

Stability Evaluation of Zinc Oxide Nanofluids Using Mangifera Odorata Extracted Peels and Leaves

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Abstract

The most widely utilized materials to produce nanofluids (NFs) are metal oxides, which have limitations regarding dispersion stability. Producing stable NFs is a crucial step in enhancing their functionality in real-world applications. This study aims to determine the ideal conditions for NFs utilizing a more ecologically friendly technique by synthesizing zinc oxide (ZnO) NFs using Mangifera Odorata (MO) peel and leaves extract. The stability of NFs was assessed using an optical photograph, an ultraviolet (UV) – visible spectrophotometer, and a Fourier Transform Infrared Spectrophotometer (FTIR) under different conditions, including varying extracted MO to ZnO ratios and extracted MO pH. The results show that Zinc Oxide-Mangifera Odorata peel (ZnO-MOp) and Zinc Oxide-Mangifera Odorata leaves (ZnO-MOl) NFs at pH 8 and MO extract to ZnO ratios of 1:9 indicate the most stable NFs due to the low separation rate sedimentation over 18 and 24 hours of observation. Furthermore, it was discovered that the existence of Zn-O stretching functional groups was demonstrated by the strong peaks in the 585–590 cm^{-1} spectrum. These findings would eventually serve as a benchmark for future study, resulting in more stable and high-quality ZnO-MO NFs for use in wastewater treatment.

1. Introduction

Nanofluids (NFs) are stable suspensions of nanoparticles in a base fluid with sizes ranging from 1 to 100 nanometers [1]. This makes them valuable for applications that necessitate efficient heat transfer. Nanomaterials, such as nanoparticles and NFs, have distinct features owing to their atom configurations and tiny surface area, making them beneficial in a variety of industries, including chemistry, manufacturing, medicines, and cosmetics [2]. These materials can be synthesized physically or chemically, with colloidal precipitation being a common procedure. These nanofluids can improve processes such as oil recovery, heat transmission, lubrication, and the administration of pharmaceuticals. The application of ZnO nanofluids has been widely used in various areas such as wastewater treatment, biomedical materials, paint industry, cosmetics and detergents. Previous researchers, Pauzi et al. [3] revealed that ZnO nanofluids have potential in health-related applications with effective antibacterial activities. NFs can be produced through a single or two-step process. According to Mahian et al. [2], researchers tend to employ two-step approaches more frequently than one-step methods. In the one-step technique, base fluids and nanoparticles are mixed. The two-step process, on the other hand, involves first

preparing the nanoparticle and then employing a greater shear mechanism to disseminate it throughout the fluid [4].

The constraint of nanofluids is that they are unstable when dispersed in water due to ZnO aggregation caused by the strong polarity of the water, resulting in sedimentation. NFs in base fluids are typically subjected to Van der Waals forces and Ostwald ripening interactions, which cause the particles to coalesce. As a result, it will result in minimal dispersion and stability, which may lessen the impact of nanoparticles in the solution [5]. The dispersion's stability is essential to the nanofluid's thermal conductivity. Thus, many researchers are interested in identifying potential control mechanisms to increase the stability of ZnO nanofluids, such as using alternative capping or stabilizing agents [3, 6].

The green production of nanoparticles often entails the extraction of bioactive substances with non-toxic solvents, such as distilled water, prior combining them with metal precursors. Numerous researchers have investigated utilizing algae, plants, fungi, and bacteria for developing green synthesis nanoparticles and nanofluids. Plants typically contain numerous kinds of phytochemicals, including flavonoids, phenolic compounds and alkaloids, which can act as a capping and reducing agent for metal oxide nanoparticles [7]. The plant extract's phytochemicals can also operate as a stabilizing agent, preventing the resultant nanoparticles from agglomerating [8]. Green synthesis is a viable substitute for traditional methods since it is an environmentally friendly and sustainable way to produce stable nanoparticles [9]. Conversely, conventional methods may be hazardous due to the use of toxic compounds and solvents [9].

Hai et al. [10] discovered that silver nanoparticles (AgNPs) were successfully produced employing mango (*Mangifera Indica*) leaf extract as a green reducing agent, stabilizer, and agglomeration inhibitor. Horta-Piñeres et al. [9] also reported an ecological approach for the synthesis of silver nanowires (AgNW) decorated with silver nanoparticles (AgNP) using *Mangifera Indica* leaf extract obtained small nanoparticles with sizes < 10 nm. According to the study of Salahuddin et al. [11] investigated a comparative of total phenolic contents of *Mangifera Odorata* contained high total phenolic content (257.17 ± 27.72). These phenolic compounds act as reducing, stabilizing, growth limiting and agglomeration prohibiting agents, which could possibly be used for the fabrication of nanoparticles [12]. Hence, it is believed that MO is a potential capping agent to be applied in the synthesizing of ZnO nanoparticles due to their properties.

