus, the dire need for developing alternative methods for greener development has led to the concept of green nanotechnology. Green nanotechnology in the petroleum industry and EOR majorly use plants, fungi or bacteria as its source for NP synthesis, which is eco-friendly.

The utilization of plants for NPs production has drawn attention in recent years because its eco-friendly, rapid, economical, and non-pathogenic protocol delivers a single-step technique for the biosynthetic process. The primary benefits of using plant extracts for NP synthesis are that they are readily available, harmless, and non-hazardous in most cases, contain a diverse range of metabolites that can aid in silver ion reduction, and are faster in synthesis than microbes [29]. Our research used a similar technique to synthesize Si-NPs from bamboo leaves. Bamboo Leaf Ash was processed 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.

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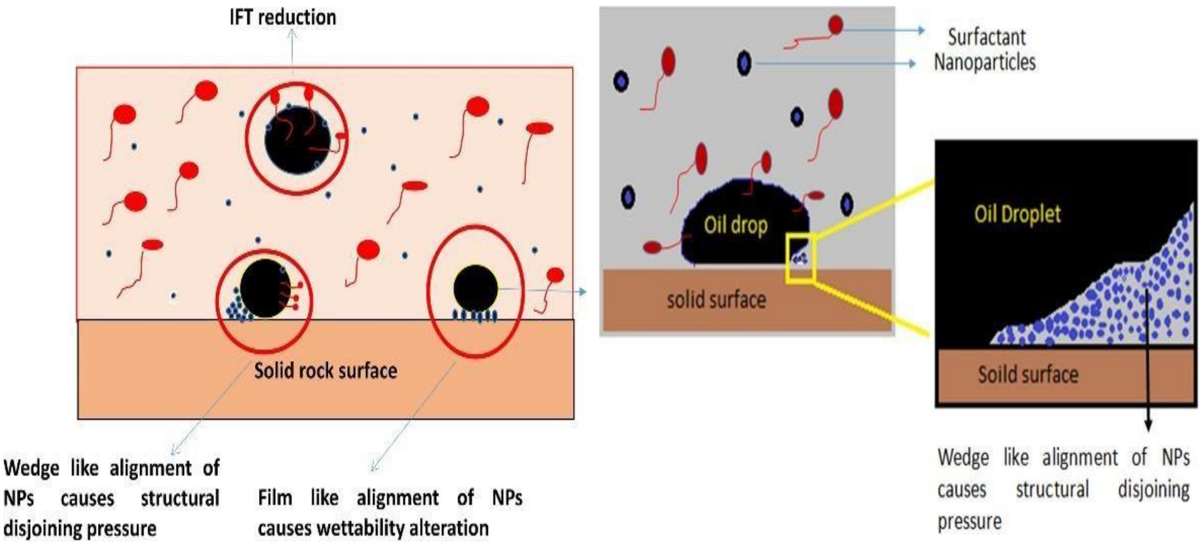

Figure 1: Shows Surfactant-NP Arrangement for Wettability Alteration and IFT Reduction [4]

3. **Microemulsion and its Characterization:** Surfactants and co-surfactants combine with oil and salt to form emulsions. MEs is a thermodynamically stable, clear, and a lucent analogous mixture of oil and water containing surface-active agents. It can be a critical choice for its application in EOR processes due to its exceptional ability to reduce IFT at the oil-water interface and change wettability. The ME flooding is feasible in a wide range of subsurface settings. The use of ME in support of oil revival is not a modern advancement in petroleum expertise. The desirablesolubility proportion and similarly low IFT of ME make it more proficient and sensible for the EOR execution. Winsor, 1954 characterized MEs into three types Type I, Type II, and Type III. Type I is an O/W ME in which oil is solubilized by an additional surfactant, as shown in Figure 2. At the same time, Type II is a W/O ME in which the surfactant solubilizes the water present.

   Furthermore, in Type III of the ME, both the oil and water fragment are solubilized by a surfactant and it may be potentially alluded to as in balance with overabundance of oil and water stage. As made sense by Winsor, Type III is more thought of and ideally picked as a replaceable liquid for EOR [30]. The most crucial point to remember about the ME study is the phase behavior. The phase characteristics might direct us as a mark of ultra-low IFT [31]. The phase behavioral studies of MEs support the quick evaluation of the most ideal and positive surfactant plans. The highlights of MEs have exhaustively been broken down by a few researchers and numerous studies has been performed to determine the parameters affecting it behavior and design accordingly for EOR application.

