



# **RETRACTED:** Recent Development of Renewable Diesel Production Using Bimetallic Catalysts

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Chia SR, Nomanbhay S, Ong MY, Chew KW, Khoo KS, Karimi-Maleh H and Show PL (2021) Recent Development of Renewable Diesel Production Using Bimetallic Catalysts. Front. Energy Res. 9:769485. doi: 10.3389/fenrg.2021.769485 Renewable diesel as a potential sustainable energy source in future requires catalysts to convert the feedstocks into end products. Among various type of catalysts, bimetallic catalysts are widely applied in the renewable diesel production due to their unique catalytic properties and enhanced catalytic activities, which differ from their parent monometallic catalysts. This mini review comprised of the brief introduction on technologies in producing renewable diesel and aims to discuss the underneath knowledge of synergistic interactions in bimetallic catalysts that synthesized through various techniques. The novelty of this review reveals the recent development of renewable diesel production, highlighting the mechanisms of bimetallic catalysts in the enhancement of the catalytic activity, and exploring their possibilities as practical solution in industrial production.

Keywords: renewable diesel, bimetallic catalyst, synergistic effect, bioenergy, catalytic process

# INTRODUCTION

Increment of population, industrial activities and hazardous gas emissions has urged the world to explore potential solution in shifting main energy sources to renewable energy due to the huge energy demand (Mofijur et al., 2013a; Peter et al., 2021). The introduction of renewable diesel (RD) as fossil fuel replacement is attractive due to its similarity of chemical structure with petroleum diesel. RD is producible using biodiesel feedstocks such as microalgae, date seeds, *Jatropha curcas* and lignocellulosic biomass (Mofijur et al., 2013b; Bharath et al., 2021; Low et al., 2021; Oladipupo Kareem et al., 2021). However, the production often involves catalytic processes that are governed by catalysts.

Noble metal catalysts such as Pd and Pt are applied due to high selectivity to deoxygenation reaction, but their expensive and scarcity have limited their availability for commercial scale of RD production. The rapid deactivation of pure Pd catalysts hinders the practical application in conversion process (Cheah et al., 2020). Hence, an efficient and cost-effective catalyst leading to the high yield of end products with long lifespan and reusability is utmost important in the current industries.

The combination of noble metals with low-cost metals forms bimetallic catalysts that can enhance the catalytic properties of catalysts during the conversion process. Such catalysts are relatively cheaper with promising catalyst efficiency. Bimetallic catalysts are prominent catalysts as high selectivity, activity and stability are observed to yield high hydrocarbons content and purity (Malins,

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2021). The synergistic effects between two metals are the major factor controlling the catalyst's efficiencies, which mainly depends on the types of metals.

Numerous reviews have discussed about the recent development of RD processes and Ni-based catalysts in producing RD, but less reviews highlighted the comprehensive mechanisms of bimetallic catalysts in current RD development even technical investigation on bimetallic catalysts are plenty. The aim of this mini review is to highlight the current status of bimetallic catalysts in RD production with their respective interaction with and without support, revealing the suitability of bimetallic catalysts in RD production from recent studies.

# RENEWABLE DIESEL PRODUCTION TECHNOLOGIES

The production of RD is achievable through thermochemical processes and biochemical process. Vegetable oil and animal fats are subjected to hydroprocessing or pyrolysis while lignocellulosic biomass undergo pyrolysis, Fischer-Tropsch (FT) process, biological conversion of sugars (i Nogué et al., 2018; Okeke et al., 2020). Among the technologies, hydroprocessing is performed directly on natural triglycerides to obtain RD, which involved a series of complex reactions. Major reactions produce the hydrocarbons,  $C_{15}-C_{18}$ are hydrodeoxygenation (HDO), decarboxylation and decarbonylation, depending on the type of catalysts and reaction conditions (Veriansyah et al., 2012). It was reported that the sonochemically treated catalysts have better triglycerides conversion compared to conventional synthesized catalyst and high selectivity of diesel range hydrocarbons was observed (Ameen et al., 2017). Hydroprocessing allows fuel with lower NO<sub>x</sub> emissions, better oxidation stability and higher energy density as compared to the transesterification of fatty acid methyl esters (Šimáček et al. 2010).