Mangifera odorata (MO) and *Mangifera indica* are mango species. According to Hai et al. [13], the *Mangifera indica* extracted leaves cause the particle size to progressively decrease. However, Khoiriah and Putri [14] found that the peel extract has a significant concentration of phytochemical components that provide capping and reducing agents in the formation of nanoparticles. Saleh et al. [15] discovered that extracted mango peels contained increased polyphenol contents. Thus, the goal of this work is to compare the potential of extracted peel and leaves that may provide a significant quantity of stabilizer, which can prevent ZnO-NF formation from self-agglomerating.

There has been inadequate research on the stability of ZnO NFs using extracted MO peel and leaf as a stabilizing or capping agent. Further research is required to determine the significance of regulating characteristics affecting the stability of NFs. Consequently, the current study investigates the stability of ZnO–MO NFs with the presence of extracted MO peel and leaves under various pH levels and extracted MO to solution ZnO ratios.

2. Materials and Methods

2.1 Materials

The *Mangifera Odorata* peel and leaves were collected near a garden at Johor, Malaysia. The chemicals required including Urea (2 M)(Chemiz/Malaysia), Zinc acetate dehydrates (0.01 M), Choline chloride (1 M) (Chemiz/Malaysia) and Sodium hydroxide (0.02 M)(NaOH) that were purchased from BT Scientific in Malaysia.

2.2 Preparation of *Mangifera Odorata* Peel and Leaves Extract

Mangifera Odorata (MO) fruits were washed and then the skin was peeled. Next, the peels were rinsed with water and left to dry for a day under the sun. Dried peels then were grinded by using a heavy-duty blender and sieved until a smooth textured powder was produced in the range of 100 microns. After that, the powder was mixed with deionized water according to the ratio of 1:10 and were placed inside an incubator shaker at 80°C and 180 rpm for 60 minutes [16]. Lastly, the extract is then filtered by filter paper. The filtered MO peels (MOp) extract was kept in a closed container and preserved in a freezer at 4°C to maintain the sample purity and readiness. A similar method was carried out for extracting MO leaves (MOL).

2.3 Synthesis of Zinc Oxide – *Mangifera Odorata* Peels and Leaves Extract

A solvent called deep eutectic solvents (DES) combines urea and choline chloride, which serve as base fluids to create nanofluids [17]. DES is prepared in which is going to be used as a medium reaction. The 1 M choline chloride

(13.96 g) and 2 M urea (12.01 g) with 1:2 ratio w/w% were mixed and heated using water bath at 80°C and 180 rpm for 30 minutes [18]. Then, it was left to cool at room temperature. 0.01 M of zinc acetate dehydrates solution were dissolved in 25 mL of DES using magnetic stirrer. 0.02 M of sodium hydroxide (NaOH) is then added into the solution. Afterward, the 10 mL of MOp extract is added according to ratio of 1:1, 1:3, 1:5, and 1:9 with ZnO solution. The solution was stirred for a day to increase ZnO–MOp NFs production. The produced ZnO–MOp NFs were stored in sealed containers at 25°C. The process was repeated by using different pH of extracted MOp (6, 7, 8). The selection of the factors' experimental range was predicated on the findings of earlier research by Sidik et al. [16] and Umar et al. [19]. The same approach and parameters are used in the comparative investigation of ZnO–MOI. Fig. 1 summarizes the preparation technique of ZnO–MOp and ZnO–MOI NFs.

