

## Using Recycled Magnetite from Concrete Waste as a Pigment for Flexographic Ink

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### ABSTRACT

**M**agnetite can be used as an additive and colorant for ink preparation. This material can be produced using several methods; however, its recycling is a safer and more environmentally friendly approach due to resource scarcity. Waste management and recycling are now major concerns due to the rise in industrial production. The current work offered a viable method for recycling magnetite particles, considered residues of micro-silica preparation in concrete production factories. The resulting magnetite was utilized as a pigment in flexographic water-based ink as a replacement for the virgin pigments in an ink formulation. X-ray diffraction (XRD) patterns, scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), particle size analysis (PSA), rheology measurement, and color characteristics were used to characterize the ink and the recovered pigment. The results showed that the recycled magnetite has a larger particle size than the original pigment. In addition, the blackness and printability of the prepared ink are superior to the original black ink. Results showed a small reduction in optical density (O.D.) of the ink made from recycled pigment in comparison with virgin ink. Despite these lower O.D. values, it is still suitable in the appropriate range for use in applications without changes in viscosity parameters. Prog. Color Colorants Coat. 19 (2026), 1-8© Institute for Color Science and Technology.

### 1. Introduction

There are numerous types of iron oxide, which is the strongest naturally occurring magnetic material [1]. The magnetic characteristics, color, density, and hardness of magnetite (Fe<sub>3</sub>O<sub>4</sub>) make it useful in both science and industry [2]. Magnetite is used as a pigment in building materials and in printing to control the charge of the toner [3, 4]. Magnetic particles can now be produced using a variety of methods, including chemical co-precipitation [8], sonochemical synthesis [7], laser pyrolysis [6], and micro-emulsions [5]. Finding a clean and environmentally friendly method

of producing these particles is crucial because of the high demand for magnetite, resource scarcity, and environmental concerns. Waste recycling has become a significant challenge for developing nations [9]. Waste recovery is an important societal concern in underdeveloped countries since it recycles resources and protects the environment [9].

Concrete is a widely utilized and well-known material in the construction sector. Its development has been associated with attempts to maximize its performance, which is usually accomplished by altering its properties [10]. These changes are initially caused by

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rebalancing the materials and then by the addition of new components. Over 500,000 MT of micro-silica is purchased by the global construction sector for usage in oil-well drilling, fiber cement, concrete treatment, and refractories [11]. Microsilica is an industrial byproduct generated during the production of ferrosilicon alloys. A comparatively high amount of gases including CO and SiO are produced from the reaction zone during the ferrosilicon manufacturing process. Then, these gases leave the loading surface of the furnace and are converted into CO<sub>2</sub> gas and silica (SiO<sub>2</sub>) dust. The latter is separated via several mid-stage processes. Microsilica did not begin to be traded as a byproduct of this process until technology advanced and various uses for these wastes (especially, in concrete) were discovered. About 220 kg of micro-silica is produced for every ton of ferrosilicon throughout the production process [12]. Traces of organic deposits and heavy metal oxides are present in microsilica as impurities [13]. Iron oxide is one of the metal oxides that are present in microsilica as waste and impurities [14].

The author has previously reported the impact of various colorants on printing ink [15, 16]. This study aims to determine the potential of the iron oxide (magnetite) obtained as waste for curing and manufacturing microsilica to be used in flexographic ink. The recovered pigments were utilized to make the printing ink to investigate the relationship between the pigment characteristics (such as morphology and particle size distribution) and the ink characteristics (such as color and optical density).

## 2. Experimental

### 2.1. Materials

The flexographic ink (FI) was a cold-set water-based black ink purchased from the Iran Ink Company. A typical formulation of the used ink is shown in Table 1. To apply the ink onto the paper sheets, a K Hand-type bar Coater (UK) with a thickness of 4 µm was used. Also, the uniformity of the film was measured by Film-Thickness Measurement. for the preparation of the water-based ink as follows: mixing all components at room temperature by stirring for 20-30 minutes through a high-speed dispersion machine at the rotating speed of 6000-9000 rpm, obtaining a dispersed color paste, milling the color paste in a sand mill containing glass balls for 5-7 hours, and filtering the obtained water-based ink.

