Print ISSN : 0022-2755

Journal of Mines, Metals and Fuels

Contents available at: www.informaticsjournals.com/index.php/jmmf

An Overview of Nano-Catalysts in Biodiesel Production

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Abstract:

Energy consumption and dependence on non-renewable resources is increasing over the years. The combustion of fossil fuels resulting in the emission of substantial amounts of CO_2, NO_X, SO_X and some greenhouse gases. Biofuels are evolving as the primary alternatives to fossil fuels since they can be readily synthesised from discarded bioresources and yield lesser emission during the combustion process. However, the extraction of biofuels has thrown up new challenges that have widened the scope of the use of nano-particles in the synthesis of biofuels. From the literature, distinct findings concerning the use of nano-particles as a catalyst and process reactant during biodiesel production have been identified; this is majorly attributed to the fact that nano-catalysts enhance thermophysical properties, reaction speed and mass transport properties. Henceforth, the present paper aims to review, summarise and provide an insight into the research findings of effectively using nanocatalysts in biofuel production and consider the significance and its relevance for further researchers in the domain of biofuels.

Keywords: *Biodiesel, Nanoparticles, Reaction speed, Mass transport properties.*

1.0 Introduction

Environmental pollution, coupled with the diminishing availability of fossil fuels, is the contributing factors for research on finding alternative sources of energy [1]. The surge in energy consumption is proliferating, and also the factors like change in climatic conditions, the swift decline of inaccessible fossil fuel energy is facilitating the need for cutting edge research in the domain of biofuels [2]. Moreover, there is another primary concern in this modern era, pertaining mainly to the emission of toxic gases from automobile and industrial processes [3 & 4]. Among the major air pollutants, molecular contaminants viz., ozone, carbon monoxide, sulphur, nitrogen oxides, and lead are specified as poisonous pollutants by the World Health Organization (WHO). These pollutants have a detrimental effect on the health of human beings resulting in respiratory and cardiovascular disorders, neuron-related psychiatric disorders, ophthalmological difficulties, soft muscle ailments, and carcinogenic ailments like malignancy, etc.

A relationship has been established between the poisonous air contaminants and the ever-growing rate of diseases in humanity [5]. This problem of air contamination arises due to the incomplete burning of crude oil, necessitating the evolution of sustainable renewable energy resources resulting in cutting-edge research in the domain of biofuels [6 & 7]. Biofuels are a distinct class of fuels derived from biological processes on agricultural and animal biomasses. These biofuels are renewable sources of energy and eco-friendly, especially the sulphur emissions, which are lesser than fossil fuels. However, there are challenges in the synthesis of biofuels. There are necessarily two methods for synthesis of biofuels viz., biochemical and thermochemical processes; among these two methods, thermomechanical techniques of converting the biomass into biodiesel, bio-oil, biogases, bioethanol is of significant significance and facilitates the need for substantial research in the domain of optimising the synthesis process [8 & 12]. The thermomechanical process of biodiesel synthesis includes the most critical step of catalytic liquefaction, which effectively enhances the reaction parameters resulting in optimised yield faster. Thus, the selection of catalysts for the transesterification process of obtaining biofuels is an important aspect that involves a deep understanding of chemicals that can be used as catalysts [13 & 14].

The objective of the present paper is to review and arrive at an understanding of the need for evolving methods of utilising nano-catalysts in the form of zinc oxide, magnesium oxide, calcium oxide, and heterogeneous combination of metal oxides viz., KF–CaO–Fe_3O_4, Au - ZnO.

2.0 Need for Biodiesel

The global scenario of biodiesel production has been increasing since the past decade and has increased, as illustrated in Figure 1 [15]. This tremendous increase in biodiesel production has triggered the keen interests of researchers across the globe, especially in the domain of newer methods of synthesis of biodiesel [16 & 17]. It is widely assumed that the increase in biofuel production will sustain in the coming years, mainly due to the optimised technological innovations resulting from cutting-edge research. The researches aim at broadening the base for supply sources, moderate the effects of instability in the extraction of unrefined crude oil, and reduce the cost involved in biofuel production [18].

