Structural, Optical and Magnetic Properties of α -Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂ Composites Produced by a Facile Method

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We prepared SiO₂, Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ composites by an enhanced method and reported the result of their structural, optical and magnetic properties. In the x-ray diffraction results of the Fe₂O₃-SiO₂, Fe₂O₃ and the SiO₂ it is evident that these composites are crystallized in rhombohedral and trigonal structures, respectively. In the Dy₂O₃-SiO₂ composite, SiO₂ transforms into a trigonal structure with the addition of Dy. The absorption bands belong to Fe₂O₃, and Dy₂O₃ were obtained using the Fourier transform infrared spectra. In ultraviolet–visible spectra, the photocatalytic properties of Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂ were determined as a function of time at room temperature. Maximum transmittance change at 800 nm was 75% and 40% for composites Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂, respectively. The photocatalytic property of Dy₂O₃-SiO₂ composite increases gradually from short to long in the wavelength region where it exhibits a maximum value in the visible region. In magnetic measurements, a weak ferromagnetic behavior was observed in the Fe₂O₃-SiO₂, while Dy₂O₃-SiO₂ exhibited paramagnetic behavior as expected. The saturation and coercivity values for Fe₂O₃-SiO₂ were found to be 0.15 Am²kg⁻¹ and 40 mT, respectively.

Key words: Silica, Fe₂O₃-SiO₂, Dy₂O₃-SiO₂, optical properties

INTRODUCTION

Composite materials are widely used in various applications in industry such as filtration in wastewater treatment,¹ biotechnology/biomedicine,^{2,3} magnetic paper,⁴ and catalysis.^{5,6} Micro- and nano-scale silica and silicon dioxide particles are important for these composite materials because of the ease of preparation and ability to control the size, a high surface-to-volume ratio, and biocompatibility.² As an example, silicon dioxide is used with photocatalytic materials. In the presence of light, the catalytic property of a material is referred by the photocatalyst property, and here the photocatalyst provides an increase in the rate of chemical

radiation by activating a catalyst by ultraviolet (UV) or visible light.⁷ Here, the light causes electrons and holes in the photocatalytic reaction. The catalysts are semiconductor materials such as TiO₂ and Fe₂O₃, so an electron transfers from the valance band to the conduction band with the absorption of the light.

To develop new composite materials, many studies are carried out.^{8–17} There are many techniques to improve the unique properties of the materials such as surface modification, formation of composites, doping with different materials such as rare earth metals, and different production methods.⁷ Synthesis and investigation of these new materials are still interesting for researchers. Zinatloo-Ajabshir et al.¹⁸ presented the Dy₂Sn₂O₇-SnO₂ with a simple, cost-effective, convenient, and eco-friendly method and compared with SnO₂. They found better photocatalytic performance in the Dy₂Sn₂O₇-SnO₂

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than in SnO₂ alone. Mahdiani et al.¹⁹ investigated a new nanostructure, CuFe₁₂O₁₉/CNT (carbon nanotube) and CuFe₁₂O₁₉/graphene and reported that the graphene-based nanocomposites led to increasing the photocatalytic activity under UV irradiation. The relation between the photocatalytic materials and the materials with large surface area and high porosity such as SiO₂ and graphene is essential to increase the photocatalytic property.^{1,19}

Fe₂O₃ nano- and microparticles are of interest for various applications in industry as catalysts, gas sensors, pigments, photo-anodes in photo-electrochemical cells or contrast agents in magnetic resonance imaging because of their hardness, catalytic activity, and surface resistivity, as well as magnetic, optical, and electronic properties.^{5,6} Because of these unique properties, Fe₂O₃ has been combined to produce a composite structure with SiO₂ that has a large surface area and high porosity particles.^{20,21} Bogatrev et al.²¹ investigated the Fe₂O₃/SiO₂ nanocomposite produced by chemical reaction, and they showed that the reaction and treatment conditions allow the production of this nanocomposite of a high specific surface area that can be of importance for practical applications. Balbuena et al.²⁰ reported the α -Fe₂O₃/SiO₂ composites for enhanced photocatalytic NO oxidation. In the studies of Fe or Fe₂O₃/TiO₂ composites, Fe⁴⁺ ions are reduced to Fe³⁺ by the photochemical reactions. Therefore, the composites absorb the photons, according to transformation of all Fe⁴⁺ to Fe³⁺ ions.^{22,23} The rare earth element, Dy³⁺ ion, is identified as an active luminescence center and also as *f*-localized trap-creating ion.^{24,25} When the *d* orbitals of rare earth elements are excited by the UV light, the *d*–*f* transition occurs because of the absorption of photons. As a result, the rare earth elements emit the light to return the initial state. The rare earth elements are generally used in various fields such as in preparing light-emitting diodes, optical fibers, amplifiers and biomedicine for its optical characteristics and its luminescence emission capabilities. Therefore, this property can be utilized for the preparation of a photocatalyst by rare earth or rare earth oxides onto oxides such as SiO₂ and TiO₂. In the previous studies,^{26–28} Dy₂O₃ has been doped on ZnO and studied for various applications other than photocatalysis induced by UV light irradiation.