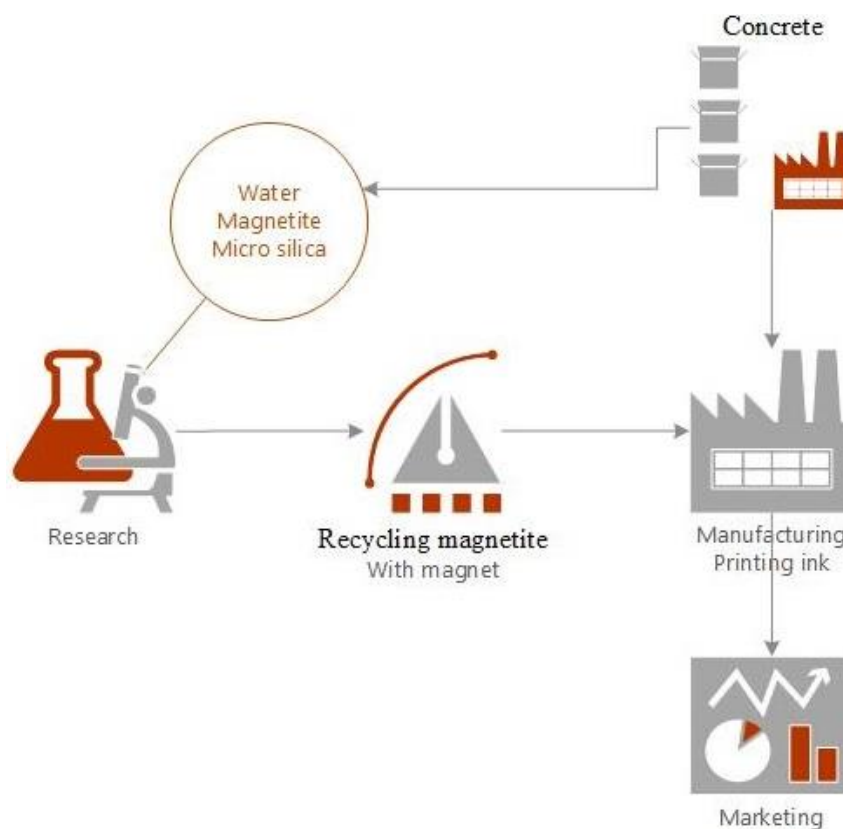
Microsilica was obtained from Ferrosilicon Company (Iran). Before preparing and treating the micro-silica for the concrete industry, its surface must be modified using ionic stabilizers and mineral salts. The waste iron oxide precipitates as black sediments and should be removed from the microsilica solution. A magnet extracts such particles from the slurry because of their magnetic properties. The full recycling method is reported in our previous work [15]. Figure 1 shows the preparation scheme.

### 2.2. Characterization

The recovered pigment and the ink were subjected to an X-ray diffractometer (STOE STADI P, Germany) employing CuK<sub>α</sub> radiation ( $\lambda=1.54 \text{ \AA}$ ). SEM-EDX was used to characterize the pigment morphology (Cambridge Instruments Stereoscan 360, UK). PSA (Mastersizer2000, Malvern, UK) was used to measure the particle size and distribution. The optical density of the printed ink was determined using an Ihara S900spectrophotometer (USA). The Ihara SpectroCam (Japan) was used to assess the color properties. The spectral reflectance factor was calculated and converted into CIELAB colorimetric coordinates (L\*, a\*, and b\*) using a CIE standard colorimetric observer and CIE standard illuminant D65 [17]. The rheological properties of the inks were studied using a US200 Anton Paar MCR 300 with a cone and plate geometry cell in controlled shear stress mode. In each case, the shear rate was increased from 0.01 to 10 Hz and reversed back down to 0.01 Hz, with 7 measurements distributed on a logarithmic scale for increasing and decreasing shear rates.