3.0 Scope of Biodiesel

The researches in biodiesel have thrown open a broad scope in the domain of the production of biodiesel. The International Energy Agency (IEA) quantifies the yearly rate of increase of production of biofuels. It has inferred that the biofuel production rate will increase by 7% by 2030 and meet nearly 25% of the fuel requirement needs of the automotive industry. This increase in production has prompted an expansion in the interests of researchers in biofuels generation [16 & 19]. In the late eighties and mid-nineties, biofuel production combined the inspiration to synthesise biofuels as a conceivable substitution for non-renewable energy sources, particularly in nations that are overwhelmingly rich in biomass as in countries like Brazil [20]. The extensive scale of the utilisation of biofuels started in Brazil, where bioethanol was first blended with diesel. Around then, the fundamental driver for biofuels development was not the substitution of petroleum products but rather the decrease of uneven characteristics and difficulties encountered during the performance of IC engines [21]. This further triggered interests in the enhancement of properties of biofuels, especially with the entry of developed nations like the USA, which is the world's biggest customer of biofuels. European countries further led the rapid development of bioethanol, expanding the research horizon in biofuels

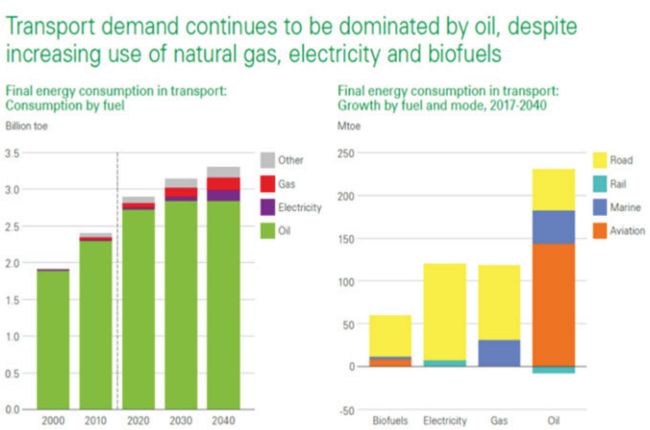

Figure 1: Transport demand depends on oil, despite increasing use of other alternates [15]

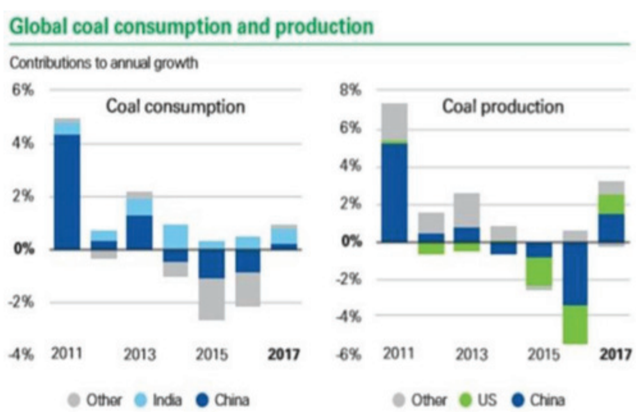

Figure 2: Coal consumption and production in selected countries

[22]. This biodiesel production scope has further widened due to challenges faced by the whole world in coal usage, as in Figure 2 [23 & 24]. This is a significant concern, and essential innovations like the extraction of ethanol from sugar or starch crops, biodiesel from vegetable oil, and robust biofuels from waste or vegetable sources can help humankind overcome several challenges. Still, the problems associated with biofuels and catalytic reactions have extended the scope of research on biofuels.

