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PRODUCTION OF BIOFUEL USING MAGNETIC NANOCATALYST ZNO-NI_{0.5}ZN_{0.5}FE₂O₄-FE₂O₃: A MINI-REVIEW OF RECENT STUDIES

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ABSTRACT

As conventional fossil fuels are becoming limited, it is critical to find alternative clean and renewable energy sources. Biodiesel is a degradable and renewable form of fuel made up of long-chain fatty acid esters that can be generated from either plants or animals. It is a mixture of long-chain alkyl esters made through a transesterification reaction with alcohol and catalyst. Due to the high cost of petroleum, the concept of biofuel production as an alternative source has increased in recent years. The present mini-review summarizes some recent studies on alternative methods for the production of biofuel. The review concludes that biofuel production using magnetic nanocatalysts is one of the best alternative approaches. Magnetic nanocatalysts can easily be recovered from the reaction mixture by using a magnetic field. It reduces the reaction time and increases the yield.

Key words: Biodiesel, esterification, combustion method, wet impregnation, magnetic nanocatalyst.

INTRODUCTION

For up-to-date and developed societies, the availability of reliable and cost-effective energy sources is considered very important. Most advanced world societies rely on fossil fuels such as natural gas and oil to fulfill their energy needs. The energy demand is increasing day by day with the increasing population. To fulfill the increased energy demand use of fossil fuels has become increased which has a very adverse impact on the environment, such as the burning of fossil fuels destroying the environment by emitting many harmful gases. The burning of fossil fuels not only increases the environmental pollution but also the temperature of the earth which causes global warming, the greenhouse effect, and cause acid rain (Saoud, et al., 2018; Voloshin et al., 2019; Alaei et al., 2020).

Since petroleum sources are decreasing day by day with increasing demand, a serious challenge to science and technology has emerged. To fulfill the world energy demand and to reduce the pollution level, many researchers have been working for last many years to find out an alternative way of energy source. Biodiesel has been considered the most excellent elective for petroleum (Ali et al., 2020). Biodiesel has many merits like it is a renewable source of energy, being easily available, low in cost, emits very less amount of gases, and is biodegradable (Galchar, 2017; Voloshin., 2020).

Trans-esterification (TES) reaction produces biodiesel, in the presence of alcohol and catalyst from a mixture of long chain alkyl esters (Alaei et al., 2020). It can be synthesized by different types of feedstock that may be first-generation or second-generation. First-generation biodiesel is obtained from edible vegetable oils including sunflower and palm oil etc. Non-edible oils, such as castor oil and animal manure, etc. are used to make second-generation biodiesel. Biofuel can be classified into different generations based on its feedstock and processing method (Pathaka et al., 2017).

As a biodegradable fuel, biodiesel has attracted the attention of many researchers as an alternative to diesel fuel. The major hurdle in biodiesel synthesis was the high cost of vegetables which is the primary source of biodiesel. So, it was economically disadvantageous and there was a need for the production of biodiesel in acceptable and economically profitable ways. However, this type of feedstock mostly contains a higher amount of free fatty acids (FFAs) that cause a crucial hindrance in the direct conversion of waste into biofuel by the traditional transesterification method. Waste fat and oil contain a significant amount of impurities. It is important to clean the feedstock and convert it into biodiesel using the traditional transesterification procedure to make it appropriate for biodiesel production (Dantas et al., 2018; Ali et al., 2020). These procedures can take place with or without the use of a catalyst but without catalyst it takes more time and energy. Hence, the type and quality of the catalyst directly influence the quality of the final product.

