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Long-term stability of novel surface-modified Fe_3O_4 nanoparticles used for preparing water based nanofluids

Su bazlı nanoakışkanların hazırlanmasında kullanılan yeni yüzey modifiye Fe_3O_4 nanopartiküllerinin uzun vadeli kararlılıklarının incelenmesi

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Long-Term Stability of Novel Surface-Modified Fe_3O_4 Nanoparticles Used for Preparing Water Based Nanofluids

Highlights

- *Modified nanoparticles were synthesized.*
- *Water based nanofluids were prepared using surface-modified Fe_3O_4 nanoparticles*
- *Long-term stability of novel nanofluids was analyzed.*
- *$Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] / Water nanofluid was prepared and proposed as stable nanofluid.*

Graphical Abstract

In this study, novel surface-modified fe_3o_4 nanoparticles were used for preparing water based stable nanofluids. Nanofluid preparation process has been demonstrated in following figure.

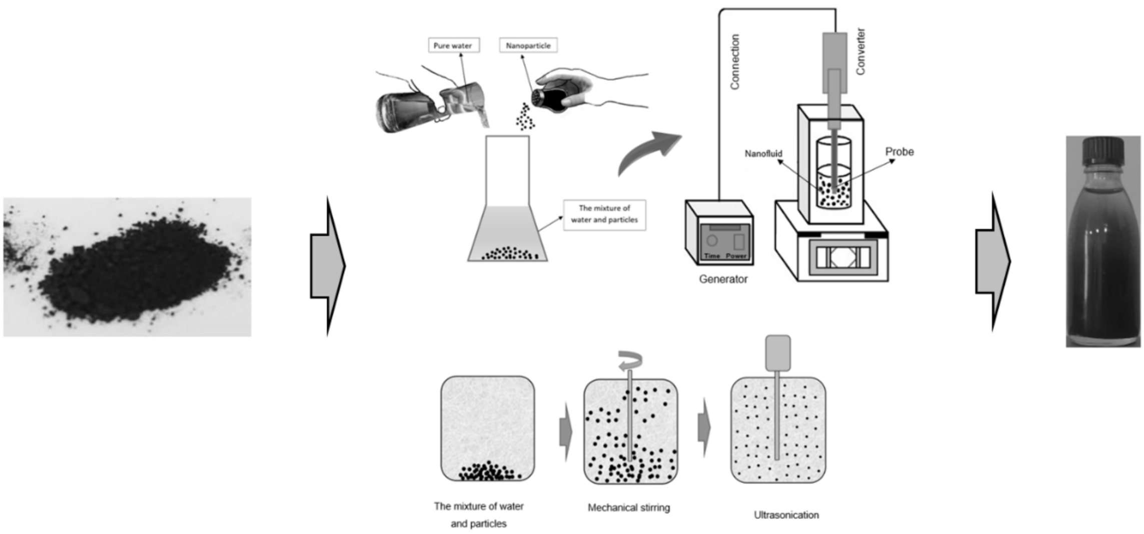

Figure. Nanofluid preparation process using novel nanoparticles

Aim

In this study, it was aimed to prepare nanofluids with high stability to use in heat exchangers as heat transfer fluid.

Design & Methodology

Design and methodology have been performed in two stages. Initially, chemical process was performed and surface-modified Fe_3O_4 nanoparticles were synthesized. In the next step, water based nanofluids were prepared and their stabilities were characterized.

Originality

The originality of this present study is preparing highly stable nanofluids which can be used as heat transfer fluid.

Findings

Stable nanofluids can be prepared by applying surface-modification method of nanoparticles.

Conclusion

Among prepared nanofluids, $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] / Water nanofluid showed the best stability performance after 3 months.

Declaration of Ethical Standards

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

Su Bazlı Nanoakışkanların Hazırlanmasında Kullanılan Yeni Yüzey Modifiye Fe_3O_4 Nanopartiküllerinin Uzun Vadeli Kararlılıklarının İncelenmesi