4.0 Role of Catalysts in Biofuel Production

The role of catalysts in the synthesis of biodiesel is to speed up the chemical reaction of triglycerides like animal fat or vegetable oil and an alcohol group. For speeding the reaction, it requires that the catalyst selected is a potent reaction agent with a strong base or an enzyme. The chemical reaction under the influence of catalyst produces a new compound called biodiesel. This conversion process is transesterification; thus, biodiesels are mono-alkyl esters of long-chain fatty acids [25 & 26]. Since biodiesels are prepared altogether from bio-mass of plant and animal sources, it doesn't contain any sulphur, toxic hydrocarbons, metals or natural petroleum deposits. The non-appearance of sulphur implies a decrease in acid rain by sulphuric acid in the environment. Also, a cetane rating between 49 to 62 is another feature that enhances the performance capability of the engines running with biodiesel. An engine that runs on 100% biodiesel might have no toxic emissions. Thus, it would be more secure to operate the engines; further biodiesel blends have decreased discharges of poly hydrocarbons, which are carcinogenic [27].

Heterogeneous catalysts have accomplished huge significance given their better surface qualities for enhanced reaction rate [28 & 29]. Nano-particles are utilised as a proficient catalyst in bioprocesses to synthesise biofuels [30 & 31]. Nano-material application has resulted in a tremendous revolution in sustainable energy development as it includes green technologies in different procedures. Nano-catalyst tends to enhance the proficiency of transesterification response, leading to increased efficiency in biodiesel production [32 & 33].

5.0 Overview on Biodiesel Production using Nano-catalysts

5.1 ZnO Nano-catalyst

An extensive work on the application of ZnO catalyst in biodiesel synthesis; among them, few are discussed. The biodiesel derived from castor oil with significant levels of fatty acid content using a heterogeneous Ni catalyst doped with ZnO Nano-catalyst is carried out resulted in better catalytic activity. Response surface practises used to enhance the optimised catalysis reactions. By applying RSM, optimum conditions were determined for transesterification as well as three-cycle reusability. More significant catalytic activity was found to be the greater surface area, thus yielding 95.20% of biodiesel as predicted by RSM techniques [34]. The synthesis of biodiesel using ZnO catalyst improves the yield to 96% at isothermal condition with an abridged reaction time of fifteen minutes. The gas chromatography analysis of the obtained biodiesel depicts the effectiveness of ZnO catalysts on conversion triglycerides into methyl esters [35].

Rajesh Madhuvilakku et al. [36] has done extensive work on the transesterification process with palm oil for biodiesel production where a metal oxide in the form of compounds of TiO_2-ZnO complex and ZnO catalyst is effectively employed. The TiO_2 used for the present work exhibits very high catalytic activity. The reaction time is 5 hours at 60ºC and the reaction rate reduced by 8%. The effect of ZnO Nano-catalysts is further analysed by XRD, FT-IR and FE-SEM. The molar ratio of methanol attained was 6 times that of petroleum-based fuel.

G. Baskar et al. [37] also investigated the effect of heterogeneous catalyst like ferromagnetic zinc oxide nano-composite for the production of biodiesel. Castor oil has maximum oil yield potential, high viscosity, stability and density. Nano-particles were used as a catalyst due to their efficiency and the lack of difficulty in separation from the biofuel extract. Iron doped Zinc Oxides generated by the co-precipitation method were applied in the processing of castor oil using methanol. Extreme biodiesel yield was 91% with optimal conditions of 50 minutes of reaction time. The temperature was set at 55 degrees Celsius. Further, there was an improvement in the cold flow characteristics of the biofuels extracted.

5.2 MgO Nano-catalyst

A. Ashok et al. [38] considered the reaction of unused cooking oil with methanol over MgO catalysts by coprecipitation methods. The average dimension of MgO Nano-catalyst was 7.86 nm and was moulded at a calcination temperature of 500ºC. Spectroscopy with the help of Fourier-transformed infrared established the development of the MgO phase with the typical vibrational mode of Mg–O. The yield obtained was 93.3% with 2 wt% of the magnesium oxide Nano-catalyst, 24:1 alcohol to oil molar ratio was obtained, response time was reduced by an hour at a temperature of 65ºC.

