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Green Synthesis of Iron Nanoparticles from Aqueous Extract of Fenugreek Seeds (*Trigonella foenum-graecum*): Synthesis Mechanism and Characterization

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***Abstract*—**Green synthesis method refers to the synthesis of metal and metal oxide nanoparticles using plant extracts. Fenugreek seeds (*Trigonella foenum-graecum*) are a rich source of numerous biological and chemical components that are frequently used in cooking and as traditional treatments for diabetes in Asian countries. In this study, the performance of fenugreek seed extract was evaluated for the synthesis of iron nanoparticles (FeNPs). The extraction of fenugreek seeds was prepared with the 1:10 ratio of fenugreek seeds powder to distilled water and the extraction was carried out at 65-70°C for 60 minutes. The aqueous extract of fenugreek seeds can be transformed Fe^{3+} into iron nanoparticles (Fe^{0}) at room temperature. The synthesized FeNPs were evaluated utilizing Scanning Electron Microscope (SEM) analysis, Energy Dispersive X-Ray (EDX) analysis, X-ray diffraction (XRD) analysis and UV-Visible Spectroscopy (UV-Vis) analysis. The particles were visible on the nanoscale in SEM pictures and the particle morphology displayed spherical forms with a particle size range of 20–40 nm. The atomic percentages of synthesized FeNPs were revealed by EDX analysis and which also revealed that the majority of the nanoparticles are in metal form. The crystal structure of the synthesized FeNPs was visible in the XRD spectrum. The UV-Visible analysis identified the synthesized FeNPs peak of absorption in the range 250–295 nm and 305–375 nm regions with the largest peak being found at 268 nm.

***Keywords*—**Green synthesis, Iron nanoparticles, Fenugreek seeds, Room temperature, Eco-friendly.

I. Introduction

Nanotechnology is the science of controlling and utilizing matter through chemical or physical approaches resulting in materials with specific qualities with different applications. It not only creates small buildings, but also mechanized technology that can manage the structure of matter thoroughly and cheaply. Nanotechnology can be explained as a technology, which creates the controllable path to create nanomaterials and also functions with them (Kathiervelu, 2003; Modi *et al*., 2022). Nanotechnology requires research and advancement at atomic and subatomic (atomic, molecular, and macromolecular) levels in a range of 1 to 100 nanometres to get an elementary understanding of nanoscale procedures and constituents. Nanomaterials are characterized using measurements of nanometric scale concerning the sample of the material as a whole and its structural elements. The nanomaterials have a high surface area to volume ratio and which is the most crucial attribute that explains why these nanomaterials are so widely used (Salata, 2004; Samarawickrama *et al*., 2021). Nanotechnology is a fast-developing field of study that is expected to have applications with a broad scope in all fields of science and technology such as material science, material processing technology, mechanics, electronics, medicine, energy and aeronautics, plastics and textiles. Its extensive influence on society has been considered to provide a great inducement to be used in a second industrial revolution (Hinestroza, 2004; Sonali Pradhan, 2013). Nanomaterials are generally classified into nanoemulsions, nanoclays and nanoparticles. The nanoparticles are the most fundamental component of nanomaterials. The nanoparticles can be further divided into subgroups such as carbon-based nanoparticles, lipid-based nanoparticles, metal nanoparticles, semiconductor nanoparticles and polymeric nanoparticles (Sonali Pradhan, 2013; Heera and Shanmugam, 2015). Furthermore, nanoparticles can be classified into numerous categories based on their size, morphology and properties. The nanoparticles exhibit distinct and unique physical and chemical properties at the nano-scale compared to their corresponding particles at higher scales (Cho *et al*., 2013; Military *et al*., 2017).

The physical characteristics of nanoparticles include absorption, reflection, light dispersion, colour, hydrophilicity, hydrophobicity, suspension, and dispersion. Also, nanoparticles have unmatched chemical properties such as sensitivity to humidity, stability in the presence of air, non-toxicity, biodegradability, anti-bacterial, and anti-fungal (Satyanarayana, 2018; Ghorbani, 2014). They benefit from their favourable