In this study, we prepared the α -Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂ composites by a facile method. In the preparation of the composites, various methods are used, such as chemical reaction and sol-gel. The preparation of these is difficult, and the contamination risk is high. As an example, sol-gel method is used in the most of study; however, this technique cannot achieve its full industrial potential due to some limitations such high permeability, weak bonding, and hard porosity control.²⁹ The sol-gel process uses several steps such as mixing and

casting, and many chemical reactions take place in the formation of metal alkoxide particles to a composite in these steps. Also, the residual precursor must be burned to remove the organic materials produced in the chemical reactions, and the burning temperature plays a crucial role in the purity of the final composite. Lack of scientific understanding of these complex reactions is a major drawback of the sol-gel technique.^{29,30} The morphological and structural analysis was performed by x-ray diffraction (XRD) and scanning electron microscopy (SEM) at room temperature. The optical properties were measured by Fourier transform infrared (FTIR) spectrometry in the range of 400–4000 cm^{−1} and ultraviolet–visible (UV-Vis) spectrometry between 200 nm and 800 nm. Also, the magnetic behavior of these composites was investigated by hysteresis curves under 2 T at room temperature.

EXPERIMENTAL

Synthesis of Composites

SiO₂, Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ composites were prepared with a heat-treatment process by using a high-temperature atmosphere oven. Five grams of pure Si powder (99.99%) was placed in an atmospheric oven with an alumina combustion boat and heated for 5 days at 1673 K. The powder was manually mixed once a day. The color of the powder changed from gray to blue throughout the process.

For Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂ composites, pure Fe (99.9%) and pure Dy (99.9%) powder were separately mixed with SiO₂ powders produced in the initial process. The mixed ratio is 1:4 (pure Fe:SiO₂ and pure Dy:SiO₂) and total powder weight is 2g for each composite. The same heating (1673 K for 5 days) and mixing processes were applied on prepared samples. All composites were slowly cooled to room temperature in the oven.

Structural, Morphological and Magnetic Characterizations

The structural analyses were performed by x-ray diffraction (XRD). The patterns were collected at room temperature in the range of 20° < 2 θ < 75° using Cu K α radiation. Their crystal lattice and parameters were determined by FullProf and QUALX 2³¹ software. Moreover, the morphological analysis was conducted by scanning electron microscopy with energy dispersive x-ray (SEM-EDX). The field dependencies of the magnetization curves, *M*(*H*), were measured by a vibrating sample magnetometer (Microsense EZ9).

Optical Characterization

The UV-Visible and FT-IR analyses of the powder samples were carried out using a potassium bromide (KBr) pellet. The vibration states of the samples were characterized by FTIR spectra, which

was taken at room temperature using the KBr disc. Spectra in the wave-number range between 400 cm^{-1} and 4000 cm^{-1} were obtained. The dark current noises and background were corrected in the infrared spectrum using two-point baseline correction.

The UV-Visible transmittance spectrum measurements were performed at a wavelength range of 200–800 nm. Cary 5000 UV-Vis-NIR

spectrophotometer was used for recording the spectra with 1 nm resolution.

RESULTS AND DISCUSSION

Figure 1a, b and c shows the refined XRD patterns of SiO_2 , $\text{Fe}_2\text{O}_3\text{-SiO}_2$ and $\text{Dy}_2\text{O}_3\text{-SiO}_2$ composites at room temperature. The pattern of SiO_2 shows

Fig. 1. Rietveld refinement results. Observed (red circles) and calculated (black lines) intensities of XRD patterns (a) SiO_2 (b) $\text{Fe}_2\text{O}_3\text{-SiO}_2$ (c) $\text{Dy}_2\text{O}_3\text{-SiO}_2$. Thin lines in the lower part of each plot show the difference between the calculated and observed patterns. The vertical bars refer to the calculated allowed Bragg reflections. Here, $P32\ 2\ 1$ and $P31\ 2\ 1$ are the trigonal, $Fd-3m$ and $Ia-3$ are the cubic, $R-3cH$ is the rhombohedral and $P41\ 21\ 2$ is the tetragonal (Color figure online).

