iJRASET
International Journal For Research in Applied Science and Engineering Technology

# INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

**Volume:** 4 **Issue:** III **Month of publication:** March 2016

**DOI:**

www.ijraset.com

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Volume 4 Issue III, March 2016
ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

# Hydro processing Of Bio Oils, Effect Of Parameters – A Review

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***Abstract--Rising energy demands and growing environmental concerns coupled with decline in petroleum reserves has stimulated the growth of bio fuels, an alternative to conventional fuel. This uses renewable oil derived from biomass as feedstock for biodiesel production. Hydroprocessing technology is one of the routes to produce biodiesel and the process is catalyzed by means of an hydrocracking catalyst. Various research works has been conducted in this technology around the globe. The biodiesel produced from hydroprocessing technique is more commonly referred to as "Green diesel" or "Renewable diesel". The current review gives a clear picture on hydroprocessing of edible and non-edible oil for biodiesel production and also effect of various operating parameters on conversion and diesel yield.***
***Keywords--Hydroprocessing, Green diesel, Renewable diesel, Conversion and diesel yield.***

## I. INTRODUCTION

Biodiesel is a biofuel obtained from triglycerol containing biological sources (most commonly vegetable oil or animal fat). Conventional biodiesel production uses transesterification process where the bio-oil is converted to biodiesel using alcohols. The products obtained are monoalkyl esters (biodiesel) and glycerine. In chemical composition, biodiesel seems to be inferior to mineral diesel fuel (Petrodiesel). This led to the development of second generation fuel technology for biodiesel production. The biodiesel produced from second generation technology is more commonly referred as "Renewable diesel" or "Green diesel"[1]. They refer to petrodiesel-like fuels derived from biological sources. These fuels are not chemically esters and hence distinct from biodiesel. The chemical composition of renewable diesel is very close to mineral diesel fuel (Petrodiesel). The term "renewable diesel" will refer to all diesel fuels derived from biomass that meet the standards of ASTM D975 and are not mono-alkyl esters.

## II. RENEWABLE DIESEL (HYDROPROCESSING) VERSUS BIODIESEL (TRANSESTERIFICATION)

As mentioned earlier the properties of FAME obtained from transesterification is very inferior to properties of petrodiesel. On the other hand the properties of renewable diesel obtained from hydroprocessing technique is very much close to the mineral diesel. The below table gives the comparison of properties of renewable diesel, biodiesel and petrodiesel

| Properties | Petrodiesel | Biodiesel | Renewable Diesel |
|---|---|---|---|
| Cetane# | 40-55 | 50-65 | 75-90 |
| Energy Density, MJ/kg | 43 | 38 | 44 |
| Density, g/ml | 0.83-0.85 | 0.88 | 0.78 |
| Energy Content, BTU/gal | 129 K | 118 K | 123 K |
| Sulfur | <10 ppm | <5 ppm | <10 ppm |
| NOx Emission | Baseline | +10 | -10 to 0 |
| Cloud Point, C | -5 | 20 | -10 |
| Oxidative Stability | Baseline | Poor | Excellent |
| Cold Flow Properties | Baseline | Poor | Excellent |
| Lubricity | Baseline | Excellent | Similar |

Table 1 comparison of properties of petrodiesel, biodiesel and renewable diesel [2]

Although FAME has been used as diesel fuel for many years, the use of FAME is connected with some problems as compared to the use of petroleum-based diesel fuel. FAME damages some construction materials of the fuel system. Renewable diesel overcomes these limitations efficiently and acts as a better alternative for petrodiesel. Hydroprocessing is a petroleum refinery operation and hence existing refineries itself can be used to carry out hydroprocessing of bio oils.. For example Finland's company Neste- Oil has completed the construction of a unit for vegetable oil hydroprocessing in Porvoo (Finland). The unit with capacity 170 kt/year has two stages: hydroprocessing of vegetable oils and isomerisation of obtained hydrocarbon product to improve its low temperature properties [3,4,5]. NesteOil also declared that the emissions of $CO_2$ in this process are lower than emissions coming from


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production of FAME [6]. The coprocessing of biodiesel with petrodiesel also becomes easier as both the petrodiesel and biodiesel can be produced simultaneously in the same hydroprocessing unit and it coprocessing vegetable oils with high vacuum oil improves process yield. For example, a 5 wt.% sunflower oil to 95 wt.% HVO produces the maximum theoretical straight chain C15–C18 yield of 87%.

## III. CATALYTIC HYDROPROCESSING

Catalytic hydroprocessing constitutes a modern way to produce premium quality bio-based diesel fuels, that are also referred toas ‘‘renewable diesel fuels’’ instead of the term ‘‘biodiesel’’, which has prevailed for FAME. In Hydroprocessing hydrogen is introduced to the feedstock in the presence of a catalyst to remove other atoms such as sulphur, oxygen and nitrogen to convert triglyceride molecules into paraffinic hydrocarbons. Hydroprocessing of biomass for biodiesel production has gained much attention[7] because of the following reasons:

The chemical composition of the hydroprocessed fuel oil is very much close to mineral diesel oil and hence it can meet the requirements of the automotive industries better than diesel obtained from transesterification.