Feedstock Processing

At present, two methods are commonly used for preprocessing feedstock on an industrial scale which are the physical method, and the chemical method. The first step of preprocessing is the refining of material. The purpose of the refining process is the removal of gum from the material, especially the removal of phospholipids from vegetables which is named a degumming process. Hydrated phospholipids are removed during this process and most of the non-hydrated phospholipids can also be reduced to a very low level. Nonhydrated phospholipids include calcium or magnesium salts of phosphatidic ethanolamine and phosphatidic acid. This process is followed by the physical method of refining that is used for the removal of free fatty acid from the feedstock. The physical method involves steam refining using vacuum distillation (Sanek et al., 2015). Biofuel is considered environmental friendly as it does not produce CO₂ and other gases that are produced during the combustion of fossil fuel. Production of biofuel also uses plants and edible crops as feedstock. There is a need for well-planned and controlled manners as uncontrolled and misuse of plants as a biomass source can cause deforestation which directly affects our environment badly. The appropriateness of biofuel as an energy source depends on many factors. It depends on the production process of biofuel, the nature of biomass used, reaction conditions, reaction reagent, and the presence or absence of a catalyst. Biofuel can be used as a direct or indirect energy source. The primary energy source is also called a direct energy source. It may be dried animal dung fuel or wood fuel that can directly use to produce electricity and heat. On the other hand, a secondary energy source or indirect energy source can be obtained from plants or animal origin. It requires very efficient technology for its processing and conversion into biofuel. To move from first-generation biofuel to secondgeneration and reduce environmental pollution and make it economically applicable, biofuel researchers began to speed up their efforts. Two methods can be employed to produce biofuel. It may include homogeneous catalytic processes and heterogeneous catalytic processes. Heterogeneous catalysis is preferred over the homogeneous process because of some advantages. These are: 1) it reduces the corrosion problem as compared to the homogeneous catalytic process. 2) Heterogeneous catalysts can be recovered at the end of the reaction and can be reused. 3) Solid catalyst may be multifunctional and decreases the number of steps involved in the conversion of biomass. In this way, it increases the amount of energy produced and decreases the cost (Dantas et al., 2018). Commonly manufactured biofuels include bio-alcohol, bio-diesel, bio-ether, and bio-syngas (mixture of CO and H₂).

The heterogeneous type of catalyst may be acidic or basic. It can carry out both esterification and transesterification on an equal time however; a homogeneous process can carry out just transesterification (Zuliani et al., 2018; Alaei et al., 2020). During esterification

and transesterification using the methyl and ethyl route, ferrites $Ni_{0.5}Zn_{0.5}Fe_2O_4$ show catalytic behavior. These catalysts have been shown to have an efficiency of about 95%. These catalysts can be synthesized by the combustion method. Zinc oxide (ZnO) shows a conversion rate of 96.20% (Silva et al., 2020; Vieira et al., 2014). All types of organic material obtained from living organisms are called biomass. Biomass includes the organic material obtained from plants, animals, microorganisms, wood, vegetables, algae, etc. The burning of wood as a heat source is known as traditional biomass. The best fuel can be produced by photosynthetic plants (Voloshin et al., 2016). As of late, nanoscale particles pulled into consideration due to their large surface area to volume ratio and show large activity compared to conventional large-scale particles. The activity of a catalyst depends upon the surface area to volume ratio. Specifically, magnetic nanoparticles (MNP) have been introduced for their applications in biofuel production. Studies have shown their higher magnetic activity as compared to regularly used catalysts. The magnetic attraction between the particles creates strong ionic forces which provide them stability.

This mini-review aims to summarize the recent studies on advanced materials with high reactivity, high resistance, recovery from the reaction mixture, less reaction time, good quality, and low cost. Synthesis by combustion method gives the magnetic ceramic powder with nanomaterial characteristics (Dantas et al., 2020; Alaei et al., 2020; Silva et al., 2020). Other processes, such as the pechini method (Gerasimov et al., 2015) and the sol-gel approach, are also utilized to create nanomaterial (Savamoorthi and Raja, 2016).

METHODOLOGY

This review elucidates the different types of magnetic nanomaterial that can be used in the processing of biofuel. To analyze the articles for review, multiple databases were searched including "Google scholar", "Web of Science", "Pub Med", and "sci-hub". To take an initial sample of available articles Google scholar was searched. The broad key term "Production of biodiesel using nanocatalyst" was initially used to establish a list of research articles. More refined terms were then used to search other databases. The keywords selected for full-text articles included, "magnetic nanocatalysts", "Synthesis of magnetic nanocatalyst", "Nanocatalyst for biofuel production", "Best catalyst for biofuel production", and "Characterization of magnetic nanocatalyst". These terms were combined in various ways to obtain the most appropriate articles. Only the recently published articles with full access were included.