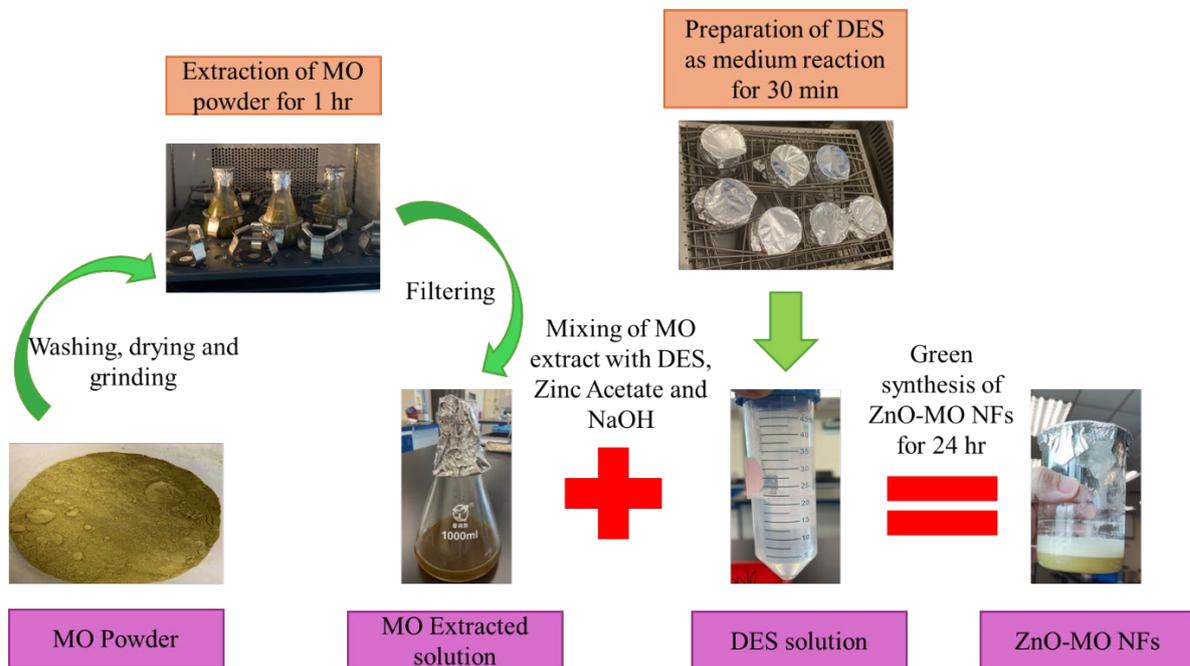


Fig. 1 Schematic illustration for green production of ZnO–MOp and ZnO–MOI NFs

2.4 Characterization of ZnO–MO Nanofluids

The assessment of ZnO–MO NFs stability involved visual observation through photography, and analysis of optical absorption spectra using a UV–visible spectrophotometer (model U–3900H). A Fourier Transform Infrared Spectrophotometer (FTIR; Agilent Tech Cary 600 series) was used to examine the chemical composition of the generated nanofluids. The liquid sample has been placed into the FTIR spectrometer's sample holder. The FTIR spectrum was then generated between 400 and 4000 cm^{-1} wavelengths [3].

3. Results and Discussion

3.1 ZnO–MOp and ZnO–MOI NFs Stability Analysis

Nanofluids' physical stability is vital for achieving uniformity as well as consistency, leading to improved future applications. Nanofluid destabilization occurs when suspended particles in the base fluid settle, causing nanoparticle sedimentation [1]. The sedimentation approach was employed to investigate the dispersion stability of ZnO–MOI and ZnO–MOp NFs over time using visual inspection. During the preparation of nanofluids, big particle agglomerates are separated gravitationally. Agglomeration may occur when the suspension's nanoparticles interfered with one another, forming huge clusters [1]. Fig. 2 (a) and (b) depict the samples for ZnO–MOp and ZnO–MOI NFs after 18 and 24 hours of investigation, respectively.

Fig. 2 (a) demonstrates that ZnO–MOp NFs 1:3 exhibits modest particle settling in comparison with ZnO–MOp NFs 1:1 for 18 hours, implying instability. Furthermore, after 18 hours, there have been no discernible changes in the sedimentation of the ZnO–MOp NFs 1:5 and 1:9 samples. The order of instability for all ZnO–MOp NFs samples over 24 hours was ZnO–MOp 1:5 < ZnO–MOp 1:3 < ZnO–MOp 1:1. In the meantime, ZnO–MOp NFs 1:9 demonstrates that no evidence of particle settling was observed after a 24-hour period, indicating that the NFs are physically stable. Therefore, ZnO–MOp NFs 1:9 yielded the most stable NFs with an extracted MOp capping.

The agglomeration of NFs is caused by van der Waals forces that exist between nanoparticles, resulting in an increase in sedimentation rate during NFs production [5]. Thus, it is possible to improve the dispersion stability of NFs with the aid of extracted MOp.