Araştırma Makalesi / Research Article

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ÖZ

Nanoakışkanlar, farklı katı nano boyutlu materyallerin (metal veya metal olmayan) bir baz sıvı içinde süspanse edilmesiyle üretilir ve genellikle enerji sistemlerinde termal performansı ve ısı transfer hızını artırmak için kullanılır. Isı transferi uygulamalarında kullanılan nanoakışkanlarda gözlenen temel problem kararsızlıklarıdır. Araştırmacılar tarafından kararlı nanoakışkanlar elde etmek için bazı çözümler geliştirilmiş ve önerilmiştir. En önemli çözümlerden biri nanopartikül yüzey modifikasyon yöntemidir. Bu çalışmada Fe_3O_4 nanopartikülleri kimyasal işlemlere tabi tutulmuş ve yüzeyleri modifiye edilmiştir. $Fe_3O_4@SiO_2@Si(CH_2)_3$-IM [Cl], $Fe_3O_4@Si(CH_2)_3$-IM [Cl], ve $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] nanopartiküller olmak üzere üç farklı modifiye nanopartikül sentezlenmiştir. Üretilen partiküller arıtılmış su olan baz akışkan içerisinde kullanılarak %0,2 hacim oranında nanoakışkanlar hazırlanmış ve nanoakışkanın kararlılığı 3 ay boyunca gözlemlenmiştir. Homojen nanoakışkanlar elde etmek amacıyla nanoakışkanlar 3.5 saat ultrasonikasyona tabi tutulmuştur. Su bazlı modifikasyonsuz Fe_3O_4 nanoakışkanı yaklaşık 1 hafta içinde tamamen çökmesi gözlemlenmiştir. Modifiye edilmiş nanoakışkanlarda ise, çökelme meydana gelmesine rağmen 3 ay sonra bile partiküllerin belirli bir miktarının süspanse kaldığı gözlemlenmiştir. Bu çalışmada, Taramalı Elektron Mikroskobu, X-Işını Kırınımı ve İletim Elektron Mikroskobu gibi önemli analiz yöntemleri kullanılmıştır.

Anahtar Kelimeler: Nanoakışkan, yüzey modifikasyonlu nanopartiküller, kararlılık, Fe_3O_4, sedimantasyon, yığılma.

Long-Term Stability of Novel Surface-Modified Fe_3O_4 Nanoparticles Used for Preparing Water Based Nanofluids

ABSTRACT

Nanofluids are produced by suspending different solid nano-size materials (metal and nonmetal) in a base liquid and are often used in energy systems to increase thermal performance and heat transfer rate. The main problem observed in nanofluids used in heat transfer applications is their instability. Researchers have developed and proposed some solutions to obtain stable nanofluids. One of the most important solutions, is the nanoparticles surface modification method. In this work, Fe_3O_4 nanoparticles were subjected to chemical processes and their surfaces were modified. Three different modified nanoparticles were synthesized, which are $Fe_3O_4@SiO_2@Si(CH_2)_3$-IM [Cl], $Fe_3O_4@Si(CH_2)_3$-IM [Cl], and $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] nanoparticles. The nanofluids were prepared in 0.2% Vol. fraction by using the produced particles in base fluid which was distilled water, and stability of nanofluids were observed for 3 months. Nanofluids were subjected to ultrasonication for 3.5 h to obtain homogeneous nanofluid. Not modified water-based Fe_3O_4 nanofluid completely collapsed in approximately 1 week. In modified nanofluids, although sedimentation occurred, it was observed that a certain amount of the particles remained suspended even after 3 months. The most important analyses in this study are Scanning Electron Microscope, X-Ray Diffraction, and Transmission Electron Microscope.

Keywords: Nanofluid, surface-modified nanoparticles, stability, Fe_3O_4, sedimentation, agglomeration.

1. INTRODUCTION

The nanofluids obtained by dispersing Nano-sized solids in the liquid are used to increase thermal performance in heat transfer applications. In the last few decades, many researchers have studied the heat transfer performance of different nanofluids. Compared to the conventional heat transfer fluids, nanofluids have higher thermal conductivity, making the use of nanofluids advantage in heat transfer applications [1,2]. However, sedimentation and agglomerations occur in nanofluids over time. The solids settle to the bottom because their density is higher than liquids. Another problem is the formation of agglomerations as a result of Van der Waals interactions between nanoparticles. Many methods have been proposed to obtain more homogeneous dispersions and to keep the produced nanofluid stable for a longer time. Some of these methods are ultrasonic vibration, pH control of suspension, and use of surfactant [3,4]. In studies evaluating the stability of nanofluids, the effect of

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ultrasonication duration has been observed and many investigations have proven that as the sonication time increases, the clusters formed by the nanoparticles break up and more stable nanofluids are obtained [5-8]. The pH values of nanofluids have a significant effect on their stability. Many studies have been carried out in which the stability of various nanofluids at different pH values has been observed. In these studies, the effects of pH in terms of stability and thermophysical properties have been demonstrated [9-12].