Table I. Crystallographic data for the structural refinement

Sample	Phases	Crystal system	Space group	$a = b$ (Å)	c (Å)
SiO ₂	SiO ₂ (1)	Trigonal	$P32\ 2\ 1$	4.853	5.332
	SiO ₂ (2)	Trigonal	$P31\ 2\ 1$	4.535	5.204
	Si	Cubic	$Fd - 3m$	5.430	$= a$
Fe ₂ O ₃ -SiO ₂	Fe ₂ O ₃	Rhombohedral	$R - 3cH$	5.032	13.749
	SiO ₂ (1)	Trigonal	$P32\ 2\ 1$	4.847	5.338
	SiO ₂ (2)	Trigonal	$P31\ 2\ 1$	4.539	5.193
	Si	Cubic	$Fd - 3m$	5.427	$= a$
Dy ₂ O ₃ -SiO ₂	Dy ₂ O ₃	Tetragonal	$P41\ 21\ 2$	4.991	6.994
	SiO ₂	Cubic	$Ia - 3$	10.658	$= a$

the mixed phases of SiO₂ (trigonal) and of Si (cubic). Fe doped SiO₂ composite exhibits a similar crystal structure for SiO₂ and a rhombohedral structure related to Fe₂O₃ in Fig. 1b. When Dy is added to SiO₂, the obtained composite includes two phases that correspond to Dy₂O₃ and SiO₂ respectively, as seen in Fig. 1c. Here, the crystal structure of SiO₂ changes from trigonal to cubic due to the addition of Dy content. A similar result has been shown in the study of Vasanthavel et al.³² where the addition of Dy to the ZrO₂-SiO₂, transforms the structure of composition from tetragonal to cubic. The XRD result of the Dy₂O₃-SiO₂ indicates that the SiO₂ crystallizes in the cubic form, and this may be caused by the Dy addition as seen in Ref. 32. Consequently, the obtained crystal structures of the composites prove that the synthesis explained herein can be employed as an alternative preparation procedure. The crystal structures and calculated lattice parameters of the patterns are given in Table I. The results show that the expected composite materials were obtained.

The morphology of SiO₂, Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ were investigated by SEM. The measurements were performed at 15 kV. The SEM images are given in Fig. 2a, b and c. Figure 2a shows the SiO₂ sample, and the micro particles integrated each other with heat treatment. The Fe₂O₃ particles congregate regionally in Fig. 2b and they have a spherical form as seen in the inset of Fig. 2b. In the Dy₂O₃-SiO₂ (Fig. 2c), the particle sizes are close to each other, and the SiO₂ particles form soft-edged structure. The EDX measurements are performed, and the indicated points are shown in the figures. The EDX spectra are also plotted in Fig. 2. In the spectra, the Si, Fe and Dy peaks were clearly determined. The results show that the expected composites were obtained.

FTIR absorption spectral curves of Si, SiO₂, Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ are presented in Fig. 3. The FTIR absorption spectrum of Si sample (black line) has main four absorption bands at around 1380 cm⁻¹, 1637 cm⁻¹, 3446 cm⁻¹ and 3735 cm⁻¹. The peak at 3735 cm⁻¹ is due to the non-hydrogen-bonded silanol. At 1637 cm⁻¹ and 3446 cm⁻¹, the absorption peaks are attributed to stretching modes of the absorbed water molecule.^{33,34} It is known that the symmetric and anti-

symmetric stretching modes of the Si-O-Si bonds are IR active in 800–1300 cm⁻¹ region.^{35–38} In the SiO₂ sample (red line), the additional peaks at 2356 cm⁻¹, 2333 cm⁻¹, 1093 cm⁻¹, 786 cm⁻¹, and 478 cm⁻¹ were observed. The 1380 cm⁻¹, 2356 cm⁻¹ and 2333 cm⁻¹ peaks are attributed to CO₂ in the air.^{39,40} The weak absorption peak at 786 and the medium absorption peak at 478 cm⁻¹ are due to symmetric stretching vibrations and bending vibrations of Si-O-Si, respectively.⁴¹