**Table 1:** A typical formulation of the FI [17]

Component	Amount (%)
Orginal pigment or recycled pigment	5.9
Alkali-soluble acrylic copolymer	28.4
Antifoam	0.36
Antibacterial	0.1
Wetting agent	0.2
PE wax emulsion	3
Isopropyl alcohol	2
Distilled water	6.04



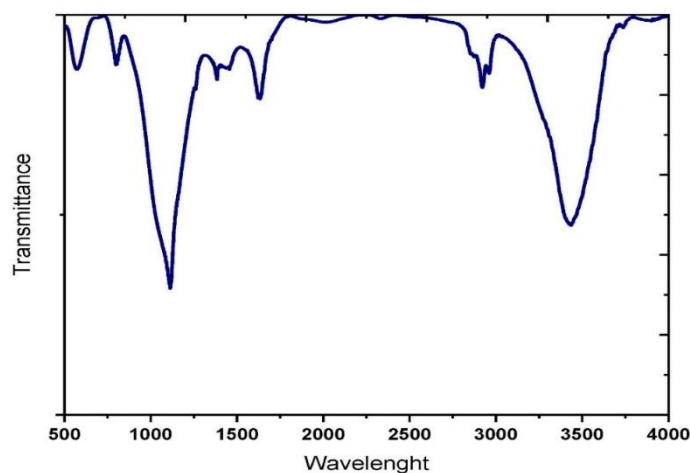
**Figure 1:** A scheme representing the recycling of pigment from concrete waste.

### 3. Results and Discussion

As seen in Figure 2, the recovered pigment exhibits an absorption peak at  $1600\text{ cm}^{-1}$  attributed to the bending of the O-H bond and a characteristic band at about  $600\text{ cm}^{-1}$  relating to the stretching of the Fe-O bond [18]. The peak at  $1000\text{ cm}^{-1}$  matches the bending vibration of the O-H bond. Both the water molecules chemically adsorbed to the surfaces of the magnetic particles and the hydroxyl groups bonded by hydrogen bonds to the iron oxide surface are represented by these two bands [19]. The intensity of FTIR bands is influenced by particle size which was explained in DLS and SEM analyses.

In Figure 2, the broad high-intensity band at  $1061\text{ cm}^{-1}$  is associated with the motion of oxygen in Si-O-Si antisymmetric stretch, due to the asymmetric stretching bonds of Si-O-Si in  $\text{SiO}_2$ . The presence  $\text{SiO}_2$  of is also confirmed by EDX. The band at  $793\text{ cm}^{-1}$  is assigned to the Si-O-Si symmetric stretch, while the band at  $451\text{ cm}^{-1}$  corresponds to the Si-O-Si or O-Si-O bending modes. The band at  $964\text{ cm}^{-1}$  is assigned to the Si-O symmetric stretch. In Figure 2, the broadband at

$1076\text{ cm}^{-1}$  is for asymmetric stretching bonds of Si-O-Si, the band at  $771\text{ cm}^{-1}$  is for Si-O-Si symmetric stretch, the band at  $454\text{ cm}^{-1}$  is for Si-O-Si or O-Si-O bending modes, the band at  $954\text{ cm}^{-1}$  is for Si-O symmetric stretch [20].