One of the ways to increase the stability of nanofluid is surface modification. Advances in the production and synthesis of nanoparticles have provided the production of nanoparticles with both hydrophobic and hydrophilic surfaces. In a study, polyethylene glycol as a hydrophilic substance and propyl chains as a hydrophobic substance were coated on the surfaces of silica nanoparticles. The oil-water interface properties of these obtained nanoparticles were investigated. The determined results showed that SiO_2 nanoparticles coated with both substances reduced the water surface tension and oil-water interfacial tension by 50% [13]. The research was carried out on SiO_2 -coated graphene nanoparticles prepared by chemical liquid deposition method. Water-based nanofluid was obtained with nanomaterials prepared by surface modification, and the stability and thermal conductivity of this obtained nanofluid were examined. In the detected results, it was observed that the stability and thermal conductivity of the modified nanofluid by coating SiO_2 increased significantly [14]. In the study carried out with aluminum oxide nanoparticles, their surfaces were modified with phosphonic acid-based oligomers of aromatic polyester, polyether, and polydimethylsiloxane. In the evaluations made, it was reported that the best results in terms of stability were obtained in the bis-phosphonic polydimethylsiloxane-based formulation [15]. The effect of the modification process on the stability of the suspension was investigated in the study with multi-walled carbon nanotubes prepared by using different surface modification methods (amide, surfactant, base, sulfate, acid). The produced nanotubes were dispersed in water and the stability of the nanofluid was observed by UV-visible spectral analysis. According to the UV results, the best stability exhibited firstly the acid mixture and then potassium persulfate [16]. In the study, which was performed by producing silver nanoparticles with an average diameter of 5 nm, oleic acid was utilized to coat the surface of the particles [17]. The study was conducted by making surface modification of carbon nanotubes with β-Cyclodextrin (β-CD). In this conducted study, carbon nanotubes were demonstrated to be stable up to 5% wt. concentration in distilled water. In addition, the two-step nanofluid preparation method was shown in the study and it was emphasized that it could be quite successful when used with surface modification [18]. The research was carried out by surface modification on TiO_2 nanoparticles. The stability of the nanoparticles dispersed in water was assessed and it was stated that the nanofluids obtained by adding Tetramethylammonium hydroxide and 3-aminepropyltriethoxysilane were more stable. However, the viscosity of the nanofluid treated with 3-aminepropyltriethoxysilane was higher than the nanofluid obtained from Tetramethylammonium hydroxide [19]. The multi-walled carbon nanotube was utilized in the experimental study performed to examine the heat transfer efficiency of nanofluids. In this study, surface modification was made by polymer wrapping process on nanotubes to obtain more stable and homogeneous nanofluid [20]. In the field of heat transfer and thermodynamics the efficient heat transfer is known as one of the first-order issues [21,22], which has been widely investigated in the literature. Currently, high technology heat exchangers such as plate heat exchangers (PHE) are proposed to enhance heat transfer and to solve this problem by using different types of nanofluids [23-27]. As an example a schematic of the experimental setup proposed in literature was presented in Figure 1.

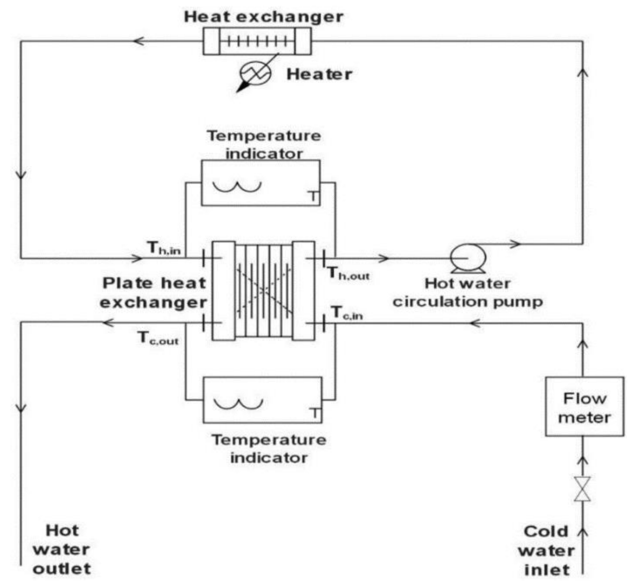


Figure 1. Experimental setup used for heat transfer analyses of nanofluids [25]

2. MATERIAL and METHOD

2.1 preparation of nanoparticle

The bare Fe_3O_4 NPs were synthesized according to co-precipitation method [28]. $FeCl_2$. $4H_2O$ and $FeCl_3$ (1:2) were heated to 80 °C in deionized water. In the next step, 1.3 mL NH_4OH was added into the stirred solution. Finally, the precipitates were collected by magnet and washed three times by DI water. The final solid NPs were obtained after drying in oven at 60 °C.