Besides, the pyrolysis of feedstocks produces bio-oil, biochar, and non-condensable gases, which bio-oil requires upgrading to become RD (Bharath et al., 2020a; Bharath et al., 2020b). The biooil can be upgraded through HDO, catalytic cracking and supercritical fluids to overcome the limitations of bio-oils as hydrocarbon fuel. However, each process possesses particular constraint, such as shorter catalyst lifespan for catalytic cracking, high hydrogen pressure requirement for HDO and high cost of solvent for supercritical fluids (Panwar and Paul, 2021).

On the other hand, the syngas obtained from gasification of lignocellulosic biomass is subjected to FT process to obtain RD. The optimization of gasifying agent to biomass ratio is important to attain optimum yield of RD production and overall energy consumption (Im-orb et al., 2015). Conversion of sugars to RD requires the assistance of microorganisms such as oleaginous yeast to produce lipid while feeding on the provided lignocellulosic sugars. The work of i Nogué et al. (2018) presented biological-catalytical hybrid process to yield 31 yeast strains using corn stover hydrolysate. The produced lipids undergo mild acid treatment and extraction, hydrogenation and isomerization to convert into RD blendstocks from *R. toruloides* as a suitable strain (i Nogué et al., 2018). Generally, catalysts are involved and aid in the accomplishment of the RD production.

# BIMETALLIC CATALYSTS FOR RENEWABLE DIESEL PRODUCTION

Monometallic or bimetallic catalysts can be used to speed up reaction rate and maximize yield. As how its named, monometallic catalysts consist only single type of metal and usually act as the parent metal for bimetallic catalyst. The common types used are Ni, Pd, Pt, Rh, Co, Fe, and Ru. Specific catalysts are required to boost the reaction activity or negative output may observe if inappropriate catalysts are used. Undesired side reaction that influences main reaction pathway due to unsuitable catalyst has to be avoided such that Ni leads to methanation in FT process (Martinelli et al. 2020). Therefore, bimetallic catalysts are introduced to overcome such issue and enhance the efficiency in catalytic processes

Bimetallic catalysts with two metal elements exhibit distinct properties from their respective parent monometallic for physical, chemical, electronic, and photonic properties (Suryawapsui et al., 2018). Compared to monometallic catalysts, they are advantageous due to synergistic effect, morphostructures, and their performances varied with the synthesis method (De et al., 2016). Besides, the distinctive and new properties may generate in bimetallic catalysts despite the combined properties of respective monometallic catalysts. The lectronic configuration, oxidation state, surface composition etc, of bimetallic catalysts have distinguished their uniqueness and benefits over monometallic catalysts as reported in literatures (Alshammari et al., 2016). Robustness of bimetallic catalysts is being studied intensively to discover their applications not only in energy production, but environmental remediation and biomedical as well (Al-Haddad et al., 2020). The wide application of bimetallic catalysts in various fields show the potential beneath and more studies are required to commercialize the usage of bimetallic catalysts in RD production.

# Synergistic Enhancement

Bimetallic catalysts are known for their synergistic effect that leads by combinations of various metals to a new feature of catalyst. The synergistic enhancement often categorized into geometric (ensemble) and ligand (electronic) effects. When the second metal (which added afterwards) modifies the electronic configuration of first metal, ligand effects occurred (Yang and Koel, 2018). This phenomenon occurs due to the changes on the electronic environment of metals, leading by the formation of heteroatoms. The altered electronic environment subsequently modifies the electronic configurations of metals and hence, the chemical properties of catalysts.