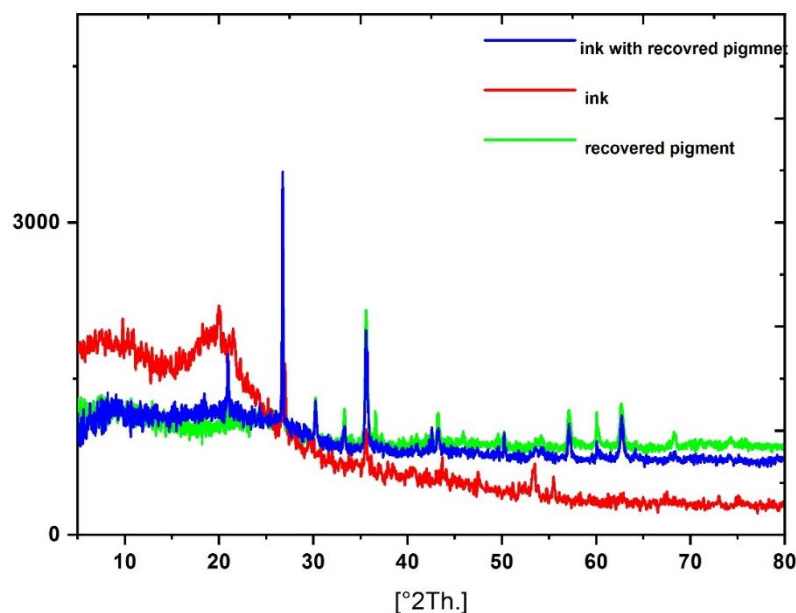
**Figure 2:** The FTIR diagram of the recovered pigment.

Figure 3 shows XRD patterns of the recovered pigment and the ink. The recovered pigment consists of silicon oxide ( $\text{SiO}_2$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), and magnetite ( $\text{Fe}_3\text{O}_4$ ) according to JCPDS card No. 01-086-1629, JCPDS card No. 00-033-0664, and JCPDS card No. 01-075-0033, respectively. The  $2\theta$  peaks of  $30.206^\circ$ ,  $35.565^\circ$ ,  $43.199^\circ$ ,  $53.579^\circ$ ,  $57.121^\circ$ , and  $62.734^\circ$  in  $\text{Fe}_3\text{O}_4$  particles represent the inverse spinel structure of  $\text{Fe}_3\text{O}_4$ . The sources of the diffraction peaks were determined to be the crystal planes located at Miller indices (220), (311), (400), (422), (511), and (440) [21]. Before and during mixing, the peak position of the recovered pigment remained unchanged. This outcome demonstrated that the ink preparation process did not alter the crystal structure of the recovered pigment. The wide diffraction peaks of the ink between  $10^\circ$  and  $30^\circ$  were thought to be caused by the amorphous polymer [22]. From Figure 3, we can observe that the XRD pattern of the  $\text{SiO}_2$  microspheres is similar to the pattern of  $\text{Fe}_3\text{O}_4$ , and the presence of  $\text{SiO}_2$  does not change the structure of the  $\text{Fe}_3\text{O}_4$  particles [19].