It can be used efficiently at low temperatures.

It has higher heating value and cetane ratio.

Lower capital and operating costs.

Various vegetable oils (both edible and non edible oils) and animal fats are used as feedstocks for hydroprocessing. First generation biofuels uses edible food crops and second generation fues uses non edible food crops owing to their economical feasibility. Recently third generation fuels that uses algae has feedstock has emerged due to the following due to the following advantages [8]:

Algae does not come under food crops and hence it is much economical.

Microalgae has higher biomass and lipid productivities, a faster growth rate than higher plants.

Requires less area almost ten times lesser than that used for other plants and also very less amount of water is required for its growth.

### *A. Hydroprocessing-Process description*

It is typically a hydrotreatment technique that is used in petroleum refining operation[9]. This type of treatment basically involves four steps

Feed preparation

Hydrotreatment reaction

Product separation and

Fractionation.

Fig 1 represents the process flow diagram of hydroprocessing of bio-oil.

In the feed preparation section the liquid biomass feedstock is mixed with the high pressure hydrogen (mainly from gas recycle with some additional fresh make-up hydrogen) and is preheated before it enters the reactor section. The reactor section consists normally of two hydrotreating reactors, a first guard mild hydrotreating reactor and a second one where the main hydrotreating reactions take place.



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Volume 4 Issue III, March 2016
ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

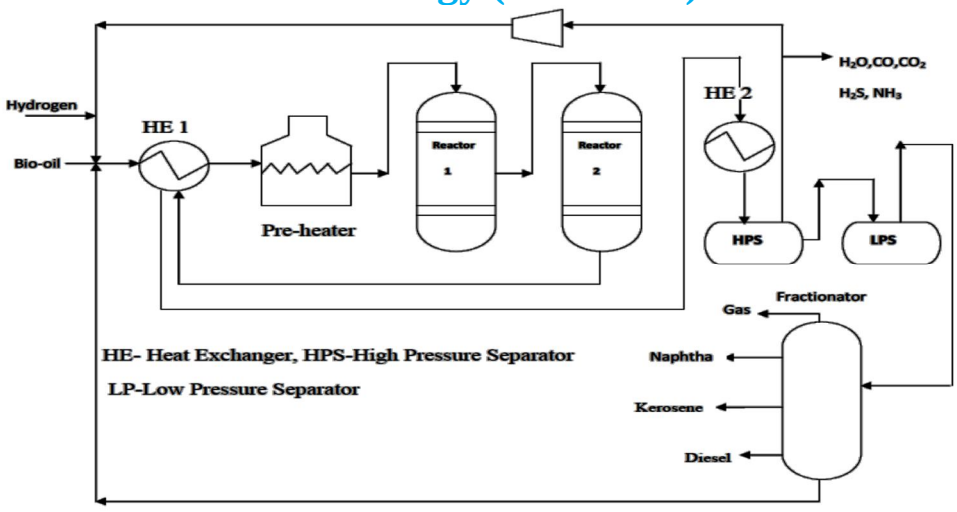


Fig 1. Hydro processing of bio-oil [9]

Each reactor contains two or more catalytic beds in order to maintain constant temperature profile throughout the reactor length. The product then enters separators where it is cooled and the gas and liquid products gets separated. The fractionator section provides the final product separation into the different boiling point fractions that yield the desired products including off-gas, naphtha, kerosene and diesel. The heaviest molecules return from the bottom of the fractionator into the reactor section as a liquid recycle stream. Catalysts for hydroprocessing are bi-functional in nature and performs dual functions: 1. cracking and 2. hydrogenation-dehydrogenation. The cracking is enhanced by an acidic support whereas the hydrogenation-dehydrogenation is imparted by metals. Some of the acid supports used include alumina, zeolite and amorphous oxides. The metal sites used can be either nobel metals like platinum, palladium, ruthenium, rhodium or transition metals like nickel, molybdenum, tungsten, cobalt, titanium. The most commonly used catalysts include CoMo, NiMo based catalysts such as NiMo/$\gamma$-$Al_2O_3$ or CoMo/$\gamma$ -$Al_2O_3$, sulfided CoMo and NiMo on alumina base, Pt/HZSM-5, Ni/HYzeolite noble metals and their alloyed catalysts including Pt, Pd, Ni, Sn, Pd/C, Ru/C on acidic support. Zeolite based hydrocracking catalysts have greater acidity, better thermal and hydrothermal stability, low coke forming tendency and easily regenerability

*B. Hydroprocessing-Reaction Mechanisms*

Several types of reactions takes place during catalytic hydroprocessing of biomass based on the type of biomass used, operating conditions and also the catalyst employed. The hydroprocessing reactions that take place during hydroprocessing of vegetable oils is as follows:

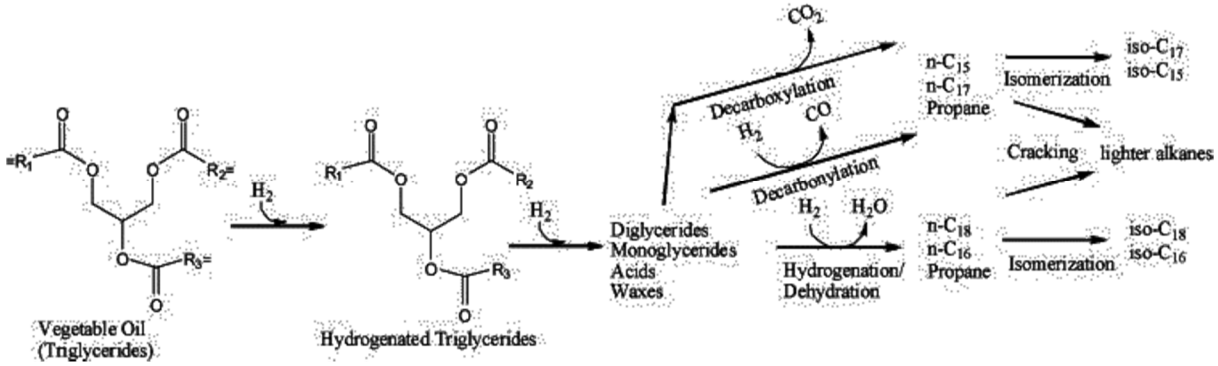


Fig 2 Hydroprocessing reactions-Hydroprocessing of vegetable oil[10]

*1) Reactions:*Cracking – Breaking of larger molecules to molecules of size similar to that of conventional fuels and their boiling range is also smililar to that of conventional fuels.


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Saturation-Introduction of hydrogen breaks the double bonds and converts to single bond thus saturating unsaturates.

Heteroatom removal- Removal of heteroatoms other than Carbon and Hydrogen. This include oxygen, nitrogen, Sulphur

Isomerization- converts Straight Chain paraffinic compounds to branched chains. This is required to improve cold flow properties of fuel.

*C. Hydroprocessing-Operating Parameters*

Operating parameters of hydroprocessing operation are

Temperature
Pressure
Liquid hourly space velocity
Hydrogen feed rate-Hydrogen/oil ratio.

*1) Effect Of Temperature:* Temperature is selected according to the type of catalyst and feedstock used. Increasing the temperature increases the conversion. Increase in temperature beyond optimum value will increase the conversion but may decrease the diesel yield. This is because the hydrocarbons formed, cracks to further lighter fractions, thereby dramatically reducing the diesel product. The effect of temperature on conversion and diesel yield for crude palm oil is shown in fig 3 [11].

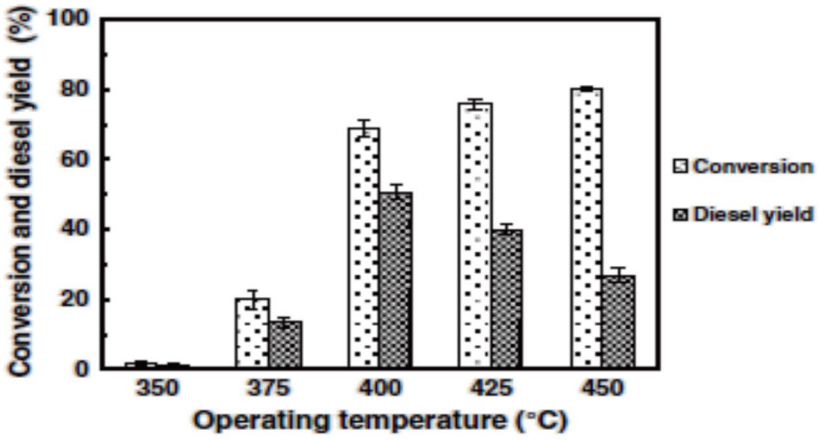


Fig 3 Effect of temperature on conversion and diesel yield for crude palm oil

Similar effects were observed in soybean oil with sulphided cobalt-molybdenum (Co-Mo) and nickel-molybdenum (Ni-Mo) on alumina base catalysts. Fig 4 shows the effect of Temperature on diesel yield for Soybean oil[12]

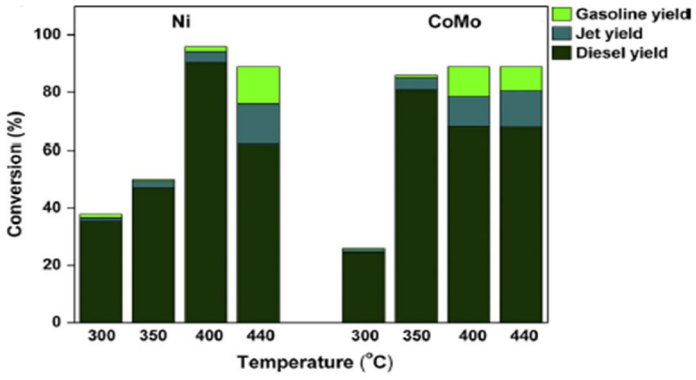


Fig 4 Effect of temperature on conversion for soybean oil

Effect of temperature on fresh and used cooking oil is shown in Fig 5. The conversion of both fresh and used cooking oil increased


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Volume 4 Issue III, March 2016
ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

with increase in temperature but fresh cooking oil showed slightly higher conversion than used cooking oil[13].