As the geometry of bimetallic catalysts are commonly differed from their parent metals, the distinction properties such as average metal-metal bond length change, change in surface bond lengths and angles are often observed (De et al., 2016; Li and Tsang, 2018). Types of geometric structures for bimetallic

nanoparticles are known as core-shell, Janus, crown-jewel, alloyed and hollow structures (Alshammari et al., 2016). In additional, the induction of lattice distortion, expansion or compression caused by alloying metals with dissimilar sizes often observed in core-shell structures (Li and Tsang, 2018). The main application of crown-jewel structure is to jewel the atoms of expensive metals at the crown of low-cost metals for using precious metals effectively. For Janus or metals with side-by-side structures, the presence of both metals on the metal surface are attractive compared to other structures. Janus structure is achievable through seed-mediated growth and three types of shapes can be generated by optimizing the experimental condition (Lyu et al., 2021). Hollow structure outstands other structures due to its high surface to volume ratio while the two metals in alloyed structure is homogenously distributed, produced under cautious monitored reaction kinetics (Alshammari et al., 2016).

The synergistic effect of bimetallic catalyst is the key factor in controlling the catalytic reactivity of the catalysts. The fundamental of the enhanced effect can be explained by the d-band theory, where the energy of adsorbate binds to the metal surface is depending on the electronic structure of surface mostly (De et al., 2016; Lee et al., 2021). Formation of bonding  $(d-\sigma)$  and anti-bonding  $(d-\sigma)^*$  is obtained through the hybridization of metal d-band with bonding orbital  $(\sigma)$  of the adsorbate. In bimetallic system, the extension of filling for  $(d-\sigma)^*$  state is subjected to the electronic structure, like surface density of state for the metals on the surface when the state of  $(d-\sigma)$  is full. Higher catalyst activities can be generated through weaker binding resulted by destabilization of metal-adsorbate interaction as the filling of  $(d-\sigma)^*$  state is increased (De et al., 2016).

## Synthesis Methods

Bimetallic catalysts are synthesized through impregnation, strong electrostatic adsorption, colloidal synthesis etc (Xhong and Regalbuto, 2013; Cho and Regalbuto, 2015; De Coster and Poelman, 2021). Optimization of synthesis methods is required to maximize catalytic activity as it is the key factor in manipulating catalytic properties of bimetallic catalysts.

Impregnation is reported as a simple and established technique to synthesize heterogeneous bimetallic catalysts. It is classified into incipient wetness and wet impregnation to synthesize supported catalysts. The volume of metallic solution is equal to pore volume of support for incipient wetness while higher volume of metallic solution is required in wet impregnation (Alshammari et al., 2016). Incipient wetness tends to produce poor metal dispersion with agglomerated and bigger alloyed particles that has weak metals' contact (Cho and Regalbuto, 2015). Strong electrostatic adsorption is advantageous as catalysts with smaller metal particles and higher dispersion were produced compare to impregnation (Cho and Regalbuto, 2015). Electrostatic interactions are involved in electrostatic adsorption while some system involves both electrostatic and chemical interactions, depending on the type of metal precursor and support used (Ewbank et al., 2014). Core-shell configuration is achievable using sequential electrostatic adsorption where the metal loading of shell is controlled by optimizing the cycles of electrostatic adsorption, showing the flexibility in altering catalyst composition (Cho and Regalbuto, 2015).

Besides, colloidal synthesis was reported to allow specific control on the distribution of metal nanoparticles on support, size distribution and size of particles compare to conventional methods of supported heterogenous catalysts (Quinson et al., 2018b). Optimization of the particles' size is achieved through separating the steps of catalysts preparation and deposition into single stages to prevent the contribution of support on catalysts formation (Quinson et al., 2018a). Enhanced structural and thermal stability was observed in the promoter-rich colloidal bimetallic catalysts via test under 800°C. The obtained result has showed potential in avoiding catalyst deactivation by presynthesizing the uniform particles and removing the extra promoter oxides (Escorcia et al., 2020). In electroless deposition, chemical agent is utilized to coat the metal ion or complex on substrate and reduced them chemically (Rao and Trivedi, 2005). This method was claimed to overcome the issues like difficulties in limiting the degree to optimize specific interactions, isolating the himetallic effects, high energy demand and high waste to product ratios (Gould et al., 2015; Egelske et al., 2020). Past study have reported that higher surface coverages of Ag and Au on Pd particles' surface are achieved and lead to better selectivity of reaction conversion (Zhang et al., 2014). However, the analysis on low and high temperature regimes have indicated the thermal instability of bimetallic catalyst at high temperature (Egelske et al., 2020).