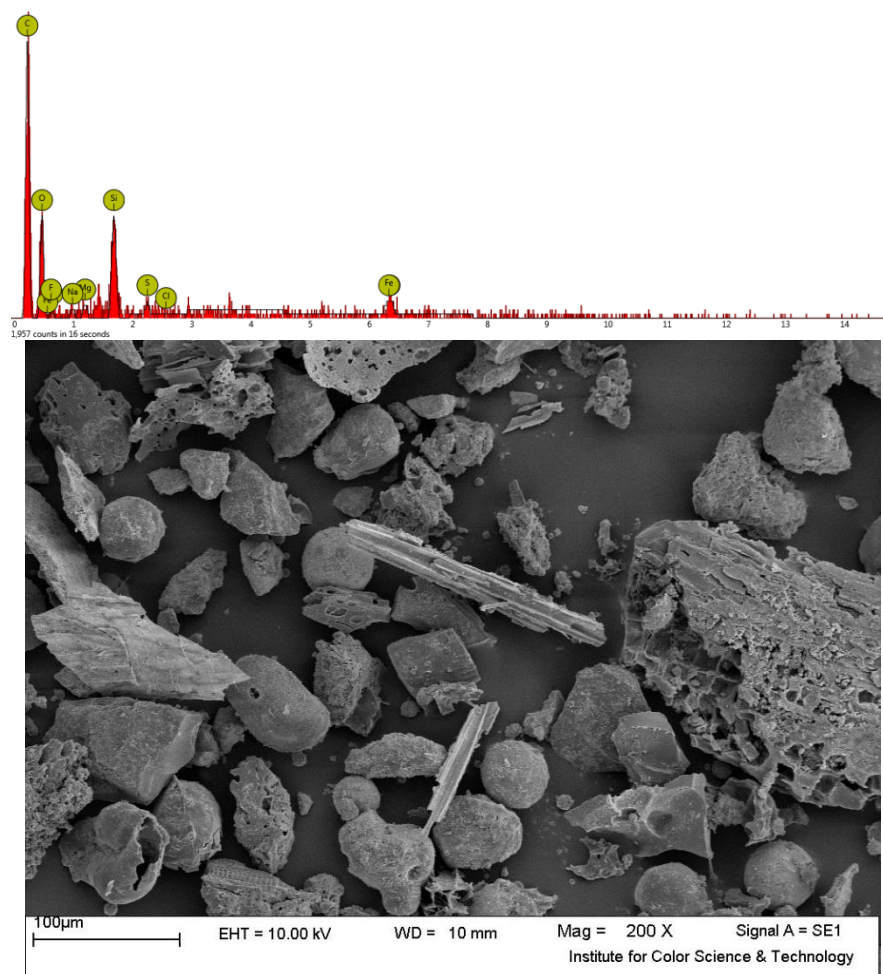
To determine the morphology and the average size of waste pigment, scanning electron microscopy (SEM) was used. In Figure 4, the SEM image of the waste pigment shows that these particles have an irregular

morphology, and the average size is about  $9\text{ }\mu\text{m}$ , which is confirmed by image j software. SEM-EDX images of the recovered pigment particles are also displayed in Figure 4. The EDX spectrum of the recovered pigment showed iron (3.1 %), silicon (6.9 %), carbon (30.5 %), and oxygen (49.5 %) elements. It is evident from the SEM image of the recovered pigment that micro-silica particles are visible next to the pigment particle. The larger particle size of the recovered pigment can also be seen in the SEM images, which is also confirmed by PSA results.

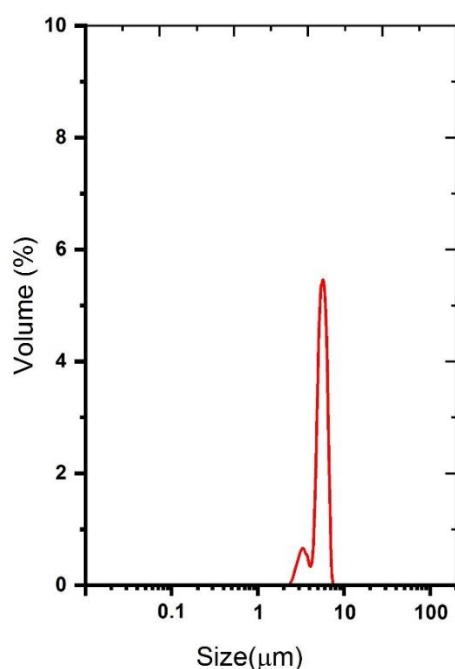
The particle size distribution of the recovered magnetite is displayed in Figure 5. The scattering power of pigments is significantly influenced by their particle size, which should ideally be under 1 micron, and preferably below 0.5 microns, depending on whether pigments with a high or low refractive index are utilized. According to the findings, the recovered pigment particles are somewhat larger than the original pigment particles, even after milling. This larger particle size mostly affects the printability (optical density) and color characteristics of the ink, while having a lesser effect on the rheological properties [23].