Metal Oxide Nanocatalyst for Biofuel Production

A nanocatalyst is a substance that has at least one dimension in the nanoscale with catalytic properties. Nanocatalyst allows more reactants to react because of the large surface area to volume ratio. Biofuel may be classified as bioethanol and biodiesel. Catalysts are classified into two groups, i.e. homogeneous catalysts or heterogeneous catalysts depending on whether the catalyst and reactants are in the same phase or different. The transesterification process is used in the production of biofuel from lignocellulose material as a feedstock in the presence of a catalyst. Catalyst enhances the rate of production of biofuel for biofuel production (Saoud et al., 2018).

Production of Biofuel using a Magnetic Catalyst $ZnO-Ni_{0.5}Zn_{0.5}Fe_2O_4-Fe_2O_3$

Ceramic oxides have been frequently employed as catalysts due to their chemical stability, corrosion resistance, and ability to be recovered and reused. Among various oxides

available, hematite (α Fe₂O₃) is one of the most commonly utilized oxides because it is easy to produce and maintains its stability in a variety of environments. The ferrites-based catalyst was preferred due to its excellent magnetic and electrical properties. It produces biodiesel in an efficient way that is inexpensive, reusable, and can easily be recovered (Farias *et al.*, 2020; Mapossa et al., 2019). Due to the magnetic properties of iron, numerous scientists have been interested in iron-based catalysts with nanoscale size. The catalytic action of the heterogeneous catalyst Fe₂O₄ indicates a conversion rate of more than 94 percent in esters. It was examined utilizing methyl and ethyl for esterification and transesterification. Another heterogeneous catalyst zinc oxide (ZnO) was synthesized by the combustion method. When tested through the transesterification process, it showed up to 80 percent and even up to 96 percent conversion in esters (Mapossa et al 2019; Farias et al., 2020).

The performance of catalysts NiFe₂O₄ and Ni_{0.5}Zn_{0.5}Fe₂O₄ by the transesterification of soybean oil into biodiesel has been investigated. XRD results confirmed the crystalline phases. Total crystallinity was found to be 43 percent with an average crystalline size of 25 nm. The crystallinity varied between 55 percent of NiFe₂O₄ to 72 percent for Ni_{0.3}Zn_{0.7}Fe₂O₄. Hematite was the most abundant crystalline phase, accounting for 55.87 percent, followed by Ni-Zn at 36.96 percent and ZnO at the lowest crystallinity (Mapossa et al., 2019; Farias et al., 2020). Crystallinity varied from 62.5 percent to 72.0 percent, and the average crystalline size was 37 to 20 nm. The magnetic assessment of the ZnO-Ni_{0.5}Zn_{0.5}Fe₂O₄-Fe₂O₃ catalyst confirmed that it reduces the cost of biodiesel production since it can be easily retrieved using an external magnetic field.

Another catalyst studied in recent years was ZnO-Ni_{0.5}Zn_{0.5}Fe₂O₄-Fe₂O₃ with magnetic properties. This is also used for producing biodiesel using the methyl and ethyl route through esterification and transesterification reactions. Results showed high catalytic activity of biodiesel production using ethanol which shows 96.5 percent conversion of esters while methanol shows 92.5 percent conversion. Reaction conditions also affect the conversion of fatty acids. For example, using 3 percent catalyst and 180 °C temperature, it showed a reduction in the acidity of biodiesel by 44 percent while if we increase the temperature to 200 ⁰C and increase the concentration of catalyst up to 5 percent. It showed a greater consumption of fatty acid conversion and caused a percentage of acidity reduction (in ethyl route 88.2 percent to 96.5 percent and in methyl route 84.4 percent to 92.5 percent) (Mapossa et al., 2020). The recovery and re-use of magnetic particles were highly reflected in literature. Magnetic catalyst ZnO-Ni_{0.5}Zn_{0.5}Fe₂O₄-Fe₂O₃ was re-used by using optimum conditions: 30 g oil, time 1 hour, and alcohol to oil ratio was 15:1. By comparing two diffractograms after two re-uses, the catalyst exhibited a 19 percent decrease in activity, but no structural change was observed and it has shown the same thermal stability. (Silva et al., 2019; Dantas et al., 2018; Mapossa et al., 2019; Hashmi et al., 2016; Dantas et al., 2012; Farias et al., 2020; Kumar et al., 2019 and Sivkumar et al., 2013).