Dividation reduction reaction is utilized for bimetallic satalysts with noble metals to reduce the usage of expensive metals and enhance the efficiency of noble metals (Zhao et al., 2020). The surface reaction is controlled to maximize the interactions between two metals. Deposition of the second metal on monometallic parent catalyst is performed through reducing parent catalyst using hydrogen at known temperature, adding pre-dissolved copper solution into the suspension of reduced parent monometallic catalyst, and obtaining synthesized catalyst after stirring and filtration (Epron et al., 2002). Besides, the synthesis of bimetallic nanoparticles is performed via top-down (laser ablation) or bottom-up technique (co-reduction, seeded-growth, and anodic dissolution) as illustrated in Figure 1. Top-down is a technique that disassemble larger objects while bottom-up is performed by metal cations reduction (Loza et al., 2020). The presence of reducing agent is necessary for the bottom-up method. Co-reduction occurs when metal precursors of both metals are presented and having close reduction potential, leading to alloyed configuration (Bhol et al., 2020). Seededgrowth is performed through sequential reduction for coreshell configuration and anodic dissolution is conducted through the deposition of a metal on another firstly dissolved metal to form hollow configuration. Laser ablation is performed using laser beam on the target immersed in liquid solution to form alloyed configuration, where surfacefunctionalization and fractionization can be further conducted on well-dispersed bimetallic nano-particles (Zhang et al., 2017; Loza et al., 2020).



## **TABLE 1** | Bimetallic catalysts with supports for RD production.

Type of bimetallic	Type of support	Synthesis method	Application (reaction)	Reference
Pd-Sb	SiO <sub>2</sub>	Reduction-oxidation	Dehydrogenation	Ye et al. (2021)
Mo-W	Biochar	Wet impregnation and reduction	Hydrotreatment	Tran et al. (2020)
Co-Mo	AC	Impregnation	Deoxygenation	Gamal et al. (2020)
Ni-Mo	$\gamma$ -Al <sub>2</sub> O <sub>3</sub> or SAPO-11	Incipient wetness	Hydro-process	Lin et al. (2021)
Ir-ReO <sub>x</sub>	SiO <sub>2</sub>	Sequential impregnation	HDO	Liu et al. (2018)
Pt-MoO <sub>x</sub>	ZrO <sub>2</sub>	Wet impregnation	HDO	Janampelli and Darbha, (2019)
Ni-La	AC	Wet impregnation	Deoxygenation	Khalit et al. (2020)
Ni-Mo	γ-Al <sub>2</sub> O <sub>3</sub>	Sonochemical	HDO	Ameen et al. (2019)
Pt-Re	Carbon support	Incipient wetness	Hydro-thermal deoxygenation	Jin and Choi, (2019)
Ni-Mo	SiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub>	Impregnation	Hydrotreatment	Malins, (2021)
Pd-Pt	γ-Al <sub>2</sub> O <sub>3</sub> -H-β	Wet impregnation	Hydrogenation and	Palanisamy and Kandasamy, (2020)
		· ·	hydrogenolysis	
CaO-La <sub>2</sub> O <sub>3</sub>	AC	Impregnation	Deoxygenation	Alsultan et al. (2017)
Mo-Ni	$\gamma$ -Al <sub>2</sub> O <sub>3</sub>	Incipient wetness	Catalytic upgrading of pyrolysis oil	Melo et al. (2021)
Pt-WO <sub>x</sub>	ZrO <sub>2</sub>	Wet impregnation	Deoxygenation	Janampelli and Darbha, (2021)
Ni-Mo	Mesostructured y-alumina	Incipient wetness co-	Continuous hydrotreatment	Afshar Taromi and Kaliaguine, (2018)
	support	impregnation	-	<u> </u>
Ni-Co	SBA-15	Wet impregnation	Deoxygenation	Kamaruzaman et al. (2020)
Pd-Cu	AC	Incipient wetness	Dehydrogenation	Cheah et al. (2020)