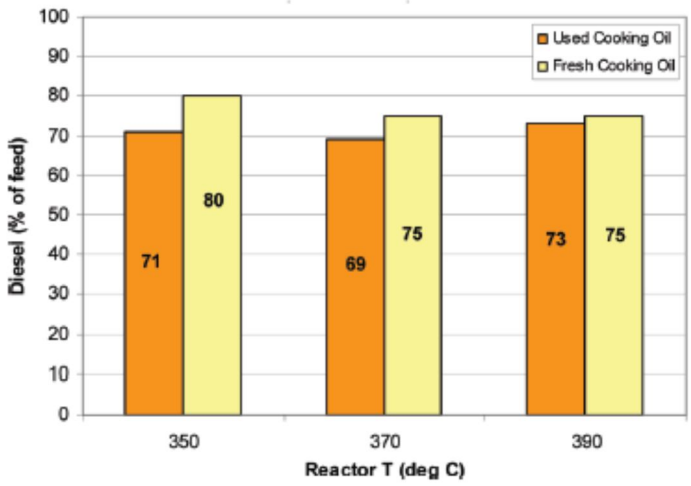


Fig 5 Effect of temperature on conversion for used and fresh cooking oil

Isomerization reactions are favoured at higher hydrotreating temperatures. Increase in temperature, decreases the paraffin content of the product decreases while increases the isoparaffin content. The effect of temperature on isomerisation reaction in case of waste cooking oil is represented in Fig 5[14]

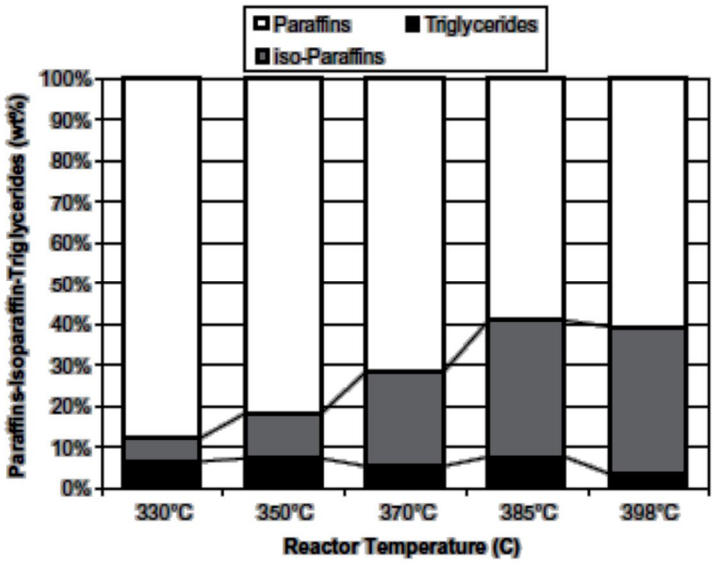


Fig 6 Effect of temperature on hydrocarbon content [14]

*2) Effect Of Hydrogen Pressure:* The pressure has significant control over equilibrium of dehydrogenation-hydrogenation reactions that takes place on the metallic sites. An increase in pressure for a given molar ratio H2/feed increases the conversion of aromatic structures to saturated products. This inturn improves the quality of jet fuel, diesel fuel and oil with very high viscosity index. Increasing the pressure increases conversion and diesel yield. But beyond optimum operating pressure conversion increases but diesel yield decreases. The effect of pressure on conversion and yield is similar to the effect of temperature. Effect of pressure on soybean oil using Ni and CoMo catalyst is shown in Fig 6[12]


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Volume 4 Issue III, March 2016
ISSN: 2321-9653


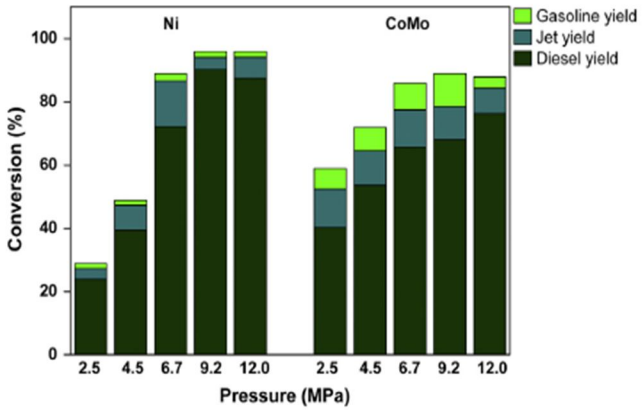


Fig 7 Effect of pressure on conversion for soybean oil

For crude palm oil using Pd/C catalyst by increasing the pressure from 20 to 60 bar, CPO conversion continually increased whereas diesel yield increased to the maximum value at 40 bar and then decreased at higher pressure. Therefore, to obtain reasonable diesel yield from hydroprocessing of CPO, The most suitable operating pressure was found to be 40 bar at 400˚C with reaction time of 3 hours. Fig 7 shows the pressure effect on yield and conversion of crude palm oil[11].