In the FTIR spectrum of Fe₂O₃-SiO₂ composite (blue line), the new additional peaks were found at 1128 cm⁻¹, 1033 cm⁻¹ and 586 cm⁻¹. The peaks at 1033 cm⁻¹ and 1093 cm⁻¹ partially overlap with each other. The peak at 1128 cm⁻¹ is due to asymmetric stretching bonds of Si-O-Si. The peaks at 1033 cm⁻¹ and 586 cm⁻¹ are an indication of the presence of Si-O-Fe.⁴²

Dy₂O₃-SiO₂ (cyan line) composite exhibits the peaks at 1128 cm⁻¹, 1093 cm⁻¹, 786 cm⁻¹, 613 cm⁻¹ and 516 cm⁻¹ in its FTIR spectrum and here, the peaks at 1128 cm⁻¹, 1093 cm⁻¹, 786 cm⁻¹ appear stronger than the other composites, and this may be caused by the structural change of SiO₂ since SiO₂ undergoes a trigonal to cubic transition with the addition of Dy content. Also, the peaks at 613 cm⁻¹ and 516 cm⁻¹ are due to Dy. The peaks at 586 cm⁻¹ and 1033 cm⁻¹ disappear in this composite. This situation indicates that these peaks are associated with the existence of Fe₂O₃.

UV-Visible absorption spectroscopy is a useful technique to explore the optical properties of semi-conducting micro and nanoparticles.^{43,44} The transmittance measurements of SiO₂, Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ were performed between 200 nm and 800 nm wavelengths.

The transmittance spectrum of SiO₂ at room temperature is given in Fig. 4a. The transmittance slowly increases up to wavelength 500 nm and then rapidly continues to grow until reaching a maximum at 800 nm. Here, the absorbance of the long wavelength in this sample is higher than the short wavelength. The spectra were measured as a function of time and generally, the obtained curves overlapped with each other. Therefore, the other curves are not given in the figure.

Fig. 2. SEM images and EDX spectra of (a) SiO_2 (b) $\text{Fe}_2\text{O}_3\text{-SiO}_2$ (c) $\text{Dy}_2\text{O}_3\text{-SiO}_2$ samples.

UV-Vis measurement of the $\text{Fe}_2\text{O}_3\text{-SiO}_2$ was performed with respect to time, and the variation of its transmittance curves are given in Fig. 4b. The transmittance in the range of 200–425 nm is lower than 20%. Between 425 nm and 800 nm, it decreases from 100% to 25% as a function of time. Here, the absorbance of the sample is increased due to the electrons movements in s and d orbitals of Fe when it is exposed to light. In the Fe_2O_3 , Fe^{3+} ions are a better electron scavenger than O_2 ,²² and therefore, the oxidation of $\text{Fe}^{3+} \rightarrow \text{Fe}^{4+}$ can occur. The photocatalytic property of this composite increases step by step the wavelength increases and it exhibits a maximum in the visible region. The variation amount of the transmittance at 425 and 800 nm are given in Fig. 4c. At 425 nm, the variation is close to zero; however, a remarkable 60% variation occurs within 200 s at 800 nm. Moreover, the $y = y_0 + A \exp(R_0 x)$ fit function was applied to these variations, and parameter A changed from -4.67 to 114.69 .

The transmittance measurements of $\text{Dy}_2\text{O}_3\text{-SiO}_2$ sample are plotted in Fig. 4d. As observed in the $\text{Fe}_2\text{O}_3\text{-SiO}_2$, the photocatalytic property was found, but in this sample, the variation occurred in the range of 425–800 nm. The transmittance in the range of 200–425 nm is about 35%. Between 425 nm and 800 nm, it decreases from 100% to 60% due to time. Fig. 4e shows the change percentage for transmittance at 425 and 800 nm. At 800 nm, the 30% variation occurs within 300 s. When these variations are fitted with the same function, the parameter A changed from 5.37 to 62.31. In the wide range in the UV region, the absorbance of the sample increases with respect to time. Moreover, the absorbance of the short wavelengths in the UV region begins to increase with Dy content. This behavior is caused by d and f orbitals of Dy, and here, the electrons are exposed by the light. Also, Dy^{3+} ions oxidate to Dy^{4+} . The absorbance of the sample increases in between nm 500 and 800 nm.