The evaluation of ZnO–MOI NFs samples at various ratios (1:1, 1:3, 1:5, and 1:9) in static settings reveals no sign of settlement during the first 18 hours, and the rate of sedimentation increases after 24 hours for ZnO–MOI NFs (1:1, 1:3, 1:5), as depicted in Fig. 2 (b). The results show that adding more ZnO solution boosted the stability of the ZnO–MOI NFs, as evidenced by the fact that ZnO–MOI NFs 1:9 demonstrate no change in sedimentation over 24 hours, indicating that it is the ideal ratio. ZnO NPs are uniformly distributed in DES and exhibit improved stability due to the bioactive ingredient in extracted MOI acting as a capping or stabilizing agent that coats the ZnO and inhibits further agglomeration [8]. Prior studies conducted by Bin Fazal and Ahmad [20] have also demonstrated that the bioactive compounds found in extracted plants, including alkaloids, polyphenols, and flavonoids, may mitigate ZnO agglomeration. In conclusion, lowering the MO extract to ZnO ratio may improve ZnO NFs stability.

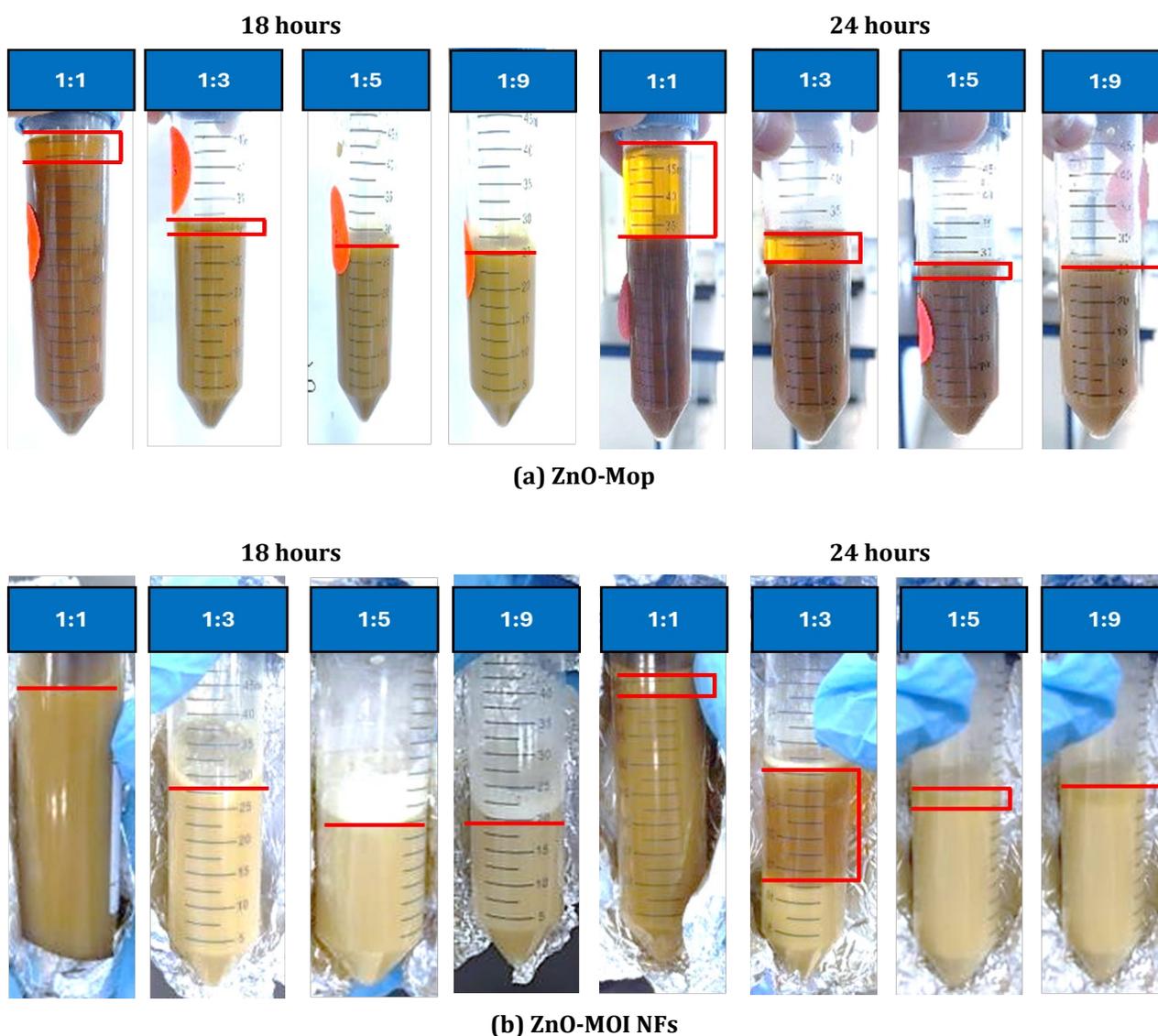


Fig. 2 Stability of a) ZnO–Mop; and b) ZnO–MOI NFs for 18 and 24 hours at varying ratio of MO extracted with ZnO solution and a constant pH of MO extracted solution