Silva et al., (2019) studied on a pilot scale, a magnetic nanocatalyst ZnO- $Ni_{0.5}Zn_{0.5}Fe_2O_4$ -Fe₂O₃ manufactured by combustion reaction method and see whether it could be used to produce biofuel from residual oil. Its conversion rate into biodiesel was about 96.16 percent using ethyl esters through transesterification reactions. Moreover, it showed a long life in recycling and showed sustainable activity even after two cycles. After several cycles, its conversion rate was found to be 95.27 percent, 93.07 percent, and 76.93 percent respectively.

The catalyst was also characterized by its structure, magnetic properties, morphology, and catalytic properties. The catalyst showed a density of 4.8 g/cm³ and a surface area of 52.9 m²/g. The result also showed a mixture of the different phases containing 7.16 percent ZnO, 36.96 percent Ni_{0.5}Zn_{0.5}Fe₂O₄, and 55.87 percent Fe₂O₃ (Table 1).

Dantas et al., (2018) synthesized Ni_{0.5}Zn_{0.5}Fe₂O₄ magnetic nano-particles (MNP) a heterogeneous magnetic nanocatalyst for biofuel production, using the combustion reaction method. A highly efficient magnetic nano-catalyst with soybean oil was used through methyl and ethyl routes for biodiesel production. It showed a conversion up to 99.38 percent \pm 0.18 percent by the ethyl route and up to 99.54 percent \pm 0.16 percent by the methyl route. A simple external magnetic field (magnet) was used for catalyst recovery. Moreover, it has been shown that the catalyst can be used again in 3 more cycles without much loss in its catalytic activities found to be stable enough (Table 1).

The performance of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite was studied doped with 0.1 and 0.4 moles of copper (Cu) respectively for the transesterification of soybean oil to biodiesel. Gas chromatography was used to identify the product of the reaction, which confirmed the conversion to methyl esters. The surface area of the materials doped with 0.1 moles and 0.4 moles of Cu was found to be 22.17 m²/g and 23.49 m²/g while particle size was found to be 50.47 nm and 47.64 nm respectively. The catalyst doped with 0.4 moles of Cu showed better performance with a conversion rate of 50.25 percent more as compared to 0.1 moles. The catalyst with 0.1 moles of Cu produced a conversion rate of 42.71 percent for biodiesel. (Dantas et al., 2012 as indicted in Table 1.

Biodiesel by using nanocatalyst Ferric-Manganese doped with sulfated zirconia (FeMn-SO₄/ZrO₂) from tannery waste was prepared. The production of biodiesel was determined to be 96.6 wt % with methanol and fat. The precise parameters were adjusted at a 12:1 methanol to fat ratio, catalytic loading of 6 wt % at 65 °C, and a stirring rate of 450 rpm for 300 minutes. Furthermore, a study was done to evaluate the performance of the catalyst in the recycling process, which revealed that the catalyst produces a yield of over 90 percent even in the fifth recycling cycle. (Kumar et al., 2019) as indicted in Table 1. Magnesium oxide (MgO) nanocatalyst using the esterification method for biofuel production was prepared. At 1.5 wt % catalyst, with methanol to oil ratio 5:1 at 550 °C, an outstanding conversion result was obtained. It showed 98 percent conversion. (Sivakumar et al., 2013) as indicted in Table 1.