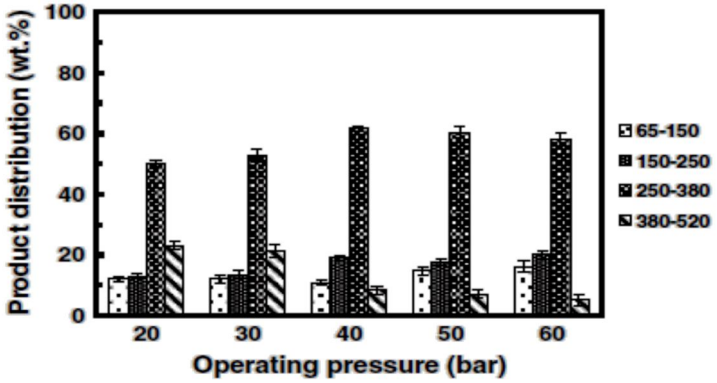


Fig 8 Effect of pressure and conversion on crude palm oil using Pd/C catalyst

The pressure effect on rapeseed oil using Ni-Mo/γ-$Al_2O_3$ at 350˚C is shown in fig 8.

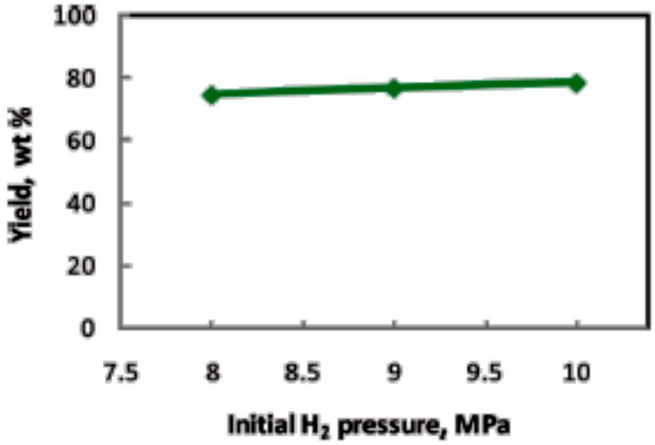


Fig 9 Effect of pressure on yield for rapeseed oil [15]

*3) Effect Of Hydrogen Feed Rate And Hydrogen/Oil Ratio:* Hydrogen feed rate has a significant influence on hydroprocessing


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ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

reactions. Higher hydrogen feed rate favors saturation and heteroatom removal. Since hydrogen feed rate adds to operating cost, optimization of hydrogen feed rate is necessary. An increase in hydrogen/oil ratio increases cetane number and calorific value. Beyond optimum value of hydrogen/oil ratio, there is no significant change in cetane number and calorific value. For hydroprocessing of chlorella using Ni-Co-Pd/γ-Al2O3 the maximum cetane value was obtained at hydrogen/oil ratio of 120, beyond which there was no significant change in cetane value. Fig 9 shows the effect of Hydrogen/Oil ratio on cetane number for Chlorella[15].

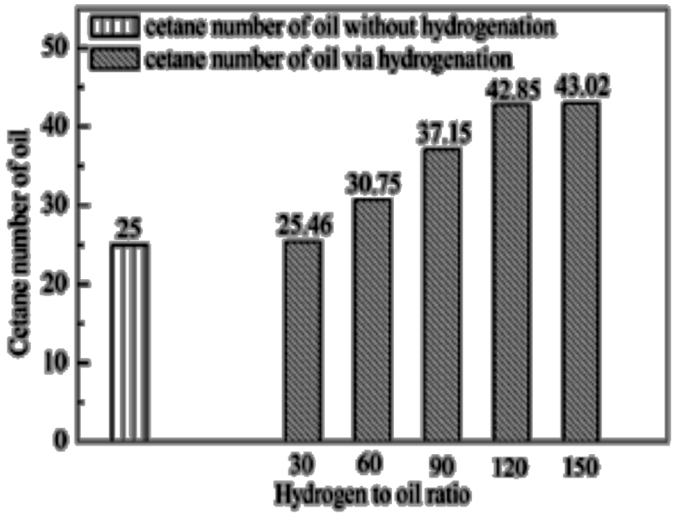


Fig 10 Effect of hydrogen/oil ratio on hydroprocessing of Chlorella [16]

*4) Effect Of Liquid Hourly Space Velocity/Weight Hourly Space Velocity:* Liquid hourly space velocity (volume basis) or Weight hourly space velocity (weight basis) greatly affects conversion.Higher liquid hourly space velocity, the less time is available for the contact of feed molecules resulting in lesser conversion. For used cooking oil it was found that as temperature increases the conversion decreases from 80% (at LHSV = 0.5 $h^{-1}$) to 74% (at LHSV = 2.5 $h^{-1}$). This conversion decrease for increasing LHSV is anticipated since high LHSVs result in smaller reaction time. Fig 10 shows the conversion and selectivities at various liquid hourly space velocities.