Establishing the ideal pH is crucial to formulate NFs that regulate colloidal particle sizes because it has an impact on the stability of surface charge and particle interactions [5]. Fig. 3 (a) and (b) illustrate the stability test results for ZnO–MOI and ZnO–MOp NFs (with ratio extracted MO and ZnO was 1:9) over 18 and 24 hours, respectively, under pH levels 6, 7, and 8. The pH value of the MO extracted solution has a direct impact on the stability of a NFs. Extracted MOp with pH 6 exhibited higher settlement growth at 18 hours and increased at 24

hours compared to pH 7 and 8 (Fig. 3 (a)). This signifies an unstable condition due to increased sedimentation development in the sample tube. This phenomenon could be attributed to the development and agglomeration of ZnO–MOp NFs caused by the steric hindrance effect under acidic medium [21].

On the other hand, after 18 and 24 hours, there was minimal evidence of NFs sedimentation for ZnO–MOp NFs under pH 7 and 8. According to Hamed et al. [22], alkaline conditions ($\text{pH} > 7$) are necessary for the majority of green ZnO nanoparticle synthesis, and these circumstances can aid in the production of ZnO. This could be explained by the presence of more OH ions in an alkaline environment, which will attract more positively charged Zn^{2+} ions to encourage the formation of Zn–O bonds. As the pH value rises, the quantity of H^+ and OH^- ions varies, which has an impact on the shape of ZnO that is generated [23].

After 18 hours, there are no signs of sedimentation in Fig. 3 (b), indicating the visual stability of ZnO–MOI NFs. The ZnO–MOI NFs under pH 7 and 8 were determined to be stable over 24 hours, since there was no alteration in the formation of sediments. The equilibrium of steric and electrostatic stabilizing forces at pH 7 and 8 may be the cause of this occurrence [21]. Meanwhile, the NFs demonstrated a decrease in stability at pH 6, as the ZnO NFs began to aggregate, leading to minimal visible settling after 24 hours. The stability of NFs is critical for constant material characteristics and performance throughout time, hence higher pH (7 and 8) is the best choice for synthesizing nanofluid. Higher pH levels resulted in improved stability and dispersion of NFs in the water due to increased reducing ability [1]. Furthermore, the green synthesis method used in the experimental process to generate ZnO NFs addressed the increased dispersion stability of ZnO with extracted MOI compared to extracted MOp.