2.1.1. Synthesis of Fe_3O_4@SiO_2 (F1)

The Fe_3O_4@SiO_2 were synthesized according to previously synthesized method [29-31]. 1g of Fe_3O_4@SiO_2 were dispersed in toluene and 2 mL (3-chloropropyl) trimethoxysilane was added to mechanically stirred solution. After for 24 h, 2 mL N-methylimidazole was added to mixture and mechanically stirred for 48 h. The $Fe_3O_4@SiO_2@Si(CH_2)_3$-IM [Cl] was separated by magnet, and oven dried [32-34].

2.1.2. Synthesis of $Fe_3O_4@Si(CH_2)_3$-IM [Cl] (F2)

1g Fe_3O_4 were dispersed in toluene and 2 mL (3-chloropropyl) trimethoxysilane was added to mechanically stirred solution. After for 24 h, 2 mL N-methylimidazole was added to mixture and mechanically stirred for 48 h. The $Fe_3O_4@Si(CH_2)_3$-IM [Cl] was separated by magnet, and oven dried.

2.1.3. Preparation of $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] (F3)

1g Fe_3O_4 were dispersed in toluene, 2 ml TEOS and 2 mL (3-chloropropyl) trimethoxysilane was added to mechanically stirred solution. After for 24 h, 2 mL N-methylimidazole was added to mixture and mechanically stirred for 48 h. The $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] was separated by magnet, and oven dried.

Figure 2 shows the synthetic procedure for three synthesized nanoparticles.

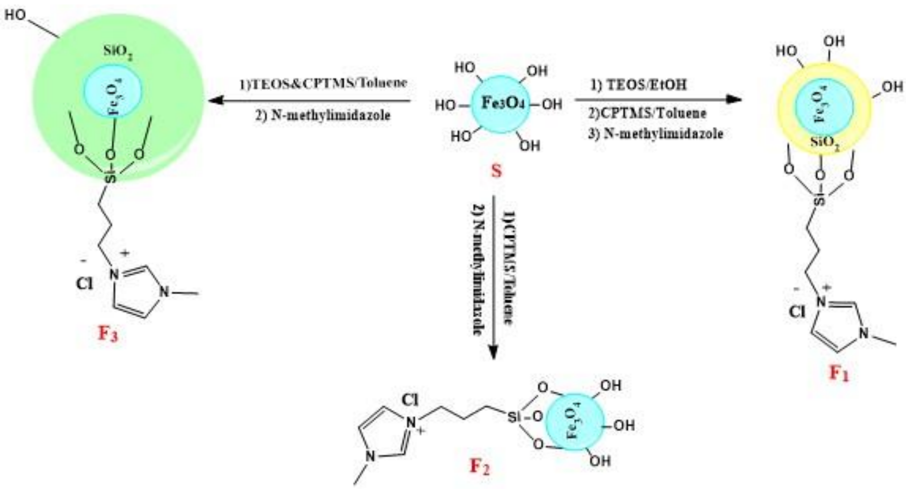
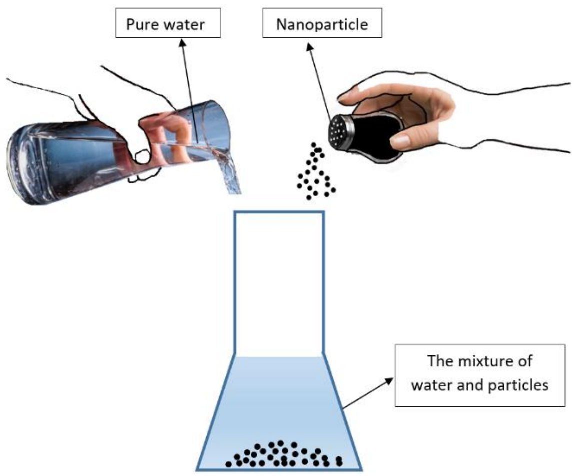


Figure 2. Synthetic procedure for nanoparticles.