Shervin Alaeia et al. [39] have successfully synthesised magnetic MgO catalyst using a combustion method that consumed far less time and cost. A change in the proportion of fuel and nitrates produced a Nano-catalyst of multidomain structure. XRD - showed an increase in fuel ratio increased the crystal size, FESEM, EDX, Dot Mapping, showed that the fuel in a 1.5 proportion has a better active surface area, FTIR, surface particle size circulation is effectively used for characterisation. The conditions used for transesterification are the temperature of 110ºC, a molar ratio of 12, catalyst concentration of 4 wt% for 4 hours. The yield that resulted from the experimentations is 91.2% biofuel from the biomass. The catalyst is effectively used for 5 transesterification reactions. The derived catalyst has many properties such as easy parting, high purity of concluding product and reusability among others. Sumreen Dawood et al. [40] synthesised methyl esters using non-edible oleander seed oil. MgO, which is a heterogeneous catalyst is used for the transesterification process to synthesis methyl ester. The reactants that affect the yield of biodiesel are the alcohol to oil molar ratio, the concentration of catalyst and the temperature.

5.3. CaO Nano-catalyst

Min Shi, Pingbo Zhang et al. [41] showed that CaO-Fe_2O_3 which are a multifunctional and magnetic catalyst, equipped by sol-gel method, exhibited noticeable catalytic actions in biodiesel production. Overall, the usage of KNO_3 is an important element to generate the magnetism of catalyst.

Hamed Nayebzadeha et al. [42] synthesised carbonated alumina doped with Calcium Oxide. The results of BET-BJH, FESEM, XRD, FTIR, EDX, and TG/DSC analysis on five variants of carbon source used for the production of carbonated calcium aluminate showed that dextrose, trimethylene glycol and sorbitol have greater reaction area, pore size and are the more active surface area for reactions to occur. Samples prepared using starch and citric acid showed properties for flash point and auto-ignition temperature. These variants were then impregnated with KOH to increase alkalinity which promoted the conversion of Canola oil into desired biodiesel. The resulting Nano-catalyst was found to have precise pore size and surface area that upgraded its activity. It was also found to have the highest steadiness in the microwave supported transesterification process.

Yatish Kalanakoppal Venkatesh et al. [43] examined the synthesis of CaO nano-particles through solution combustion of crude glycerine as the combustion fuel. Under optimum conditions, 96.2% of biodiesel was obtained. A molar ratio of 9:1 was also obtained, with a reduction in 70 minutes of reaction time.

Libai Wen et al. [44] studied the effect of KF-CaO nano catalyst to prepare biodiesel from Chinese tallow seed oil. China produces 100,000 metric tonnes of oil and it consists of 40 to 70% of fatty acid. Because of its acid value, the oil causes intoxication and saponification problems if transesterification is carried out using NaOH, KOH and CH_3ONa, which are the homogeneous catalyst. The acid value is much higher than rapeseed and soyabean oil, hence facilitating the need for utilising CaO based Nano-catalysts.

Hebbar H.R. et al. [45] explained the optimisation of biodiesel yield from Bombax Ceiba Oil (BOC) through CaO nano-particle as the catalyst. A two-stage transesterification method was used for the production of biodiesel and the percentage of yield was found to be 96.7%. The impact of three transesterification factors is the molar ratio of 10.3:1, CaO-NPS concentration of 1.5 wt% and reaction time of 70.5 minutes.

5.4 Novel Nano-catalysts Al-MCM-41

Neda Vardas et al. [46] has examined the improvement of acidity of MCM-41 by the addition of Al and used the methods to upkeep a Nano-catalyst with the inclusion of calcium for the synthesis of biodiesel. Synthesis of the catalysts was done using 2 methods: impregnation and sono-dispersion. Synthesised models of Nano-catalyst were characterised using XRD, FE- SEM, EDX, TEM, BET and FTIR analysis. The performance of Nano-catalyst was examined under conditions of a temperature of 70ºC, the methanol-oil ratio of 12 and loading of 10 wt%. It was thus found that by increasing the amount of calcium catalysts, the quality of the biodiesel was also increased. Out of the two samples, synthesis using sono-dispersion showed higher conversion for the same loading of Al-MCM-41.

a. TiO_2

Manash Jyoti Borah et al. [47] synthesised in-situ TiO_2/RGO catalyst for the chemical conversion of used cooking oil to generate biodiesel through the process of transesterification. Characterisation was carried out by XRD, FTIR, SEM, EDX, TEM, and TGA.