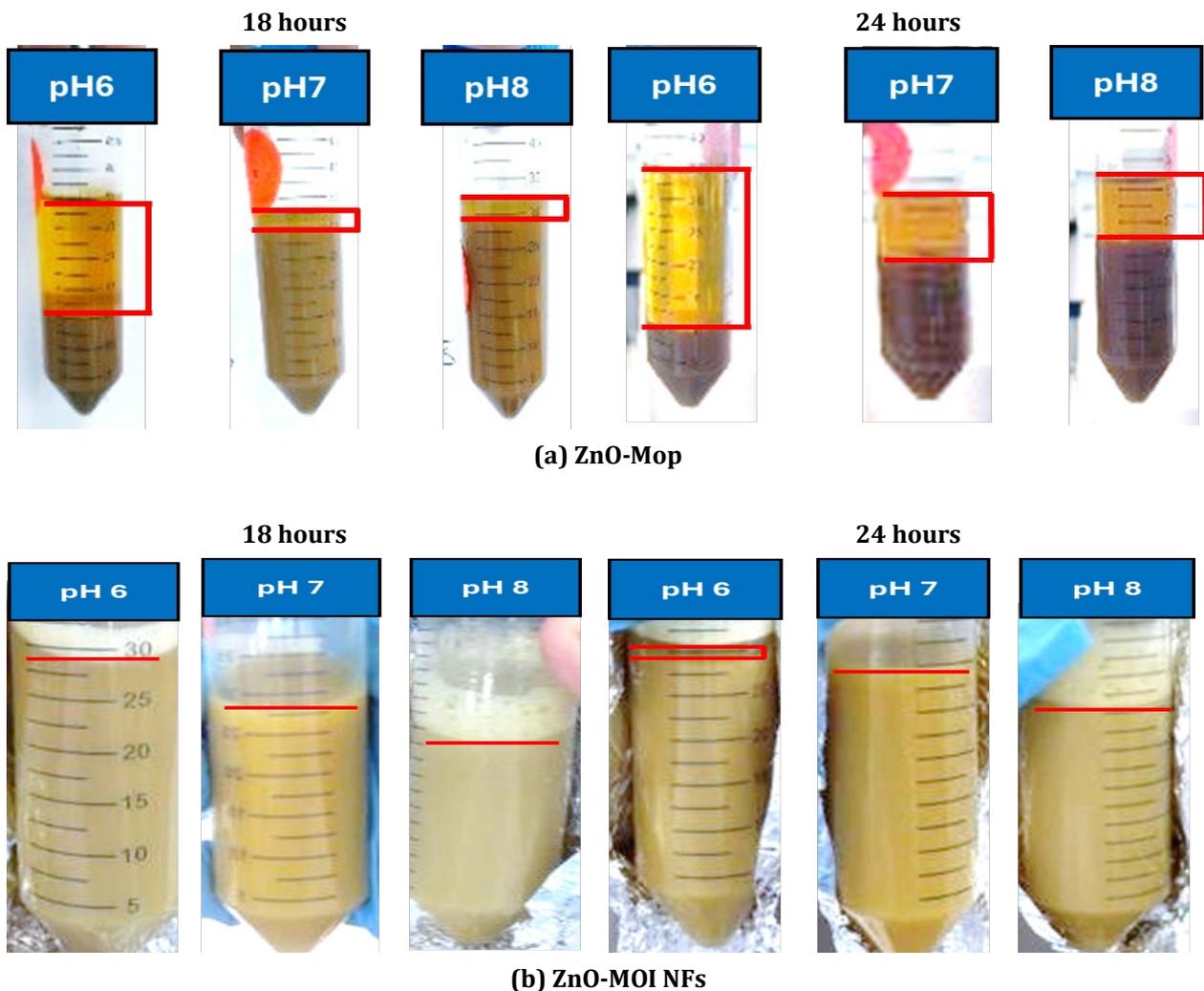


Fig. 3 Stability of a) ZnO–Mop; and b) ZnO–MOI NFs for 18 and 24 hours at varying pH values of the MO extracted solution and a constant ratio of MO extracted with ZnO solution 1:3

According to Al-darwesh et al. [7], UV–visible spectrophotometry can be employed to verify the synthesis of ZnO NFs. Table 1 lists the UV absorption spectra of ZnO–MOp and ZnO–MOI NFs that were produced with varying

extracted MO to ZnO solution ratios and extracted MO pH levels. The absorbance trend decreased as follows: ZnO–MOp 1:9 > ZnO–MOp 1:5 > ZnO–MOp 1:3 > ZnO–MOp 1:1. In contrast, the decreasing trend of absorbance for ZnO–MOI was in the following order: ZnO–MOI 1:9 > ZnO–MOI 1:5 > ZnO–MOI 1:1 > ZnO–MOI 1:3, which is compatible with the sedimentation visual photograph.

Chakraborty and Panigrahi [24] suggest that a UV-VIS spectrophotometer can be used to assess the stability of a nanofluid through investigating the absorption peak attributes. The UV-Vis spectra revealed the greatest absorbance peak at 340 nm, as shown in Table 1, confirming the synthesis of ZnO NFs utilizing MO extracted peel and leaves. This finding is similar to a previous investigation by Pauzi et al. [3], which validated the creation of ZnO in the range of 340 nm–350 nm. The absorbance peak for the production of ZnO NPs was identified in the earlier work in the region of 300–360 nm [25].

The absorbance of ZnO–MOp NFs increased from 4.71 to 6.27 a.u as the extracted MO pH increased from 6 to 8. A similar trend can be seen for the ZnO–MOI NFs effect on the pH of extracted MO, with the greatest absorbance value at pH 8, which corresponds to the stability result of visual settling of the NFs sample. Similar findings demonstrate increased absorbance for ZnO production utilizing plant extract ranging from (2–10 a.u) depending on the specific conditions [20]. In accordance with Ramteke et al. [6], higher absorbance values indicate more homogeneous and stable particles. An increase in absorbance signifies the stability of the nanofluid suspension since absorbance and particle concentration in the solution are proportionate [1]. Furthermore, the strength of the absorbance peaks can reveal information on the size and concentration of the NFs; larger or more concentrated NFs are indicated by higher absorbance. The optical characteristics of optoelectronic NFs and nanomaterials can be properly characterized using UV-Vis spectroscopy. This makes it possible to identify the visual nanoparticles suspended in the liquid phase, as well as their relative concentration and the behavior of electronic transitions [26]. According to this discovery, extracted MOI is preferred over ZnO–MOp NFs due to its stability.