Fig. 3. The FTIR spectra of Si (black), SiO₂ (red), Fe₂O₃-SiO₂ (blue) and Dy₂O₃-SiO₂ (cyan) (Color figure online).

This increase could be due to the presence of color centers.⁴⁵ On the other hand, the change of absorbance is lower than the Fe doped sample, but the absorbance of the light occurs at the wide range wavelength.

To check the performance of the Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂, the sample was left for 30 min inside the spectrometer in the dark, and a new measurement was performed shown in Fig. 5a and b. Here, the dash lines indicate the first and last measurements taken from Fig. 4.b and the straight lines show the new measurements. As seen in the figure, the absorbed light causes an increase at the transmittance in the 200–750 nm wavelength region in the new measurements. This shows that in the dark, the new absorbed light could trigger the change in the short wavelength and the absorbed light from the initial measurement may not emit out of the sample. The absorbed light causes the color centers to occur in the composite. These are due to

Fig. 4. At room temperature, the UV-Vis spectra depending on time of (a) SiO₂ (b) Fe₂O₃-SiO₂ (d) Dy₂O₃-SiO₂ and the variation of transmittance at 425 and 800 nm wavelength for (c) Fe₂O₃-SiO₂, (e) Dy₂O₃-SiO₂.

Fig. 5. After 30 min in the dark, the UV-Vis spectrum of (a) $\text{Fe}_2\text{O}_3\text{-SiO}_2$ (b) $\text{Dy}_2\text{O}_3\text{-SiO}_2$.

Fig. 6. Hysteresis curves of (a) $\text{Fe}_2\text{O}_3\text{-SiO}_2$. The black line with dots is the measured data and the blue line with hollow circle is the obtained data after the removed paramagnetic part. The red line is the fit curves and (b) the paramagnetic curve of $\text{Dy}_2\text{O}_3\text{-SiO}_2$ under 2 T magnetic field at room temperature (Color figure online).

the electron and hole trapping in the Fe^{3+} and Fe^{4+} ions. As a result of this, these color centers might be permanent after the initial irradiation.⁴⁶

Similarly, the $\text{Dy}_2\text{O}_3\text{-SiO}_2$ sample waited in the dark for 30 min and in the light for 5 min, separately. In Fig. 5b, the dash lines indicate the first and last lines from initial measurements in Fig. 4d. The transmittance increases a bit more between 200 nm and 800 nm, but this change is more prominent than other regions between 500 nm and 800 nm. This shows that the sample emits the absorbed light in the initial state and, therefore, it recovers in the dark. However, when the sample is

exposed to light for 5 min, the absorption begins between 550 nm and 800 nm wavelength.

It is very well known that the transition element, Fe exhibits strongly magnetic behavior and moreover, iron oxides have both magnetic property and high coercivity due to their hysteresis.^{47,48} In Fig. 6a and b, the hysteresis curves of $\text{Fe}_2\text{O}_3\text{-SiO}_2$ and $\text{Dy}_2\text{O}_3\text{-SiO}_2$ composites are given under 2 T magnetic field at room temperature. In Fig. 6a, the magnetic behavior of the $\text{Fe}_2\text{O}_3\text{-SiO}_2$ is a mixture of paramagnetic behavior related to SiO_2 and the ferromagnetic behavior related to Fe_2O_3 . To obtain the ferromagnetic part of this curve, the linear fit function was applied, and it was removed from this curve. The saturation value for $\text{Fe}_2\text{O}_3\text{-SiO}_2$ was found to be $0.15 \text{ Am}^2 \text{ kg}^{-1}$. The obtained coercivity of the sample is 40 mT. In Fig. 6b, the $\text{Dy}_2\text{O}_3\text{-SiO}_2$ shows the paramagnetic behavior at room temperature. Here, both Dy_2O_3 and SiO_2 are the paramagnetic and Dy_2O_3 exhibits ferromagnetic behavior at low temperature.^{49,50}