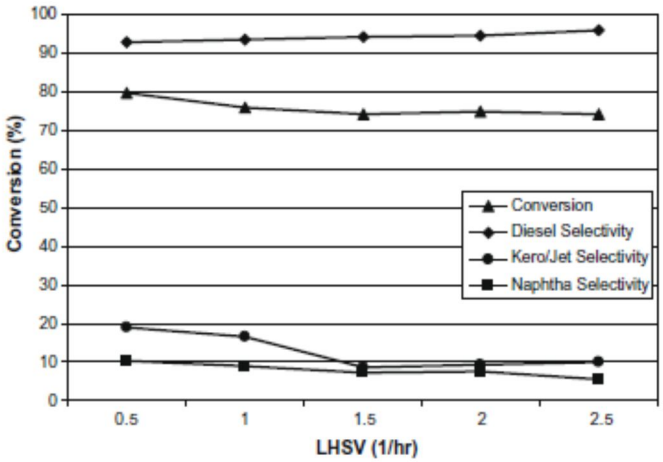


Fig 11 Conversion and selectivites for various LHSV's for used cooking oil [17].

The effect of Weight hourly space velocity on sunflower oil-Gas oil blend for various catalysts is shown in Fig 12[18]


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Volume 4 Issue III, March 2016
ISSN: 2321-9653
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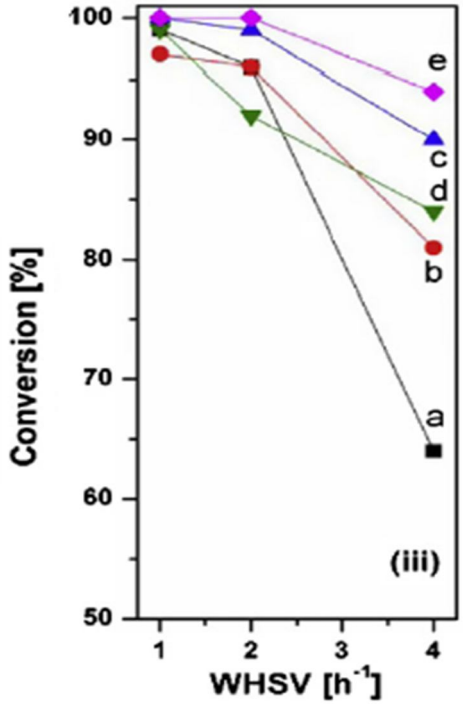


Fig12 Effect of WHSV on conversion for sunflower oil-gas oil blend : (a) 20% oil–diesel blend; Ni–Mo–Al; (b) 20% oil–diesel blend, Ni–Mo–15BEA; (c) 20% oil–diesel blend, Ni–Mo–30BEA; (d) 40% oil–diesel blend, Ni–Mo–30BEA; (e) 20% oil (+10% oleic acid)–diesel blend, Ni–Mo–30BEA.

*4) Effect Of Residence Time*: Incresing the residence time of reaction increases the conversion to a certain extent, but beyond the optimum residence time, the diesel cut products undergoes further cracking and reduces the diesel yield. For crude palm oil using Pd/C the optimum reaction time was found to be 3h beyond which there is decrease in diesel yield[4]

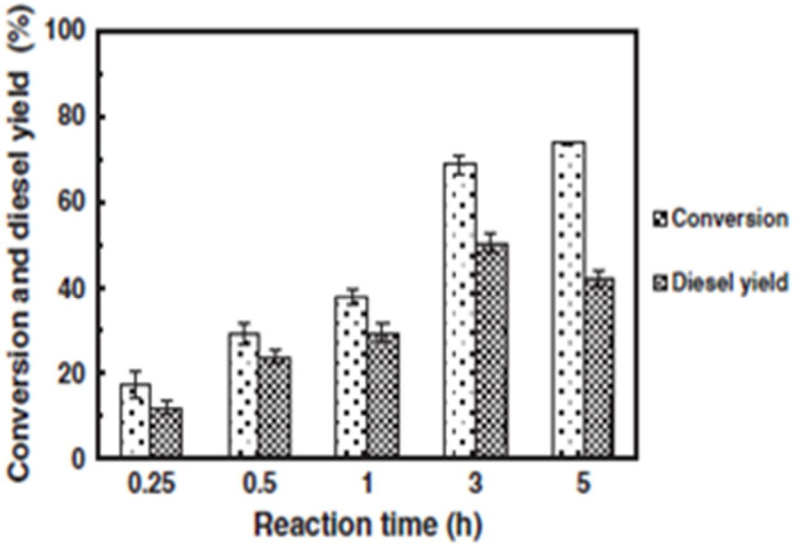


Fig 13 Effect of reaction time on conversion and diesel yield for crude palm oil


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Volume 4 Issue III, March 2016
ISSN: 2321-9653