2.2 Preparation of nanofluid

The nanoparticle agglomerates that occurred in the nanofluid employed in energy systems and heat exchangers [36-39] significantly affect total thermal performance. Ultrasonication of the nanofluid is a common method used to break the agglomerations and obtain a homogeneous mixture. The most significant parameter in the mentioned method is the exposure time of the nanofluid to ultrasonication. The effects of ultrasonication Time on the stability, thermal conductivity and viscosity of the nanofluid were widely investigated. It has been reported that thermal conductivity increased with the ultrasonication time. Additionally, the optimum ultrasonication should be considered to achieve the maximum thermal conduction and heat transfer values [40-41]. In this context, sonication times between 10 and 160 min were examined and the stability of nanofluids was compared. It can be stated that, as the sonication time increased, the nanoparticle clusters in the base fluid were broken and a more homogeneous nanofluids can be obtained [42-43]. Amounts of pure water and nanoparticles required for 50 mL nanofluid with a volume ratio of 0.2% were calculated. Hielscher UP400S ultrasonic homogenizer was used to stir synthesized nanoparticles into water uniformly (operated with a power of 200 W at 10 kHz for 3.5 h). In Figure 3, the process of nanofluid preparation has been presented.



3. RESULTS of ANALYZES and DISCUSSION

The Scanning Electron Microscope (SEM) image of Fe_3O_4, which is used as a pure material for the modification application, was taken and the diameter of the nanoparticles was obtained. According to these results, the dimensions of the nanoparticles used were in the range of 9-14nm, as seen in figure 4.

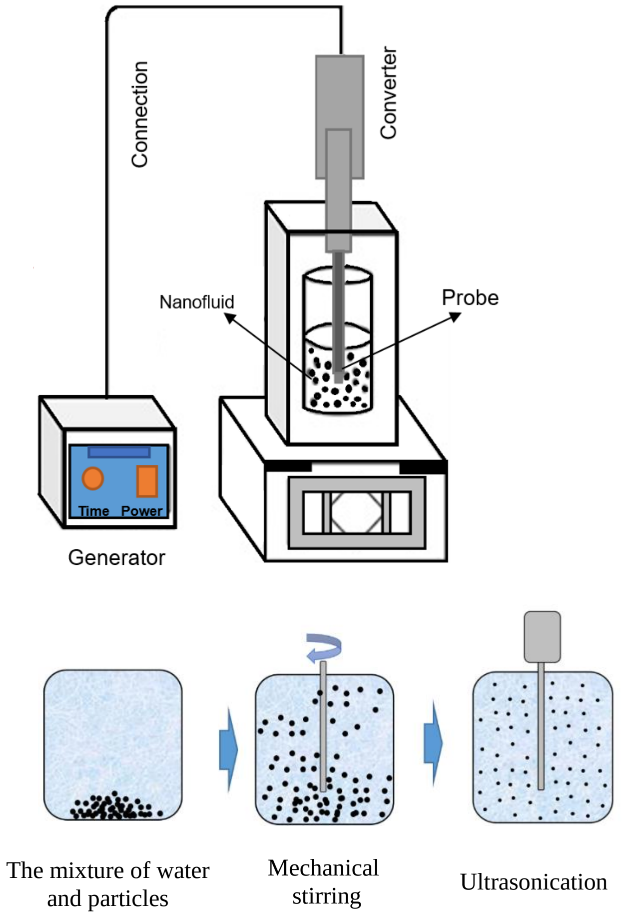


Figure 3. Nanofluid preparation process mechanical stirring

Energy-Dispersive X-Ray Spectroscopy (EDS) results of the modified nanofluid samples were obtained as shown in figure 5. According to the results, long-term suspension and higher stability can be observed in samples 1 and 3 after 3 months. On the other hand, in sample 2 (F2), complete precipitation occurred and the nanoparticles were completely settled in the liquid. The reason why aluminum metal appears in the EDS results is the presence of aluminum metal on the test plate of the EDS device. The occurrence of iron (Fe) particles in F1 and F3 samples was demonstrated using circle dash lines