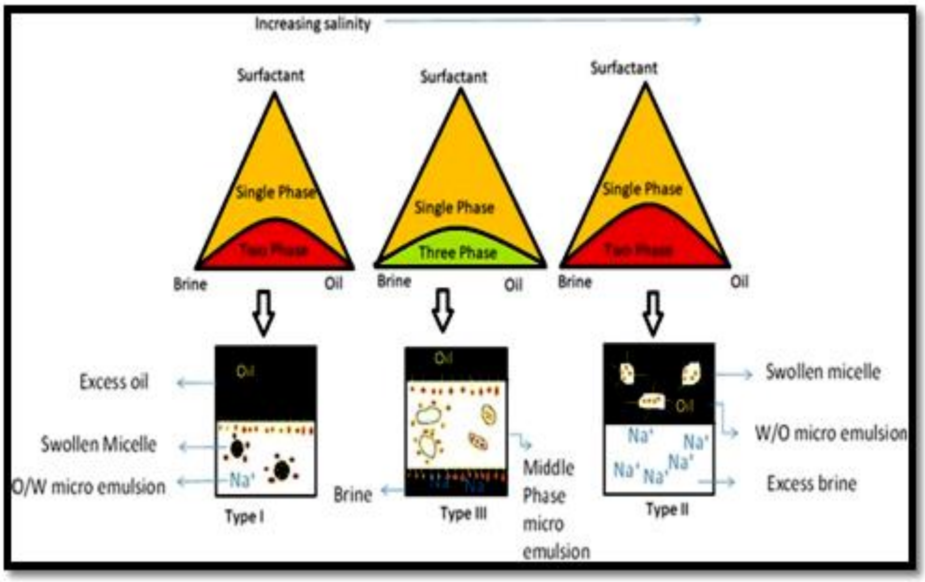

Figure 2: Shows Phase transition of ME under varying salinities [4]

For the creation of a successful ME, proper characterization is the first and most crucial stage. Analyzing a MEs characteristics makes it easier to determine its chemical make-up and molecular structure. It is necessary to characterize the phase behavioral behavior of the ME to determine better optimal formulation for efficient EOR execution and EOR. ME formulation is characterized by key factors, including wettability, IFT, viscosity, coalescence time, O/W solubilization ratio, and adsorption behavior, as depicted in Figure 3. This characterization aids in comprehending the unique characteristics of the ME for improved core flooding performance. These characterization studies revealed a more effective technique to formulate a formulation for a reservoir that will successfully stimulate the residual oil. The right amount of water, oil, surfactant, and co-surfactant is needed for the intended formulation. Use of the optimal amounts of the required could result in a formulation that is both economical and effective. An optimized 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.

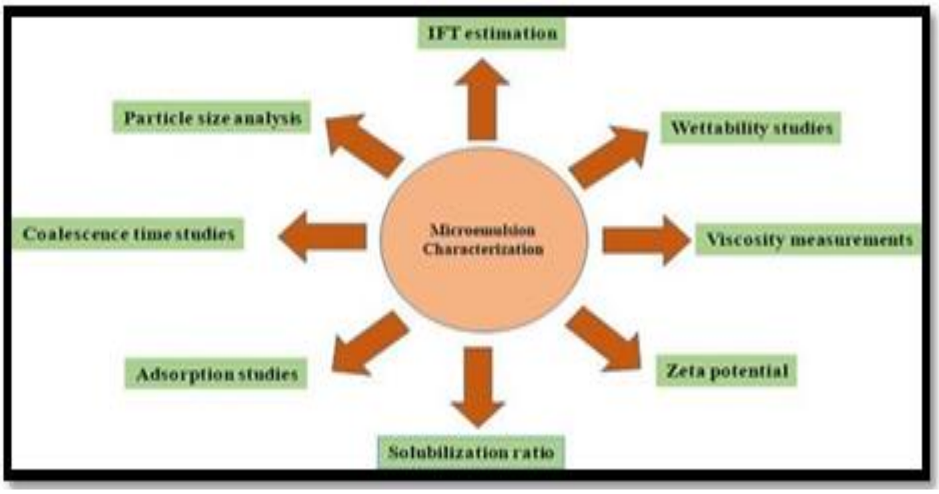

Figure 3: Shows Various ME Characterisation Techniques [5]

4. **Application of GNPs as Stability Enhancer for me:** MEs can sometimes be unstable in the reservoir environment; hence adding NP to these MEs can enhance its stability. The use of NPs to stabilize ME has many 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, andparticular chemicals, among other things. Even though silica is the particle most frequently employed to stabilize ME, only a small number of alternative NPs, such as hydrophilic silica NPs, partially hydrophobic modified SiO_2 NPs, and partially hydrophobic clay particles.