CONCLUSION

In this work, SiO_2 , $\text{Fe}_2\text{O}_3\text{-SiO}_2$, and $\text{Dy}_2\text{O}_3\text{-SiO}_2$ composites were prepared by a facile method and their structural, optical and magnetic properties were investigated. The XRD results show that the expected composites have been prepared successfully. The SiO_2 crystallizes in a mixture of two trigonal crystal structures and $\text{Fe}_2\text{O}_3\text{-SiO}_2$ possesses a mixture of rhombohedral (related to Fe_2O_3) and trigonal (related to SiO_2) crystal structures. For $\text{Dy}_2\text{O}_3\text{-SiO}_2$ composite, the crystal structure is found to have a mixture of tetragonal Dy_2O_3 and cubic SiO_2 . The SiO_2 transforms from trigonal to cubic with the addition of Dy. In the FTIR spectra, the absorption bands were determined at room temperature for all composites. The peaks at 1033 cm^{-1} and 586 cm^{-1} due to Fe_2O_3 were found and the peaks at 613 cm^{-1} and 516 cm^{-1} were attributed to Dy_2O_3 . In UV-Vis analyses, the photocatalytic properties of all composites were determined as a function of time at room temperature.

The maximum transmittance variation occurred to be 75% at 800 nm wavelength for Fe₂O₃-SiO₂, and the 2% at 425 and the 40% at 800 nm variations were observed for Dy₂O₃-SiO₂. Here, the Fe₂O₃-SiO₂, and Dy₂O₃-SiO₂ rapidly absorbs the light around 300 nm. Moreover, the dark and light sensitivities of these composites were measured after waited in the dark and the light, separately. The absorbed light causes an increase at the transmittance in the 200–750 nm wavelength region. Consequently, in the dark, the new absorbed light could trigger the change in the short wavelength and the absorbed light from the initial measurement may not emit out of the sample. The magnetic behavior of these composites was defined by the hysteresis curves under 2 T magnetic field. The weak ferromagnetic behavior was observed in the Fe₂O₃-SiO₂ and the composite can not reach a saturation. Therefore, we obtained the ferromagnetic part of this curve. For the ferromagnetic part, the saturation value for Fe₂O₃-SiO₂ was found to be 0.15 Am²kg⁻¹. The obtained coercivity of the sample is 40 mT under 2 T magnetic field. Dy₂O₃-SiO₂ exhibited paramagnetic behavior as expected. The results show that these compositions can be produced with this facile method, and the Fe₂O₃-SiO₂ and Dy₂O₃-SiO₂ composites can absorb the UV-light at long wavelengths due to their photocatalytic properties. Hence, the results specify that the present composites could be applied for the developing photocatalytic materials, the optical fibers and electronic devices for light protection.