| Feedstock | Catalyst used | Operating conditions T (°C), P (MPa), and WHSV ($h^{-1}$) , Time (h) | Reference |
|---|---|---|---|
| Palm Oil | 1. Ni-Mo/γ- $Al_2O_3$<br>2. Pd/C | T=400℃ , P=2-6 bar, time=3h | [11] |
| Soybean Oil | 1. sulphided cobalt-molybdenum (Co-Mo)<br>2. sulphided nickel-molybdenum | T=400℃, P=0.9-1 bar<br>T=400℃ P=1-1.2 bar | [12] |
| Canola Oil | $Mo_2N$/γ.$Al_2O_3$ | T = 400, P = 8.3, WHSV = 0.9, H2/oil = 810 L/L | [19] |
| Canola Oil | NiMo/γ.$Al_2O_3$ | T = 300−400, P = 1.8−8.5, batch | [20] |
| Olive-oil derived byproduct | Sulfide CoMo/meso-silicates | T = 250, P = 3, WHSV = 2.7 | [21] |
| RSO | NiMo/$Al_2O_3$ | T = 260−280, P = 3.5, WHSV = 1−4, H2/oil = 50 mol/mol | [22] |
| RSO | NiMo/$Al_2O_3$ | T = 260−340, P = 7, WHSV = 1.0 | [23] |
| RSO | NiMo/$Al_2O_3$, Mo/$Al_2O_3$ Ni/$Al_2O_3$ | T = 260−280, P = 3.5, WHSV = 0.25−4.0 | [24] |
| SBO | NiMo/γ.$Al_2O_3$ | T = 350−400, P = 1−20, batch | [25] |
| SBO | NiMo/γ.$Al_2O_3$ | T = 360, P = 14, batch | [26] |
| SBO | NiMo/γ.$Al_2O_3$, Pd /γ.$Al_2O_3$, CoMoS /γ.$Al_2O_3$,Ni/SiO2-Al2O3Pt/γ.$Al_2O_3$, Ru/γ.$Al_2O_3$ | T = 400, P = 9.2, batch | [27] |
| SFO | NiMo/γ.$Al_2O_3$/F | T = 280−380, P = 2−8, LHSV = 0.75−3, H2/oil = 400−600 N m3/m3 | [28] |
| SFO | Pt/HZSM-22/Al2)3Pt/SAPO-11 | T = 280−380, P = 3−8, WHSV = 1−4, H2/oil = 250−400 N m3/m3 | [29], [30] |
| SFO | CoMo/Al2O3 | T = 380, P = 4−6, WHSV = 1.0, H2/oil = 500−600 N m3/m3 | [31] |
| SFO | Pd/SAPO-31 | T = 310−360, P = 2, WHSV = 0.9−1.6, H2/oil = 1000 N m3/m3 | [32] |
| SFO | Commercial hydrocracking catalyst | T = 360−420, P = 18 | [33] |
| WCO | NiMo/B2O3-Al2O3 | T = 300−350, P = 7, batch | [34] |
| WCO | Commercial hydrocracking catalyst | T = 330−398, P = 8.3, WHSV = 1.0, H2/oil = 4000 scfb | [14] |
| WCO | Commercial hydrocracking catalyst | T = 350−390, P = 13.8, WHSV = 1.5, H2/oil = 6000 scfb | [17] |

Table 2: Operating conditions for hydroprocessing of various vegetable oils

## IV. TWO STEP HYDROPROCESSING ( HYDROCRACKING+ISOMERISATION)

Hydroprocessing can be carried out either in single stage (hydrocracking) or two stage (hydrocracking+isomerisation). Several research activities in laboratory- and pilot-scale have also targeted on optimizing lipids conversion via catalytic hydroprocessing, most of which however have focused on the first hydrotreatment step[17-19]Isomerisation improves cold flow properties of the fuel and thereby increases the hydroprocessing efficiency. The first step of hydrotreatment converts the triglycerides of the lipid feedstock into an oxygen-free hydrocarbon- based fuel consisting of primarily normal paraffins, which even though has an


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impressively high cetane number, it also has degraded (high) cold flow properties. The product of the first step then undergoes a second hydroprocessing step where isomerization/ dewaxing enables the conversion of normal paraffins into iso- and cyclo-paraffins, leading to an improved fuel with sufficiently low cold flow properties (20–30°C) lower than that of the product of the first hydroprocessing step) [35]. This type of two step hydroprocessing is preferable particularly in cold climate, where the isomerisation unit addresses the cold filter plugging point limitations. For waste cooking oil using NiMo catalyst the effect of isomerisation is depicted in the graph below