**Figure 3:** XRD patterns of the original ink, the recovered pigment, and the ink containing the recovered pigment.



**Figure 4:** EDX pattern and SEM image of the recovered pigment.



**Figure 5:** Particle size distribution diagram of the recovered pigment.

Figure 6 shows the shear stress and viscosity diagrams versus the shear rate for both the original ink and the ink containing the recovered pigment. Newtonian behavior is evident in the difference between the loading and unloading curves of the viscosity diagram [24]. The findings indicate that while both inks exhibit identical rheological behavior, their viscosity values differ because of their varied pigment particle size. As expected, all inks exhibit mild thixotropy between increasing and decreasing shear runs. They also exhibit shear-thinning behavior to approximately the same extent over this shear rate range.

The colorimetric coordinates ( $L^*$ ,  $a^*$ , and  $b^*$ ) were measured using the CIE standard illuminant D65. An increase in the  $L^*$  indicates higher lightness. The  $b^*$  axis exhibits blue and yellow extremes, whereas the  $a^*$  axis exhibits a green hue at its negative extremity ( $-a^*$ ) and a red hue at its positive extremity ( $+a^*$ ) [21]. Both inks showed  $a^*$  and  $b^*$  values close to zero, which provided

the appropriate blackness of the samples within the original ink range, despite changes in lightness values. However, the nonuniformity of the ink results from the greater particle size of the recovered pigment which alters the  $L^*$ ,  $a^*$ , and  $b^*$ . The optical density (O.D.) of the ink can be calculated from Eq. 1:



$$\text{O.D.} = \log L_W/L_R \quad (1)$$

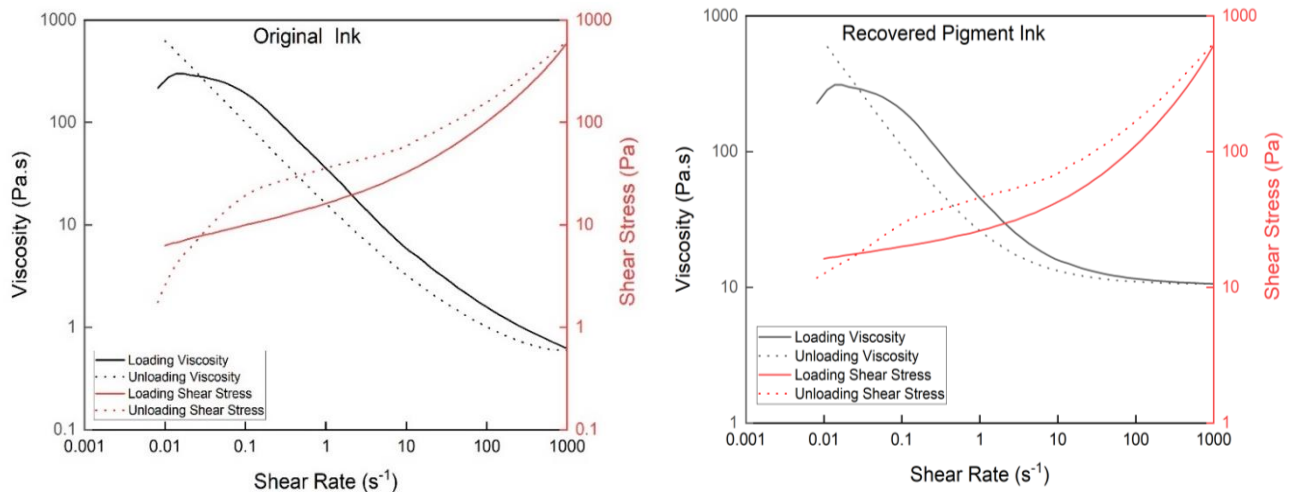
$L_W$  and  $L_R$  are the intensity of the reflected light from the white area of the paper and the printed ink, respectively. Better optical density and higher print quality are indicated by density values of 1 and above [22].