Optimum operating settings were oil to methanol molar ratio of 1:12 and 65ºC temperature with 1.5 wt% of synthesised catalyst for a reaction time of three hours. The reaction resulted in a 98% yield. Biodiesel was obtained and confirmed by Carbon and Proton NMR, Gas Chromatography-Mass spectroscopy. TiO_2 catalyst was obtained by hydrothermal process. The catalyst can be used for 3 cycles and was established within the ASTM D6751 standards.

Chen et al., reported the transesterification of cottonseed oil at 230ºC for 8 h, using a molar ratio of 12:1 and 2 wt.% TiO_2 Nano-catalyst, from the results, it is seen that the yield of methyl esters is relatively higher at 96.5% as compared to the synthesis of the biofuel using conventional basic catalysts [48].

b. $MgFe_2O_4$

Benedict Iserom Ita et al. [49] investigated the utility of $MgFe_2O_4$ as a catalyst for transesterification of Jatropha curcas whose seeds yield 432 ml which is 36% of the oil without any catalyst. The best combination of parameters was found to be a 7:1 molar ratio of alcohol to oil, 0.5% $CoMgFe_2O_4$ as Nano-catalyst, 60ºC as reaction temperature and 180 minutes as response time. As the methanol oil ratio was increased, unreacted oil settled at the bottom and it significantly improved the properties. The derived biodiesel had a flashpoint of 120 minutes, and a density of 0.899 g/cm^3.

c. Magnetite

Vannia Cristinados Santos-Durndell et al. [50] employed Magnetic Fe_3O_4 or Fe_3O_4/SiO_2 produced by a simple and inexpensive process as a catalyst for the esterification of palmitic acid under the action of methanol under solvothermal conditions without the need for the presence of catalytic species. The catalysts could be used for 4 cycles. The paper revealed the advantage of the Silica coating. Reusability of the derived solids was also found. This catalyst can be used in place of mineral acids for fatty acid transesterification. XRD and SEM analyses showed identical particle distribution in both fresh and reused catalysts.

d. $SrO/CoFe_2O_4$

Milton S. Garcia et al. [51] carried out a study on the magnetically recoverable catalyst of SrO immobilised on $CoFe_2O_4$ support which is used for babassu oil transesterification and was found to be used for up to 4 cycles. Characterisation was executed by VSM, TG, XRD, FTIR, SEM, TEM, and EDS. For the first run, a yield of 96% was obtained. A magnetic stirrer was employed for the co-precipitation method and further modified with SrO immobilisation using a wet-impregnation approach. The best catalytic activity for the methanolysis reaction of babassu oil was observed for the catalyst synthesised in acetone for SrO/CoFe2O4 ratio of 5:1.

e. ZrO_2

Fengxian Qiu et al. [52] have carried out investigations on biodiesel production through chemical transesterification of soybean oil alongside methanol by using ZrO_2 loaded through $C_4H_4O_6HK$. The biodiesel yield is 98.03%. The solid base catalyst used are regenerative and are less corrosive. Hence it is safer and environmenta-friendly. The Nano-catalyst obtained is then characterised by XRD, FTIR, TEM, Hammett indicator method.

f. SO_4/Fe-Al-TiO_2

Jabbar Gardy et al. [53] produced biodiesel from waste cooking oil by transesterification process using a solid acid magnetic catalyst SO_4/Fe-Al-TiO_2. The catalyst used for the transesterification process is characterised by XRD, FTIR method and 96% yield is obtained. The magnetic property of the catalyst helps the process to be stable and recyclable.