    - **Visual Stability Analysis:** The MEs formulated by synthesized NPs were initially screened employing visual inspection and classifiedinto different groups according to Winsor classification. In the present application of GNPs, initially, MEs formulations were prepared in a 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. The figure shows that 0.05% and 0.075% concentrations of silica NPs resulted in better visual stability and Winsor Type III Figure 4 shows ME formation.

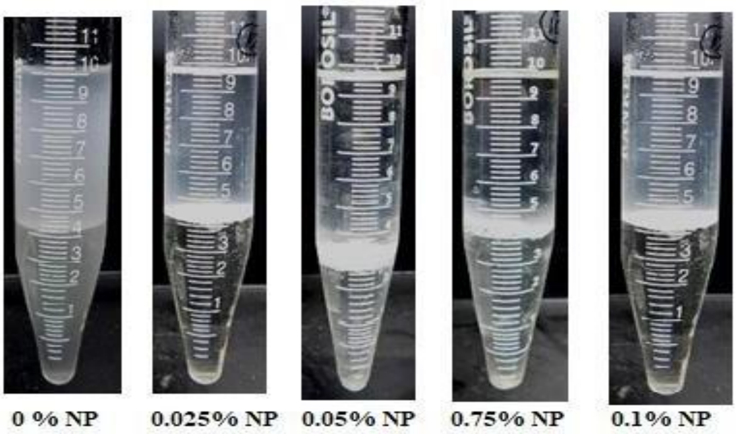

Figure 4: Decane Oil/ Surfactant Microemulsion with Varying Synthesized NP Concentration

    - **DLS and Zeta Potential for Droplet Stability Analysis:** Dynamic Light Scattering (DLS) is a technique for quickly and accurately determining the size or distribution of a distinct species dispersed in a continuous phase and the translational particle diffusion coefficient. The Stokes-Einstein relation can be used to determine the hydrodynamic size of the dispersed species if the diffusingspecies are spherical, non-interacting bodies. The DLS of the synthesized MEs were investigated withvarying time and concentration to determine the droplet size and stability to understand its coalescence behavior. The droplet size with varying concentrations shows that the hydrodynamic radius was lowest in the case of 0.75% compared to other concentrations. It was observed that it decreased with increasing concentration to 0.75%, and after that, it increased, which may be due to an increased coalescence rate. The zeta potential is a physical property exhibited by any particle in suspension, macromolecule, or material surface. 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. It can also be used to forecast long-term stability. For a ME to be stable, the zeta potential value should be +/-35mV. Since the prepared ME has values corresponding to -35 mV, it could be concluded that the prepared ME was stable, as shown in Figure 5.

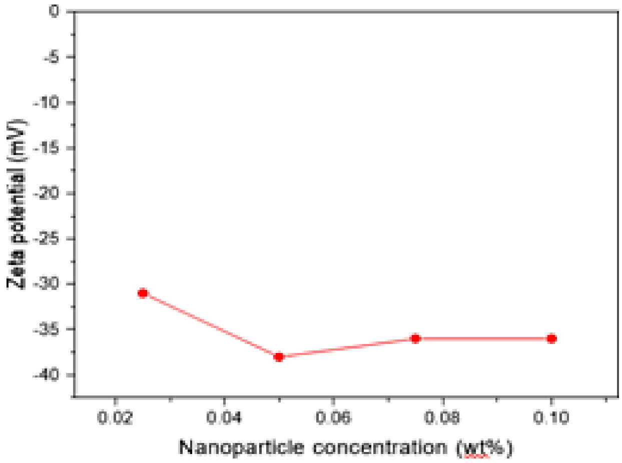

Figure 5: Shows variation of Zeta Potential with respect to change in NP concentration

- **Shift towards Sustainability:** The sustainability challenges currently faced in the global platform are complex, and nanotechnology has emerged as a technical solution. 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 also enable the development of functional materials with superior properties that can be applied to harsh reservoir conditions. Using green NPs in the petroleum industry could reduce the potential risk to the environment and the risk of potentially adverse effects on natural systems.

  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 techniques, as well as strategies for producing biological and biologically active materials using similar materials and methods. It is time to take stock and look forward to some of the most exciting developments in the coming years.

III. SUMMARY

In summary, we have discussed various biological or eco-friendly green nanomaterial synthesis and their petroleum applications. Though physical and chemical methods for producing nanomaterials are available, biological processes 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 synthesized NP and its stability was verified by DLS and zetapotential studies. The application of ME for recovery of residual oil from the reservoir could be applied in later stages to analyze its potential. However, numerous concepts, such as methods for large-scale production at a lower cost, must be investigated further for their industrial application.

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