In general, the optical density reading increases with the quality of the ink film. It was discovered that the average optical density of the recovered pigment

ink fell within that of the original ink (Table 2), although it has a larger particle size. The hue and opacity of the pigment are directly influenced by the size of the particles. The black hue seems more intense when smaller particles scatter lights more effectively. Larger particles, on the other hand, only slightly scatter light, producing a softer black shade. Furthermore, smaller particles have greater opacity, which enhances their ability to conceal and provide superior coverage in inks and other applications. Due to a more uniform particle size distribution and a smaller mean particle size, the original sample exhibits greater printability when compared to the waste sample based on the optical density of the printed samples. Although the optical density of both samples remains within a reasonable range [25].

**Table 2:** Spectrophotometric parameters of the original and the recovered pigment inks.

Sample	O.D.	$L^*$	$a^*$	$b^*$	Image
Original ink	1.31	41.47	0.58	-0.82	
Recovered pigment ink	1.04	32.30	1.12	1.02	



**Figure 6:** Viscosity and shear stress behavior of the original ink and the ink containing the recovered pigment.



## 4. Conclusions

In this study, for the first time, magnetite particles recycled from microsilica wastes in concrete factories were used as pigment for printing applications to overcome resource scarcity and environmental concerns. The recovered pigment particles exhibit  $\text{Fe}_2\text{O}_3$ ,  $\text{Fe}_3\text{O}_4$ , or  $\text{SiO}_2$  phases according to XRD and SEM-EDX data. They were used in flexographic water-based ink and compared with the original ink.

## 5. References

1. Cornell RM, Schwertmann U. The Iron Oxides: Structure, Properties, Reactions Occurrences and Uses. Wiley-VCH, 2003
2. Ataefard M, Ghasemi E, Ebadi M, Effect of micro- and nanomagnetite on printing toner properties. *Sci World J.* 2014; In press. <https://doi.org/10.1155/2014/706367>.
3. Teja AS, Koh PY, Synthesis, properties, and applications of magnetic iron oxide nanoparticles. *Prog. Crystal Growth Charact Mater.* 2009; 55: 22-45, <https://doi.org/10.1016/j.pcrysgrow.2008.08.003>
4. Meisen U, Kathrein H, The influence of particle size, shape and particle size distribution on properties of magnetites for the production of toners. *J Image Sci Technol.* 2000; 44(6):508-513, [https://doi.org/10.2352/ISSN.2169-4451.1999.15.1.art00045\\_2](https://doi.org/10.2352/ISSN.2169-4451.1999.15.1.art00045_2).
5. Quanguo WW, Jiang HC. Magnetic iron oxide nanoparticles: synthesis and surface functionalization strategies, *Nano-scale Rese.* 2008;3:397-415. <https://doi.org/10.1007/s11671-008-9174-9>.
6. Yang H, Li S, Yang R, Bai J, Guo Z. Recovery of silicon powder from kerf loss slurry waste using superconducting high gradient magnetic separation technology, *J Mater Cycles Waste Manag.* 2018; 20(2): 937-945. <https://doi.org/10.1007/s10163-017-0656-7>.
7. Faiyas APA, Vinod EM, Joseph J, Ganesan R, Pandey RK. Dependence of pH and surfactant effect in the synthesis of magnetite ( $\text{Fe}_3\text{O}_4$ ) nanoparticle and its properties. *J Magnet Magnetic Mater.* 2010; 322: 400-404, <https://doi.org/10.1016/j.jmmm.2009.09.064>.
8. Sun S, Zeng H, Size-controlled synthesis of magnetite nanoparticles. *J Am Chem Soc.* 2002; 124(28): 8204-8205, <https://doi.org/10.1021/ja026501x>.