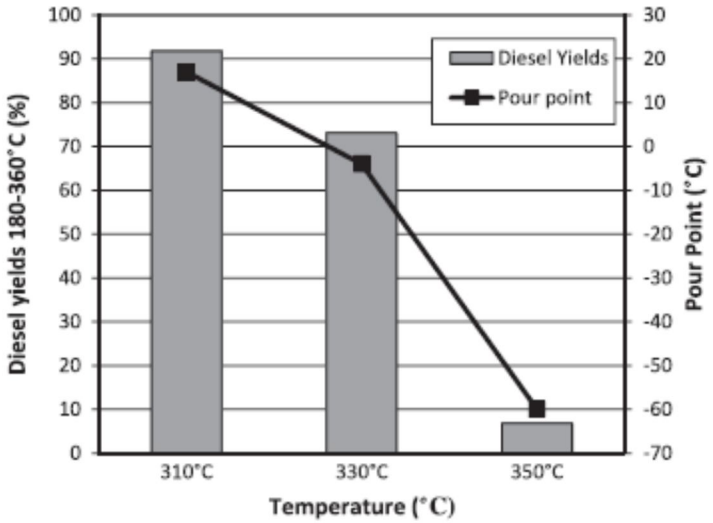


Fig 11 Effect of isomerisation temperature on diesel yield and pour point for waste cooking oil [36]

## V. CONCLUSION

Hydroprocessing technology for biodiesel production proves to be a better alternative for petrodiesel. The hydroprocessing operation, reaction mechanisms, factors affecting hydroprocessing, their effects and the superiority of hydroprocessed biodiesel over fatty acid methyl esters prouduced through transesterification has been described in this work. The research works are now mainly focussed on improving and modifying the existing catalysts so as to improve the process and to improve the yield and selectivity of diesel. However intensive studies are still required to produce commercialize the process on industrial scale.

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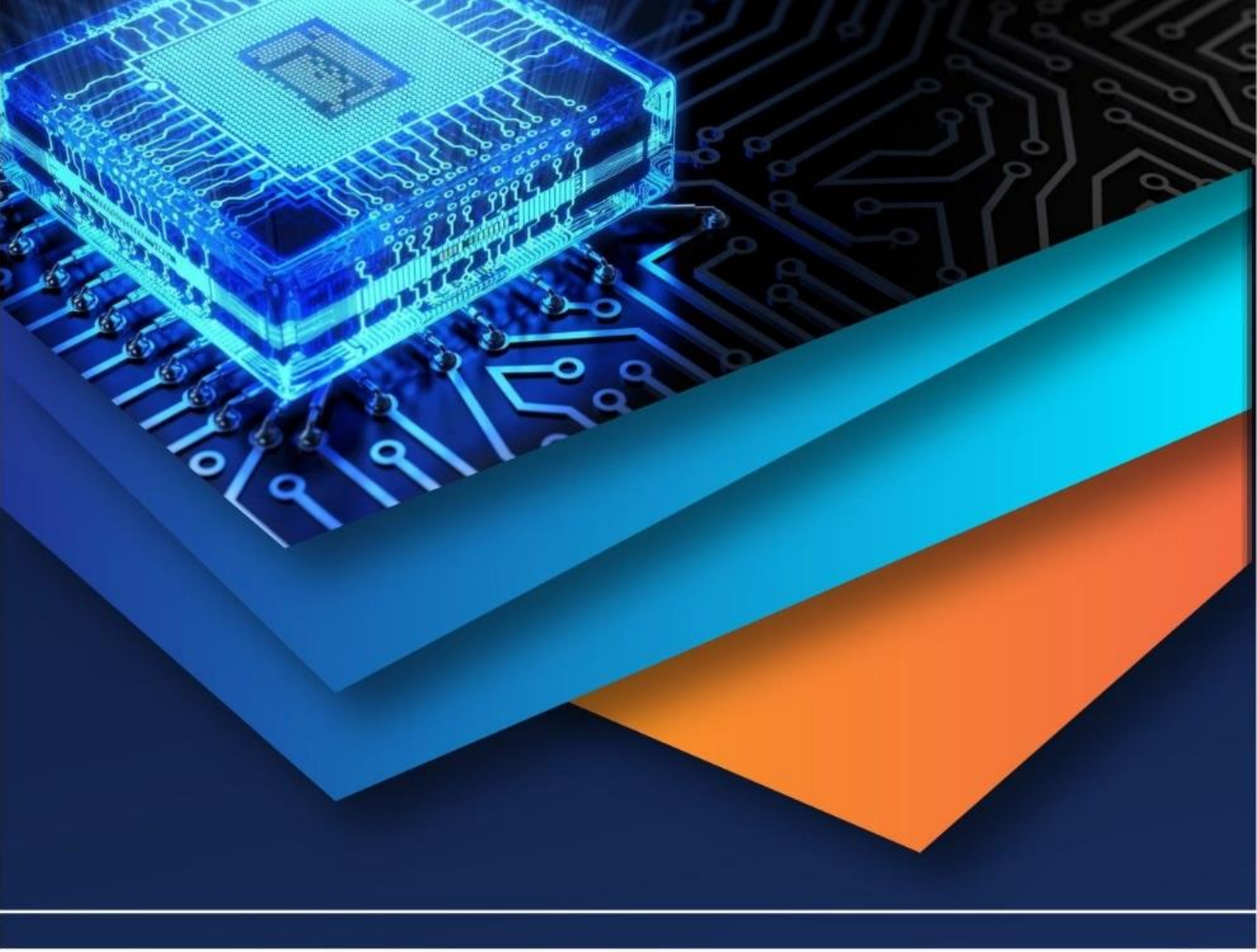

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