Table 1 *Uv-vis spectra of ZnO -MOp and ZnO–MOI NFs synthesis under different parameters*

Parameter	Absorbance (a.u.) (Wavelength 340 nm)	
	ZnO–MOp	ZnO–MOI
Different ratio of extracted MO with ZnO solution		
1:1	4.60	8.71
1:3	4.76	7.53
1:5	4.80	8.93
1:9	5.36	9.01
Different pH of extracted MO		
6	4.71	8.56
7	5.33	8.57
8	6.27	8.70

3.2 FTIR Analysis of ZnO–MOp and ZnO–MOI NFs

The FTIR measurement was carried out at the ambient temperature for wavelengths ranging from 400 to 4000 cm^{-1} in order to comprehend the functional groups of ZnO–MOp and ZnO–MOI NFs. The findings show that several identical functional groups have identical signals at varying intensities, with no substantial variance in the peak positions of the ZnO, ZnO–MOp, and ZnO–MOI NFs under various extracted MO to ZnO solution ratios and extracted MO pH levels, as shown in Fig.4. This pattern of behavior can be attributed to the lack of substantial chemical interaction between the extracted MO structures and the loaded metal oxide NFs in the examined ZnO–MOp and ZnO–MOI NFs [20].

The broad absorption peak at 3411–3463 cm^{-1} corresponds to the bending vibration of a hydroxyl (O–H) group of surface adsorbed water molecules [27]. This O–H group is commonly present in phenols or alcohols, which may result from the extraction of MO, as well as the use of sodium hydroxide (NaOH) during the synthesis of ZnO–MO NFs. Subsequently, a medium peak with a strong stretch at 1600–1650 cm^{-1} was discovered, confirming C=C bending of the alkene [28].

The functional group discovered for the peak near 1390–1414 cm^{-1} can be attributed to the stretching of C–O caused by leftover organic molecules from the plant extract which cover the NFs during the production step [20]. Additionally, there are intense peaks in the 585–590 cm^{-1} region, suggesting the presence of functional groups linked to the Zn–O stretching and similar to the bands described in earlier research by Aldeen et al. [8]. Bin Fazal and Ahmad [20] discovered similar peaks in their ZnO biosynthesis employing okra seed and amla extract. The FTIR study revealed the presence of alcohols, alkenes, and Zn–O in the produced ZnO–MO NFs.