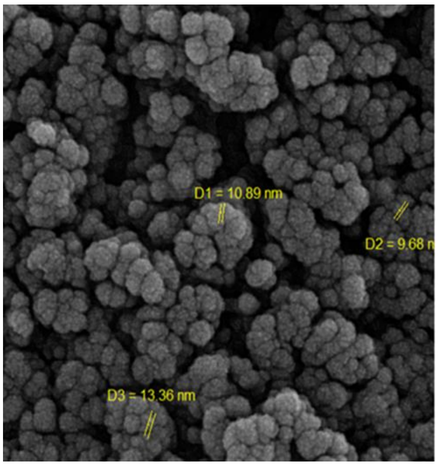
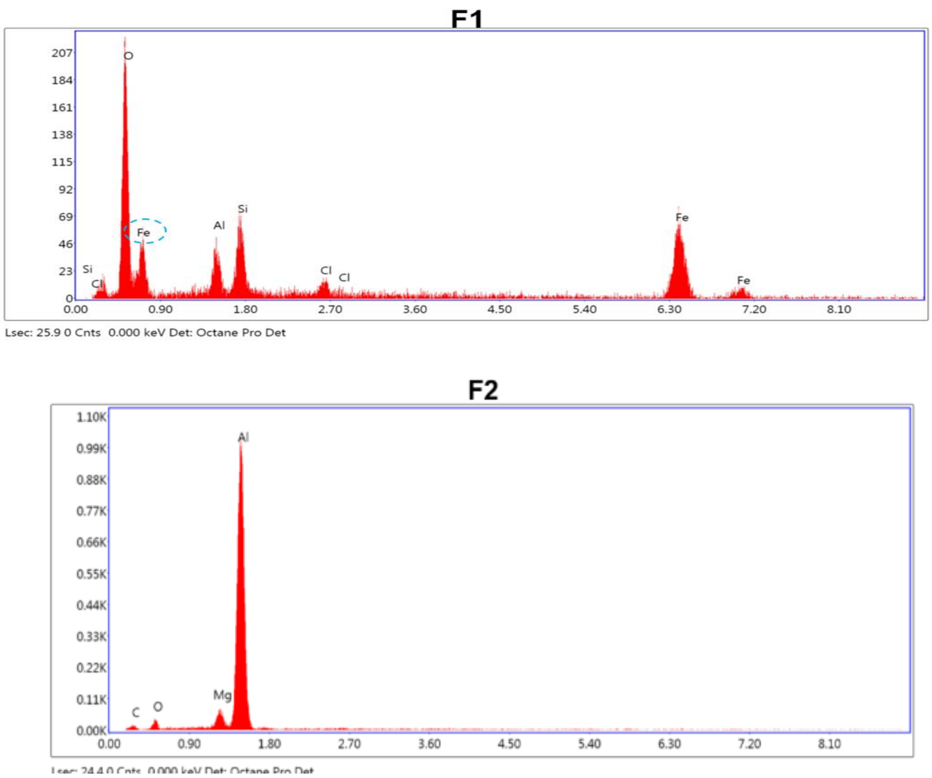


Figure 4. Dimensions of nanoparticles in SEM analysis images



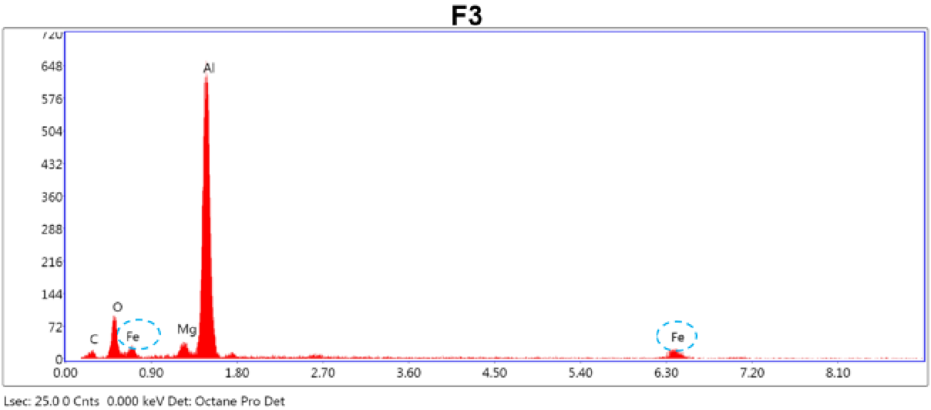


Figure 5. Energy-Dispersive X-Ray Spectroscopy results after 90 days

The agglomeration of nanoparticles decreases the stability of prepared nanofluids and this issue affects their technologically advantages and desired properties. The sedimentation of nanofluids in base fluid is illustrated in following figure, which was observed for the sample Fe_3O_4@SiO_2-mix-$(CH_2)_3$Cl@Imidazol (F_3)/water after 3 months (Figure 6). Optical microscope images of all three samples prepared were obtained after three months as demonstrated in figure 7. As can be seen from the microscope images in the F1/Water sample, a low amount of the nanoparticles remained suspended in the liquid. However, approximately no nanoparticle images were found in the F2/Water sample. This prepared sample lost its nanofluid properties at the end of 3 months and completely turned into a base liquid.