CONCLUSION

By using the combustion method with catalyst ZnO-Ni_{0.5}Zn_{0.5}Fe₂O₄-Fe₂O₃ biofuel is synthesized on a pilot scale. This method of synthesis is found to be secure and proficient. This study shows how the quantity of catalyst and temperature affect the production of biofuel. It was concluded that the catalysts with large surface ranges, great chemical properties, and high thermal stability used for synthesis give an environment-friendly way to produce biodiesel. Microscopy results show that the combustion approach encourages the creation of nanoscale materials with magnetic characteristics, which aid in the separation and recovery of catalysts from the reaction mixture.

AUTHORS CONTRIBUTION

They have made a substantial contribution to the concept or design of this article. They also revised the article critically for important intellectual contents and finally approved the version to be published.

CONFLICT OF INTEREST

The author(s) declare that there is no conflict of interest.

Ziafat et al., (2022). Production of Biofuel using ZnO-Ni_{0.5}Zn_{0.5}Fe₂O₄-Fe₂O_{3.} *J Biores Manag.*, 9(4): 162-168

Table 1: Tabulated summary of some recent studies on the topic included in the study

Reference	Objective	Method	Result
Silva et al., 2020	Production of magnetic nano catalyst with high catalytic activity, corrosion resistance, and chemical stability.	Combustion reaction	The catalyst Fe_2O_3 -Ni _{0.5} Zn _{0.5} Fe ₂ O ₄ -ZnO was created. The pilot-scale production proved risk-free, repeatable, and cost-effective. The catalyst had a high surface area and was ferromagnetic, polyphasic, and nanometric. It was successfully used in the production of biodiesel from residual oil. The catalyst worked in all settings, with conversion rates ranging from 58 percent to 96 percent.
Farias et al., 2020	To determine the impact of the secondary phase and magnetization of the Ni-Zn system on biodiesel conversion and to contribute to the consolidation of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ catalytic viability.	Combustion reaction	The particle obtained was small in size with nanometric characteristics and magnetic properties. Catalytic tests indicated that Ni-Zn ferrite promotes ester conversion of ± 94 percent, and ZnO promotes 83.9 percent. The effectiveness of ZnO was independent of the secondary phase.
Kumar et al., 2019	The reusability of catalysts was tested to see how well they performed during recycling.	Wetness impregnation	The use of a Fe-Mn-SO ₄ /ZrO ₂ nanocatalyst improved biodiesel generation from animal fat derived from tannery waste with a high FFA content. In the ideal circumstances of 12:1 methanol to fat molar ratio, 6 wt% catalysts at 65 °C, and 300 min at 450 rpm, it showed enhanced catalytic activity, yielding 96.6 wt% biodiesel. It was able to convert fat to biodiesel without the need for esterification and can easily be regenerated and recycled up to 5 times.
Mapossa et al., 2019	Production of NiFe ₂ O ₄ and Ni _{0·3} Zn _{0·7} Fe ₂ O ₄	Combustion synthesis is based on the chemistry of thermodynamic	During the transesterification of soybean oil to biodiesel, magnetic nano ferrites achieved a maximum conversion of 94 percent. Findings from this study suggest that the development of nickel-zinc ferrite nanoparticles as magnetic heterogeneous catalysts could provide a biodiesel manufacturing platform that is environmentally benign.
Sivakumar et al., 2013	The use of smoke-deposited nano-sized MgO as a biodiesel catalyst is being investigated.	Esterification reaction	At 1.5 wt% catalyst, an outstanding conversion result was obtained; methanol to oil 5:1 at 550 °C yielded 98 percent conversion in 45 minutes. This conversion was three to five times higher than that reported in the literature for laboratory MgO.
Dantas et al., 2012	To see how well $Ni_{0.5}Zn_{0.5}Fe_2O_4$ doped with Cu works as a catalyst for converting soybean oil into biodiesel.	Combustion reaction	The nanocatalyst $Ni_{0.5}Zn_{0.5}Fe_2O_4$ was more active in the methyl and ethyl esterification reactions than in the transesterification reactions, resulting in a 99.54 percent conversion of fatty acids in biodiesel. Nanoferate doped with 0.4 Cu ⁺² produced the best conversion results. The magnetic characteristics increased the recovery rate of catalyst from reaction mixture.

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