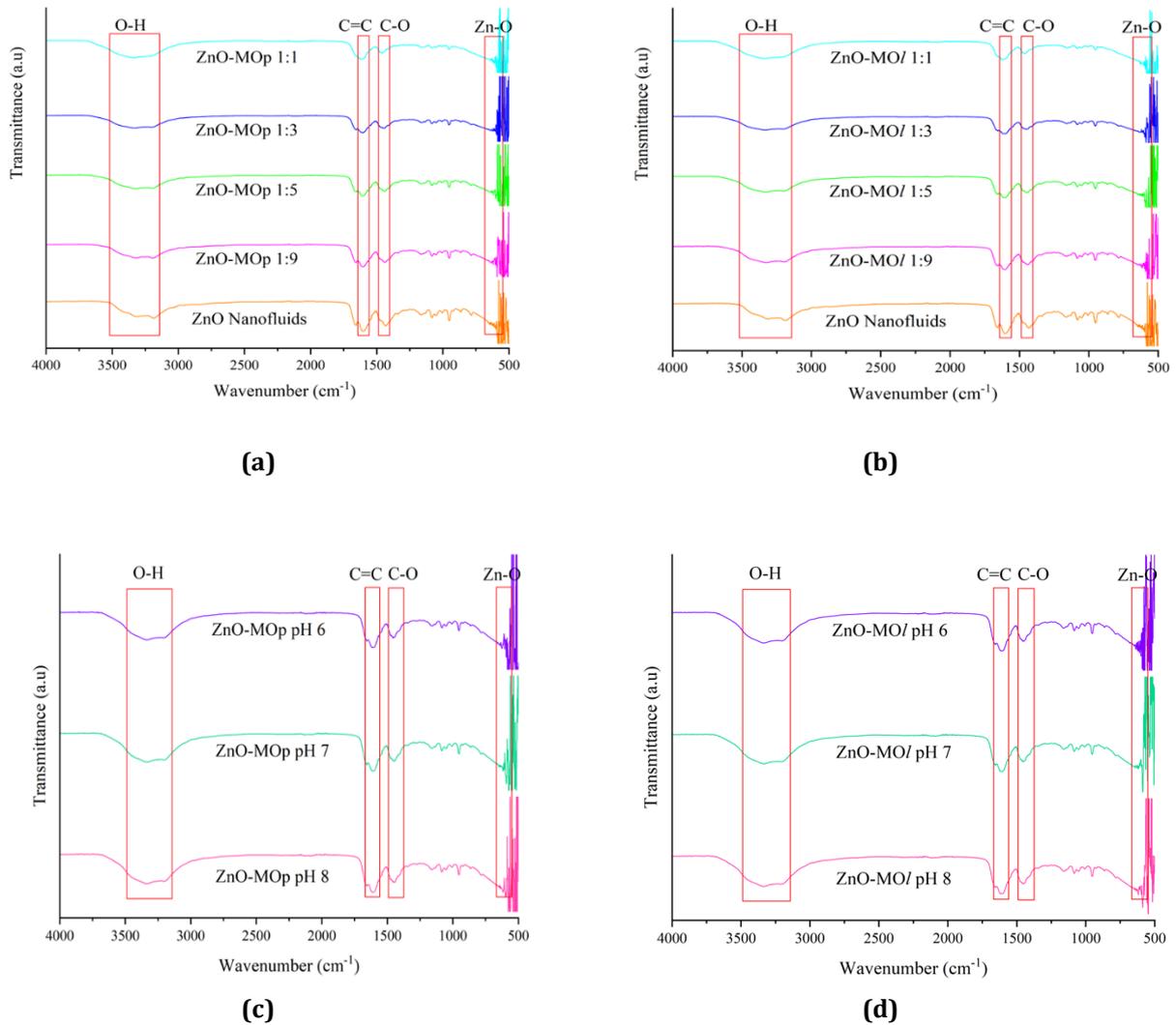


Fig. 4 FTIR spectra of green synthesis ZnO–MO NFs under different: a) ratio of extracted MOp with ZnO solution; b) ratio of extracted MOI with ZnO solution; c) pH of MOp extracted solution; and d) pH of MOI extracted solution

4. Conclusion

The work successfully achieves its goal of developing an environmentally sustainable method for producing ZnO NFs using leaf extract from MO peel and leaves. Owing to their unique characteristics, the MO extract has the potential to effectively stabilize ZnO NFs, resulting in consistent particle dispersion. This study examined the stability of these NFs under a variety of circumstances, including MO extract to ZnO ratios (1:1, 1:3, 1:5, and 1:9) and MO extract pH levels (6, 7, and 8). Additionally, the study demonstrates that ZnO–MOp and ZnO–MOI NFs are most stable at pH 8. Furthermore, for both varieties of ZnO–MOp and ZnO–MOI NFs, the optimal stability is reached at MO extract with a ratio of 1:9. All samples were thoroughly analyzed using FTIR and UV-Vis spectroscopy to validate the successful creation of ZnO–MOp and ZnO–MOI NFs, which display unique chemical bonds and functional groups of ZnO nanoparticles. The optimal formulation from this study is critical for increasing stability and preventing particle aggregation. This study successfully met its aims by investigating the structure and surface properties of ZnO NFs produced from MO peel and leaf extract. The findings emphasize how crucial MO extract pH levels and ZnO ratios are for maximizing the potential of MO phenolic compounds as natural stabilizing or capping agents, which is required to maintain the stability and usefulness of NFs. More investigation might be required to enhance the characteristics of ZnO–MOI NFs as a promising photocatalyst for palm oil secondary effluent treatment. The findings of this current study will serve as the foundation for future study.

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Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: **study conception and design:** Dilaeleyana Abu Bakar Sidik1, Nur Hanis Hayati Hairom; **data collection:** Muhammad Zarif Daniel, Muhammad Faez; **analysis and interpretation of results:** Muhammad Sufyan Naim, Amy Farhana Afiqah Azemi; **draft manuscript preparation:** Nuraina Syafiqah Md Rosdi, Nur Atiqah Syuhada Azuan. All authors reviewed the results and approved the final version of the manuscript.

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