chemical and physical properties for a variety of industrial applications. The nanoparticles are used in many industrial applications such as medicine, reducing environmental pollution, developing new energy sources, agriculture, information technology and communication (Thiyagarajan *et al*., 2020; Castillo-henr *et al*., 2020). Nanoparticles can be produced using chemical and physical methods and they can be made in different sizes and shapes of nanoparticles. But, these methods need harsh conditions such as high temperatures and pressure, which factors influence the environmental and social aspects negatively (Heera and Shanmugam, 2015; Satyanarayana, 2018). Therefore, the method of green synthesis method used the nanoparticles synthesis process. The synthesis of metal and metal oxide nanoparticles utilizing plant extracts is known as a method of "green synthesis." The plant extract acts as a reducing and stabilizing agent in the synthesis of metal and metal oxide nanoparticles. However, this method does not need high-pressure, high-temperature and toxic chemicals as required by the physical and chemical methods (Alves *et al*., 2019; Journal, Gour and Jain, 2019). The green synthesis method is growing in popularity throughout industries that focus on the synthesis of environmentally friendly nanomaterials as a result of the above-mentioned advantages. Green synthesized nanoparticles have a surprising number of applications for sustainable development in a variety of disciplines and they have distinct applications in medicine, pharmaceuticals, biomedicine, anti-microbials, optical imaging and molecular sensing etc. Similarly, green synthesized nanoparticles have been used as sorbents for environmental contaminants and in environmental restoration (Links, 2011; Hemmati *et al*., 2018). A promising future lies in green nanoparticle production as one of the most available, efficient and effective methods for obtaining nanostructured materials.

The fenugreek plant (*Trigonella foenum-graecum*) is a seasonal herb in the Fabaceae family and which has three tiny obovate to oblong leaflets that make up the leaves. This is commonly grown in Middle East countries. Since ancient times, people have utilized the seeds and leaves of the fenugreek plant as a frequent ingredient in food (Alarcon-aguilara, Roman-ramos and Perez-gutierrez, 1998). Fenugreek seeds and leaves possess a number of beneficial qualities such as anti-inflammatory, anti-microbial, anti-tumour and antioxidant capabilities. Therefore, they provide a variety of health benefits such as lowering inflammation in the body, improving digestive issues, cholesterol levels, blood sugar control and improving athletic performance etc. (Bouhenni, Doukani and Sekeroglu, 2019). Therefore, fenugreek leaves and seeds have more benefits for the world. Seeds of the fenugreek plant consist of the alkaloid trigonelline along with mucilage, tannic acid, yellow colouring matter, volatile oils, diosgenin and vitamin A. These seeds are also extremely high in protein and one of the active elements in fenugreek seeds is 4-hydroxy isoleucine and unique major free amino acid (Moradi-kor, 2014; Petit *et al*., 2018).

An aqueous extract of fenugreek seeds was employed for the metallic nanoparticle synthesis where it acts as a reducing and stabilizing agent. Aromal and Philip used the aqueous extract of seeds of the fenugreek plant and treated AuCl solution. They successfully treated Au nanoparticles in the range of 15-20 nm (Aromal and Philip, 2012). According to the results of Suganya and Devasena, the size of circular nanoparticles synthesized using aqueous fenugreek seed extract are in a range below 50 nm (Devasena and Suganya, 2014). Fragoon, Frah and Mamoun successfully synthesized spherical-shaped Au nanoparticles using aqueous fenugreek seed extract in a size range between 8-15 nm (Fragoon, Frah and Mamoun, 2016). Rizwana and a few of the researchers treated Ag nanoparticles sized between 9-57 nm with fenugreek plant seed extract to treat (Rizwana *et al*., 2021). Meena and Chouhan successfully synthesized 50-90 nm sized circular Ag nanoparticles using $AgNO_3$ solution and the aqueous fenugreek seed extract (Meena and Chouhan, 2015). In this study, the aqueous fenugreek seed extract is used synthesis of iron nanoparticles (FeNPs). Iron nanoparticles are non-toxic, easily produced, eco-friendly and inexpensive particle types. Iron nanoparticles are used in medical, wastewater treatment, coatings, nano-fibres and textile applications (Iyer, 2019; Saranya, 2017). The synthesized FeNPs were characterized using Scanning Electron Microscope (SEM) analysis, Energy Dispersive X-Ray (EDX) analysis, X-ray diffraction (XRD) analysis and UV-Visible Spectroscopy (UV-Vis) analysis.

II. Methodology

A. Collection of materials and chemicals

The fenugreek seeds were bought from the supermarket in the Colombo area. The ferric chloride hexahydrate ($FeCl_3.6H_2O$) was used as the source of iron (Fe) and which was obtained from the local chemical supplier in Sri Lanka. The dialysis membrane (Molecular weight cut off 12,000-14,000 Dalton) used in this work was purchased from local chemical suppliers in Sri Lanka. The distilled water was prepared by using Distilled Water Dispenser in the laboratory. This study used analytical grade chemicals and they could be utilized without any additional purification.

B. Extraction of fenugreek seeds aqueous extract

Fenugreek seeds were washed thoroughly using distilled water multiple times to remove dust and other matter. Fenugreek seeds were oven dried at 40°C for 6 hours and then finely ground into powder form. The aqueous extraction of the fenugreek seeds was prepared with a 1:10 ratio of fenugreek seeds powder to water and the extraction was carried out at 65-70°C for 60 minutes using the magnetic stirrer with the hot plate. The extract was then vacuum-filtered after cooling down at room temperature.