9. Kien N, Tomoaki B, Hiroshi Takahashia S. Recycling woven plastic sack waste and PET bottle waste as fiber in recycled aggregate concrete: An experimental study, *Waste Manag.* 2018; 78: 79-93, <https://doi.org/10.1016/j.wasman.2018.05.035>.
10. Pedro J. de Brito D, Evangelista L. Mechanical characterization of high performance concrete prepared with recycled aggregates and silica fume from precast industry. *J Clean Prod.* 2017; 164: 939-949, <https://doi.org/10.1016/j.jclepro.2017.06.249>.
11. Gite BE, Rath MK, Rajguru RS, P. Shaikh A. Advance construction material-micro silica in concrete. *Concrete Soc. Tech., Report No.41*, 1993.
12. Limbachiya M, Leelawat T, Dhir R. Use of recycled concrete aggregate in high-strength concrete. *Mater Struct.* 2000;33:574-580. <https://doi.org/10.1007/BF02480538>.
13. Friede B. Microsilica-characterization of an unique additive, Conference: International inorganic-bonded fiber composites conference (IIBCC), At Sao Paulo, Brazil, Volume: 10.
14. Guide for the Use of Silica Fume in Concrete, ACI Committee 234, 2000.
15. Ataefard M, Salehi MM, Preparing poly styrene-co-acrylic acid and recycled waste magnetite composite from concrete industry as a printing ink. *J Thermo-plast Compos Mater.* 2022;35:2298-2309. <https://doi.org/10.1177/0892705720962156>.
16. Ataefard M, Aarabi AM. Producing ceramic toner via emulsion aggregation method based on  $\text{ZrSiO}_4$ :Pr Ceramic Pigment. *Prog Color Colorant Coat.* 2021;14: 113-120. <https://doi.org/10.30509/pccc.2021.81703>.
17. The Printing Ink Manual, Robert Leach, Ray Pierce, 2007, Springer.
18. Mohammadia P, Sheibani H. Green synthesis of  $\text{Fe}_3\text{O}_4$ @ $\text{SiO}_2$ -Ag magnetic nanocatalyst by using flower extract and its application as recoverable catalyst for reduction of dye pollutants in water. The First Conference on the Development of Science and Chemical Industry Feb 1-2, 2017.
19. Duan Y. Novel preparation of  $\text{Fe}_3\text{O}_4$ /styrene-co-butyl acrylate composite microspheres via a phase inversion emulsion process. *Colloid Polym Sci.* 2017; 295(1): <https://doi.org/10.1007/s00396-017-4154-1>.
20. Shan J, Wang L, Yu H, Ji J, Amer WA, Chen Y, Jing G, Khalid H, Akram M, Abbasi NM. Recent progress in  $\text{Fe}_3\text{O}_4$  based magnetic nanoparticles: from synthesis to application, *Mater Sci Technol.* 2016;32 (6):602-614. <https://doi.org/10.1179/1743284715Y.00000000122>.
21. Yiu HHP, Keane MA, Lethbridge ZAD, Lees MR, Ehaj AJ, Dobson J. Synthesis of novel magnetic iron metal-silica (Fe-SBA-15) and magnetite-silica

- (Fe<sub>3</sub>O<sub>4</sub>-SBA-15) nanocomposites with a high iron content using temperature-programed reduction. *Nanotechnology*. 2008; 19: 255-606. <https://doi.org/10.1088/0957-4484/19/25/255606>.
22. Bishop CA. Process diagnostics and coating characteristics, vacuum deposition onto webs, *Film Foil*. 2011; 4: 81-114. <https://doi.org/10.1016/B978-1-4377-7867-0.00005-2>.
  23. Ghaffarzadeh O, Ataefard M, Hedjazi S, Abdulkhani A, Taherzadeh MJ. Intelligent modeling of printability of liner paper coated with modified soda lignin. *Biomass Conversion Biorefin*. 2025; In Press. <https://doi.org/10.1007/s13399-025-06817-w>
  24. Salehi MM, Ataefard M. Micro powder poly lactic acid/carbon black composite as a bioprinting ink. *J Comp Mater*. 2019;53(17):2407-2414. <https://doi.org/10.1177/0021998319828154>.
  25. Ataefard M. Electrophotographic printing of fabrics: Investigating the effect of fabrics on color reproduction. *Fiber Polym*. 2016; 17(7): 1055-1061. <https://doi.org/10.1007/s12221-016-5445-8>.

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