In the F3/Water sample, agglomeration occurred between nanoparticles and its homogeneity decreased compared to its original state, but this sample preserved its stability better than the other samples. According to these results, the nanofluid obtained from the F3 nanoparticle gave best results compared with samples F1 and F2.

Figure 6. Sedimentation of the prepared sample F3/water during time

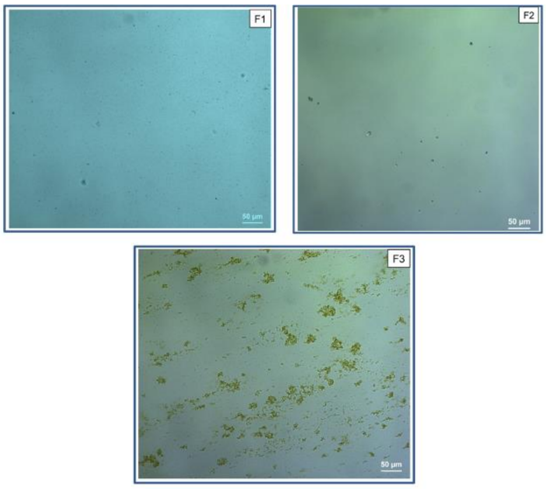


Figure 7. Optical microscope results after 90 days

Figures 8 and 9 show Transmission Electron Microscope (TEM) analyses results with 2 µm and 50 nm scales, respectively for three different nanofluid samples named F1/water, F2/water and F3/water. If the image of the F1 sample is examined, dense particle aggregation and inhomogeneous regions can be observed in different regions of nanofluid. It can be seen that the particle density for the F2/Water sample is quite low and the stability is notably poor. Likewise, in the light of the images obtained from these analyzes, it is clearly seen that the most homogeneous particle distribution and the lowest agglomeration problems are in the F3/Water sample.

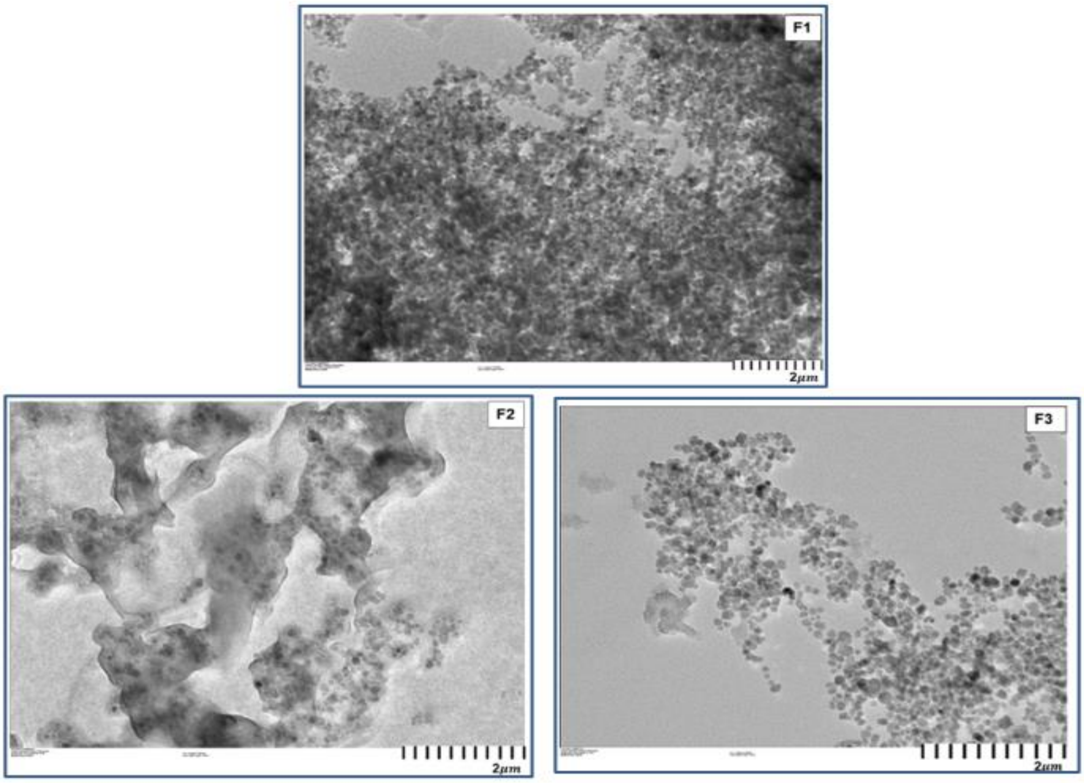


Figure 8. TEM results after 90 days (at 2μm scale)