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REFERENCES

1. S. Soltan, H. Jafari, S. Afshar, and O. Zabihi, *Water Sci. Technol.* 77, 1689 (2016). <https://doi.org/10.2166/wst.2016.343>.
2. S.C. Feifel and F. Lisdat, *J. Nanobiotechnol.* 9, 59 (2011). <https://doi.org/10.1186/1477-3155-9-59>.
3. S.H. Huang and R.S. Juang, *J. Nanopart. Res.* 13, 4411 (2011). <https://doi.org/10.1007/s11051-011-0551-4>.
4. C. Pacurariu, E.A. Taculescu, R. Ianos, O. Marinica, C.V. Mihali, and V. Socoliuc, *Ceram. Int.* 41, 1079 (2015). <https://doi.org/10.1016/j.ceramint.2014.09.031>.
5. E. Esmaeili, M. Salavati-Niasari, F. Mohandes, F. Davar, and H. Seyghalkar, *Chem. Eng. J.* 170, 278 (2011). <https://doi.org/10.1016/j.cej.2011.03.010>.
6. U. Schwertmann and R.M. Cornell, *The Iron Oxides in the Laboratory, Preparation and Characterization* (Weinheim: VCH Publishers, 1996).
7. M. Mishra and D.M. Chun, *Appl. Catal. A: Gen.* 498, 126 (2015). <https://doi.org/10.1016/j.apcata.2015.03.023>.
8. M. Salavati-Niasari, F. Davar, and M. Mazaheri, *J. Alloys Compd.* 470, 502 (2009). <https://doi.org/10.1016/j.jallcom.2008.03.048>.
9. M. Salavati-Niasari, P. Salemi, and F. Davar, *J. Mol. Catal. A: Chem.* 238, 215 (2005). <https://doi.org/10.1016/j.molcata.2005.05.026>.
10. F. Beshkar and M. Salavati-Niasari, *J. Nanostruct.* 5, 17 (2015). <https://doi.org/10.7508/jns.2015.01.003>.
11. F. Motahari, M.R. Mozdianfard, F. Soofivand, and M. Salavati-Niasari, *RSC Adv.* 4, 27654 (2014). <https://doi.org/10.1039/C4RA02697G>.
12. F. Soofivand, F. Mohandes, and M. Salavati-Niasari, *Mater. Res. Bull.* 48, 2084 (2013). <https://doi.org/10.1016/j.materresbull.2013.02.025>.
13. M. Ranjbar, M. Salavati-Niasari, S.M. Hosseinpour-Mashkani, and K. Venkateswara-Rao, *J. Inorg. Organomet. Polym. Mater.* 22, 1122 (2012). <https://doi.org/10.1007/s10904-012-9704-x>.
14. O. Amir, M. Salavati-Niasari, S.M. Hosseinpour-Mashkani, A. Rafiei, and S. Bagheri, *Mater. Sci. Semicond. Process.* 27, 261 (2014). <https://doi.org/10.1016/j.mssp.2014.06.020>.
15. H. Zeinali, S. Behnam Mousavi, and S.M. Hosseinpour-Mashkani, *Mater. Lett.* 144, 65 (2015). <https://doi.org/10.1016/j.matlet.2015.01.023>.
16. N. Mir, M. Bahrani, E. Safari, and S.M. Hosseinpour-Mashkani, *J. Cluster Sci.* 26, 1103 (2015). <https://doi.org/10.1007/s10876-014-0800-7>.
17. A. Sobhani-Nasab, Z. Zahraei, M. Akbari, M. Maddahfar, and S.M. Hosseinpour-Mashkani, *J. Mol. Struct.* 1139, 430 (2017). <https://doi.org/10.1016/j.molstruc.2017.03.069>.
18. S. Zinatloo-Ajabshir, M.S. Morassaei, and M. Salavati-Niasari, *J. Colloid Interface Sci.* 497, 298 (2017). <https://doi.org/10.1016/j.jcis.2017.03.031>.
19. M. Mahdiani, F. Soofivand, F. Ansari, and M. Salavati-Niasari, *J. Clean. Prod.* 176, 1185 (2018). <https://doi.org/10.1016/j.jclepro.2017.11.177>.
20. J. Balbuena, M. Cruz-Yusta, A. Pastor, and L. Snchez, *J. Alloys Compd.* 735, 1553 (2018). <https://doi.org/10.1016/j.jallcom.2017.11.259>.
21. V.M. Bogatyrev, V.M. Gunko, M.V. Galaburda, M.V. Borysenko, V.A. Pokrovskiy, O.I. Oranska, E.V. Polshin, O.M. Korduban, R. Leboda, and J. Skubiszewska-Ziba, *J. Colloid Interface Sci.* 338, 376 (2009). <https://doi.org/10.1016/j.jcis.2009.06.044>.
22. Q. Sun, W. Leng, Z. Li, and Y. Xu, *J. Hazard. Mater.* 229230, 224232 (2012). <https://doi.org/10.1016/j.jhazmat.2012.05.098>.
23. X. Wang, C. Liu, X. Li, F. Li, and S. Zhou, *J. Hazard. Mater.* 153, 426433 (2008). <https://doi.org/10.1016/j.jhazmat.2007.08.072>.
24. Y.B. Saddeek, K.H.S. Shaaban, R. Elsamani, A. El-Taher, and T.Z. Amer, *Radiat. Phys. Chem.* 150, 182 (2018). <https://doi.org/10.1016/j.radphyschem.2018.04.028>.
25. M.S. Rao, B. Sanyal, K. Bhargavi, R. Vijay, I.V. Kityk, and N. Veeraiyah, *J. Mol. Struct.* 1073, 174 (2014). <https://doi.org/10.1016/j.molstruc.2014.04.075>.
26. H. Huang, Y. Ou, S. Xu, G. Fang, M. Li, and X.Z. Zhao, *Appl. Surf. Sci.* 254, 2013 (2008). <https://doi.org/10.1016/j.apsusc.2007.08.041>.
27. G.A.S. Josephine and A. Sivasamy, *Environ. Sci. Technol. Lett.* 1, 172 (2014). <https://doi.org/10.1021/ez4002032>.
28. G.A.S. Josephine and A. Sivasamy, *Appl. Catal. B: Environ.* 150–151, 288 (2014). <https://doi.org/10.1016/j.apcatb.2013.11.004>.
29. A. Kumar, N. Yadav, M. Bhatt, N.K. Mishra, P. Chaudhary, and R. Singh, *Res. J. Chem. Sci.* 5, 98 (2015).
30. J.D. Mackenzie, *J. Non-Cryst. Solids* 100, 162 (1988).
31. A. Altomare, N. Corriero, C. Cuocci, A. Falcicchio, A. Moliterni, and R. Rizzi, *J. Appl. Crystallogr.* 48, 598 (2015). <https://doi.org/10.1107/S1600576715002319>.
32. S. Vasanthavel, B. Derby, and S. Kannan, *Inorg. Chem.* 56, 1273 (2017). <https://doi.org/10.1021/acs.inorgchem.6b02361>.