C. Green synthesis process of iron nanoparticles

The fenugreek seeds extract was mixed with 0.1M ferric chloride solution in a ratio of 1:1 at room temperature. The resulting mixture was stirred for 90 minutes at 300 rpm with a magnetic stirrer at room temperature. The synthesis of iron nanoparticles was confirmed by the observation of an intense black solution. The solution was centrifuged at 6000 rpm for 15 minutes. FeNPs were separated by allowing them to precipitate and then separating the precipitated mass from the supernatant (this process was repeated 2 to 3 times). The supernatant was discarded and the dialysis membrane was used to dialyze the precipitate 2-3 times with distilled water. The synthesized FeNPs were then dried at a temperature of 50°C in a vacuum oven. After the dried process, the FeNPs powder was kept in an air-tight container for future use.

D. Characterization of iron nanoparticles

1) Scanning electron microscope (SEM) and energy dispersive x-ray (EDX) characterization: The surface morphology and structure of synthesized iron nanoparticles were examined using scanning electron microscopy (SEM) analysis (Carl Zeiss eco 18 Research, Germany). Sample image analysis was performed at a 10kV voltage using different magnifications. The Energy Dispersive X-Ray (EDX) analysis (EDAX element detector, USA) attached to the SEM machine was used to determine the chemical composition of synthesized iron nanoparticles.

2) X-ray diffraction (XRD) characterization: The crystalline metallic FeNPs were examined using an X-ray diffractometer. X-ray diffraction spectrum (XRD) was recorded by the Bruker D8 Focus X-Ray Diffraction Spectrometer at the scanning rate of 2°/min from 2θ range of 5°- 60° with Cu Kα radiation (λ = 1.5418 A°) utilizing 40 kV and 30 mA for voltage and current.

3) UV-Visible spectroscopy (UV-Vis) characterization: The optical characteristics of the produced iron nanoparticles were investigated using UV-visible absorption spectroscopy. The absorbency spectrum was evaluated in the wavelength range of 200-800 nm using a Thermo scientific genesis 10S series type UV-Visible spectrophotometer.

III. Results and Discussion

A. Document Visual inspection of the synthesized iron nanoparticles

The colour of the mixture (0.1 M Ferric Chloride solution with fenugreek seeds aqueous extract) changed from pale yellow to a bluish-black at room temperature. Figure 1 shows the bluish-black coloured iron nanoparticle solution. Figure 2 shows the FeNPs in powder form which was successfully separated from the FeNPs solution after the centrifuged and drying processes.

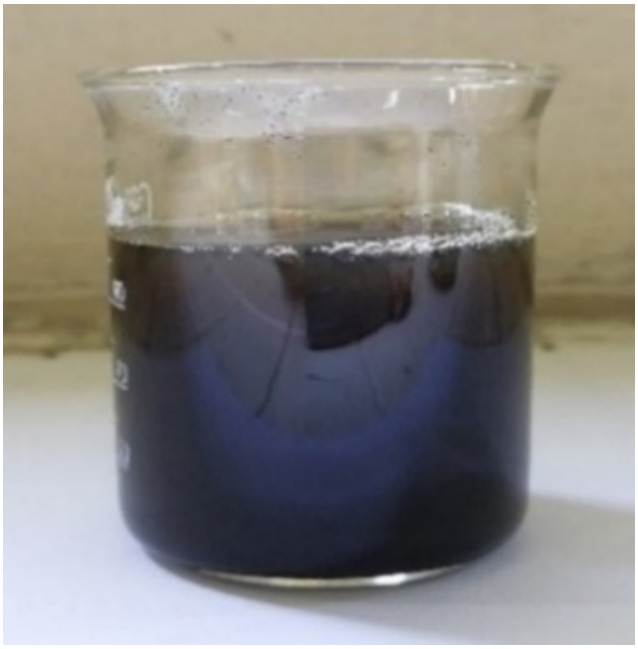

Figure 1: Synthesized FeNPs solution

Figure 2: Synthesized FeNPs in powder form

B. Scanning Electron Microscope and EDX analysis of synthesized iron nanoparticles

The SEM images of synthesized iron nanoparticles were provided with a magnification of 1 μm and 200 nm (Figure 3). This shows that nanoparticles formed are agglomerated due to their innate adhesiveness. They have a spherical morphology and a particle size range of 20–40 nm. These results have shown consistent with the results of synthesized FeNPs using the aqueous extract of fenugreek seeds (Shahwan *et al.*, 2011). The EDX diagram is shown in Figure 4 and which qualification gives atomic percentages are 4.78% of iron, 47.47% of oxygen, 0.78% of magnesium, 0.95% of sodium, 25.75% of chlorine and 19.46% of potassium. This EDX analysis from determined the atomic percentages of

FeNPs that were synthesized using the aqueous fenugreek seed extract as a reducing agent.