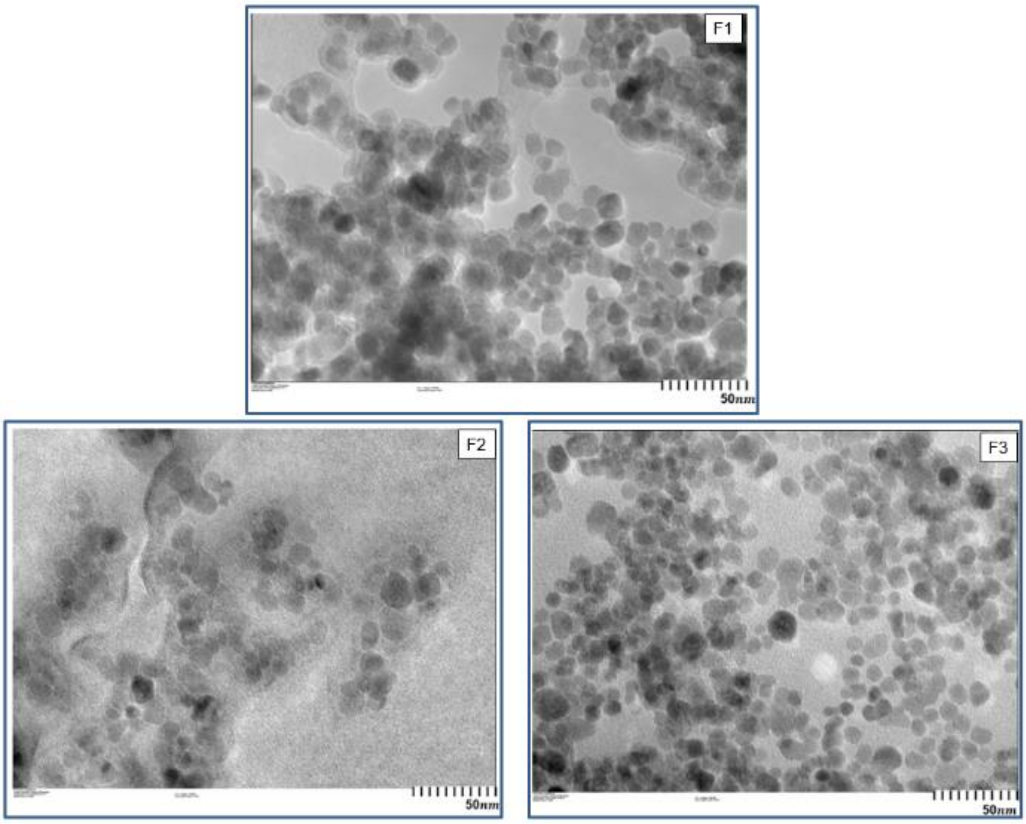


Figure 9. TEM results after 90 days (at 50 nm scale)

6. CONCLUSION

In this work, novel nanoparticles were prepared using a novel surface modification method and the obtained water based nanofluids were investigated in term of stability. Fe_3O_4 nanoparticles were utilized and surface modifications were performed using several chemical methods. With the aim comparison stability of prepared nanofluids samples, SEM, TEM, XRD and optical microscope analyses were performed. As a main conclusion it was observed that, $Fe_3O_4@SiO_2\&Si(CH_2)_3$-IM [Cl] / Water nanofluid showed the best stability performance after 3 months. Stability problems are frequently mentioned in the literature, especially for nanofluids prepared by Fe_3O_4. As a result of this present research, synthesis of more stable nanofluids for industrial applications is possible using advanced processes such as surface modification. However, hybrid methods can also be applied using methods such as using surfactant and long period ultrasonication to increase stability.

ACKNOWLEDGEMENT

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DECLARATION OF ETHICAL STANDARDS

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

AUTHORS' CONTRIBUTIONS

Faraz Afshari, Burak Muratçobanoğlu, Emre Mandev, Bayram Şahin, Eyüphan Manay: Performed analyses of the nanofluids.

Shabnam Rahimpour, Reza Teimuri-Mofrad: Performed the synthesis of nanoparticles.

Burak Muratçobanoğlu, Emre Mandev, Shabnam Rahimpour, Faraz Afshari: Wrote the manuscript.

CONFLICT OF INTEREST

There is no conflict of interest in this study.

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