33. S. Moshtaghi, S. Zinatloo-Ajabshir, and M. Salavati-Niasari, *J. Mater. Sci.: Mater. Electron.* 27, 425 (2016). <https://doi.org/10.1007/s10854-015-3770-0>.
34. A. Vazquez, T. Lopez, R. Gomez, Bokhimi, A. Morales, and O. Novaro, *J. Solid State Chem.* 128, 161 (1997). <https://doi.org/10.1006/jssc.1996.7135>.
35. A. Aronne, S. Esposito, and P. Pernice, *Mater. Chem. Phys.* 51, 163 (1997). [https://doi.org/10.1016/S0254-0584\(97\)80287-8](https://doi.org/10.1016/S0254-0584(97)80287-8).
36. W.L. Konijnendijk, *Glastechn. Ber.* 48, 216 (1975).
37. W.L. Konijnendijk and J.M. Stevels, *J. Non-Cryst. Solids* 21, 447 (1976). [https://doi.org/10.1016/0022-3093\(76\)90033-8](https://doi.org/10.1016/0022-3093(76)90033-8).
38. Y. Tsunawaki, N. Iwamoto, T. Hattori, and A. Mitsuishi, *J. Non-Cryst. Solids* 44, 369 (1981). [https://doi.org/10.1016/0022-3093\(81\)90039-9](https://doi.org/10.1016/0022-3093(81)90039-9).
39. S. Maensiri, C. Masingboon, B. Boonchom, and S. Seraphin, *Scripta Mater.* 56, 797 (2007). <https://doi.org/10.1016/j.scriptamat.2006.09.033>.
40. S. Zhang, *Nanostructured Thin Films and Coatings: Functional Properties* (Routledge: Taylor and Francis, 2010).
41. Y.B. Saddeek, K.H.S. Shaaban, R. Elsaman, A. El-Taher, and T.Z. Am, *Proc. Indian Acad. Sci.* 12A, 93 (1940).
42. V.C. Pierre and M.J. Allen, Contrast Agents for MRI. *The Royal Society of Chemistry, London.* (2018). <https://doi.org/10.1039/9781788010146>.
43. M. Arshad, A. Azam, A.S. Ahmed, S. Mollah, and A.H. Naqvi, *J. Alloys Compd* 509, 8378 (2011). <https://doi.org/10.1016/j.jallcom.2011.05.047>.
44. S. Muthu Kumaran and R. Gopalakrishnan, *J. Sol-Gel Sci. Technol.* 62, 193 (2012). <https://doi.org/10.1007/s10971-012-2708-8>.
45. J. Pisarska, *J. Phys.: Condens. Matter* 21, 285101 (2009). <https://doi.org/10.1088/0953-8984/21/28/285101>.
46. K. Kadono, N. Itakura, T. Akai, M. Yamashita, and T. Yazawa, *J. Phys.: Condens. Matter* 22, 045901 (2010). <https://doi.org/10.1088/0953-8984/22/4/045901>.
47. R. Kant, D. Kumar, and V. Dutta, *RSC Adv.* 5, 52945 (2015). <https://doi.org/10.1039/C5RA06261F>.
48. T.P. Raming, A.J.A. Winnubst, C.M. van Kats, and A.P. Philipse, *J. Colloid Interface Sci.* 249, 346 (2002). <https://doi.org/10.1006/jcis.2001.8194>.
49. D.X. Chen, V. Skumryev, and B. Bozzo, *Rev. Sci. Instrum.* 82, 045112 (2011). <https://doi.org/10.1063/1.3581224>.
50. D.J. Flood, *Phys. Lett. A* 49, 59 (1974). [https://doi.org/10.1016/0375-9601\(74\)90668-9](https://doi.org/10.1016/0375-9601(74)90668-9).

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