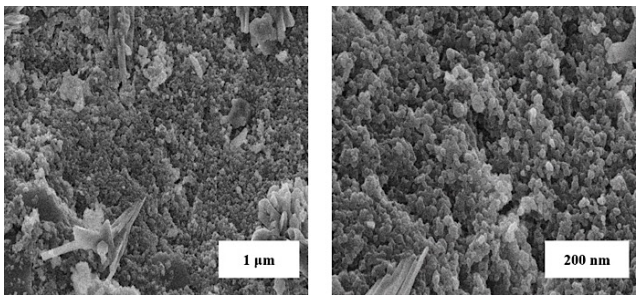

Figure 3: SEM image of synthesized FeNPs at the magnification of 1 μm and 200 nm

C. XRD analysis of iron nanoparticles

The presence of FeNPs powder synthesized by aqueous extract of fenugreek seeds was confirmed by a series of reflection angles (2) at 24.18°, 33.16°, 39.25° and 40.84°, which correspond to phase planes 012, 104, 006 and 113. The XRD spectrum of synthesized FeNPs is shown in Figure 4 and the features of the crystalline metallic iron phase have been established based on these values (Naseem & Farrukh, 2015).

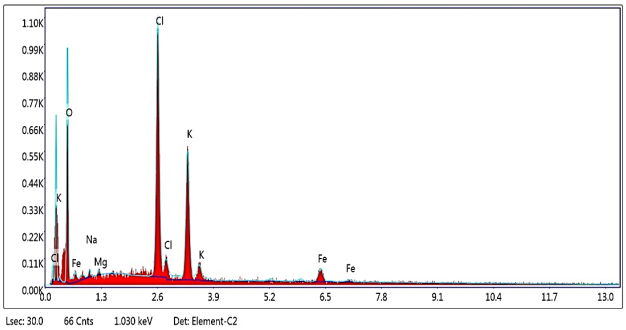

Figure 4: EDX analysis of synthesized FeNPs

D. UV-visible absorption analysis of iron nanoparticles

UV-Visible spectra of green synthesized FeNPs in an aqueous solution is presented in Figure 5. The UV-visible spectral analysis was done within the range of 200-800 nm. The absorption peaks observed were at 250-295 nm and 305-375 nm regions due to the excitation of surface plasmon vibrations in the iron nanoparticles. The maximum peak was observed at 268 nm, which is equal to the characteristic UV-visible spectrum of iron nanoparticles and it was recorded (Pattanayak and Nayak, 2013).

IV. Conclusion

Fenugreek (*Trigonella foenum-graecum*) seeds extract was utilized as a reducing and stabilizing agent for the iron nanoparticle (FeNP) synthesis. The analysis techniques of Scanning Electron Microscope (SEM), Energy Dispersive X-ray (EDX), X-ray diffraction (XRD) and UV-Visible Spectroscopy were used to characterize and confirm the existence

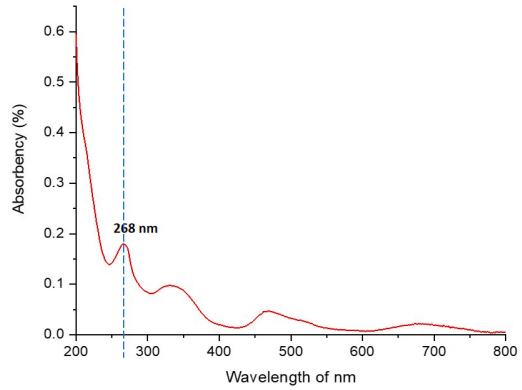

Figure 5: UV-Vis spectrum of synthesized FeNPs

of FeNPs. The particles were determined to be nanoscale based on SEM images and their morphology showed spherical forms with particle sizes varying from 20 to 40 nm. The EDX analysis showed the current atomic percentages with regard to iron and indicated that nanoparticles are primarily present in metal form. The crystalline structure of the synthesized FeNPs were analyzed using the XRD spectrum. The absorption peaks were observed using UV-visible spectral analysis at 250-295 nm and 305-375 nm, with the largest peak being observed at 268 nm, which is identical to the UV-visible spectrum of produced FeNPs. These results verify that the aqueous fenugreek seed extract can be used to synthesize iron nanoparticles. This green synthesis method is natural, fast, eco-friendly, and cost-effective. This is a step toward a more environmentally friendly future in which industrial metal nanoparticle production can be done without the use of harmful reducing and capping chemicals.

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VI. Author Contribution

Rumesh Samarawickrama: writing – original draft, methodology, data curation and visualization. Samudrika Wijayapala: writing – review editing and supervision. Nandana Fernando: writing – review editing and supervision.