g. Sodium Disilicates

Abolfazl Gaffer et al. [54] considered the chemical reaction and subsequent conversion of rapeseed oil to the methyl ester using methanol. It is carried out by using phases of layers of sodium disilicates. The catalysts are characterised by XRD, FTIR, FESEM, TGA-DTA, N_2 adsorption-desorption. The percentage yield of the process is found to be 97.8%.

h. $Ni0.5Zn0.5Fe_2O_4$

Joelda Dantas et al. [55] have effectively employed the catalyst spinel nano ferrites $Ni0.5Zn0.5Fe_2O_4$ for

Table 1: List of nano catalysts for biodiesel production

	Catalyst used	Biodiesel Yield	No. of Reaction Cycles	Reaction Time (h)	Reaction Temperature (°C)	Molar ratio
1	ZnO	93.5%	3	2.03	57.5	6:1
2	MgO	92.7%	5	2.5	88.3	
3	CaO	96.4%	5	1.17	60	
4	Al-MCM-41		4	8	70	12:1
5	TiO_2	98%	3	3	65	1:2
6	$MgFe_2O_4$	92%		3	60	7:1
7	Magnetite	97%	4	6	120	12:1
8	$SrO/CoFe_2O_4$	96%	4	3	60	5:1
9	ZrO_2	98.03%		2	60	16:1
10	SO_4/Fe-Al-	96%	10	2.5	90	10:1
11	TiO_2 Sodium Disilicates	93.7%	5	3	65	30:1
12	Hydrotalciteparticles	95.2%	8	1.5	45	3:1

ethyl and methyl esterification reaction of acidified soyabean oil and cottonseed. The spinel nano ferrites are synthesised by combustion and are characterised by XRD.

i. Hydrotalcite Catalysts

Xin Deng et al. [56] produced biodiesel from jatropha oil which uses the catalyst Hydrotalcite-particles with magnesium and calcium which are prepared by co-precipitation technique using urea as precipitating mediator and it is microwave-hydrothermally treated, followed by calcination. Maximum biodiesel yield by utilising the catalyst is found to be 95.2%.

6.0 Conclusions

From the literature, it is significantly inferred that, by using Nano-catalyst for biodiesel production, there is an increase in cycle repeatability. Also, higher reaction rates and product yields are achieved as a result of greater availability of surface area. Also, the catalyst recovery is noticeably higher for Nano-catalysts as compared to conventional catalysts. Further, it is observed from the review of the literature that the Nano-catalysts exhibit lower reaction time as compared to conventional catalysts at lower temperature ranges. Further, it is critically observed from the literature that by using ZnO Nano-catalysts, the average biodiesel yield is around 93.5%, the number of reaction cycles is restricted to 3 and the response time is around 2.03h, On the other hand, biodiesel produced using MgO Nano-catalyst is 92.7%, the number of reaction cycles is 5 and the response time is 2.5h. Better catalytic action is obtained with CaO Nano-catalyst where it yielded around 96.4% of biodiesel, number of reaction cycles as 5 and the reaction time reduced to 1.17h. Among the various novel Nano-catalysts, it is found that TiO_2, ZrO_2 and SO_4/Fe-Al-TiO_2 produced a maximum quantity of biodiesel with an average of 96% yield. The number of reaction cycles for SO_4/Fe-Al-TiO_2 is 10 which is the highest among all the Nano-catalysts. The reaction time is also less for SO_4/Fe-Al-TiO_2, when compared to its equivalents. All the reactions are accomplished in the temperature range of 60-80°C. It can therefore be concluded that Nano-catalysts are suitable for synthesis for biodiesel. There is a need to carry out comparative experimental analysis using various Nano-catalysts for particular biodiesel to effectively study the reaction time, cycle repeatability and mass transport. The conclusions are compiled and tabulated in Table 1.

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