## **Performances of Bimetallic Catalysts**

Bimetallic catalysts can be synthesized into catalysts with and without support. Those without support are rarely applied as unsupported catalysts have lower surface areas compared to supported catalysts. For such reason, most studies utilized supported catalysts in RD production. Yet, the performance of new generation NEBULA unsupported catalysts with high active site density is convincing and has been commercialized due to their outstanding catalytic activity compared to common supported catalysts (Eijsbouts et al., 2007; Burimsitthigul et al., 2021). It was reported that NiMoS<sub>2</sub> allowed 95–100% conversion of palmitic and oleic acids through HDO in obtaining high yield and selectivity of C16 and C18 (Yoosuk et al., 2019).

On the other hand, support is used to increase catalytic activity of reactions. Advantages such as higher catalysts' stability was achieved as the metal are well-dispersed on the support, governed by the interaction between metal catalysts and support material (Ferreira-Aparicio et al., 1998). The presence of support is important to reduce cost of precious metal catalysts and allow better metal recovery (Parapat et al., 2014). Reuse of precious bimetallic catalysts with maximised catalysts recovery is achievable using support. However, type of bimetallic catalysts and support influence the synergistic interaction and requires optimization for optimum catalytic properties.

From **Table 1**, bimetallic catalysts in most studies are synthesized through impregnation, either incipient wetness or wet impregnation. Ni-Mo bimetallic catalysts synthesized by deposition-precipitation impregnation are reported to have better performance as higher yield and selectivity to n-C18 were obtained compared to incipient wetness and double incipient wetness (Malins, 2021). However, the sonochemical synthesis is claimed to be more effective than impregnation to generate active sites of Mo, enhancing physicochemical properties like higher specific surface area and uniform distribution of metal particles on support's surface (Ameen et al., 2019).

Bimetallic combinations are investigated in HDO, deoxygenation or dehydrogenation reactions. Ni is widely studied as it is comparatively cheaper, yields high hydrocarbon yield and triglycerides conversion by sulfiding itself and other metals with  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> as support (Wang et al., 2014). Types of bimetallic combinations and support are key parameters to optimize. It was concluded Ni-Co is more suitable to synthesize with  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> compare to SiO<sub>2</sub> as higher conversion and diesel selectivity were obtained (Hari and Yaakob, 2015). The support,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> reported to work better with Ni-Mo than SAPO-11, despite the second metal is unsimilar with the previous work (Lin et al., 2021). Activated carbon (AC), biochar, carbon support in other forms or metals can be utilized as support too.

## CONCLUSION

Bimetallic catalysts are proven to be efficient catalyst in RD production with the flexibility, robustness and unique properties that are not possets by monometallic catalysts. Variation of metal combinations with different support favours specific reactions. Exploration of bimetallic catalysts for RD production in more studies is required, to further enhance the catalytic properties and activities in increasing the efficiency of catalyst. Conventional bimetallic catalysts have their

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advantages, but more studies of novel bimetallic catalysts are required to aid in the improvement of this field. Further investigation is required to fully control the final configuration of bimetallic catalysts through synthesis methods and reduce the possibilities of sites deactivation especially for precious bimetallic catalysts. Expenses on catalysts can be lowered through enhancing the recyclability and stability of catalysts to achieve sustainable, economical, and efficient process in practical application.

## **AUTHOR CONTRIBUTIONS**

SC and SN contributed to conceptualization and visualization. SN, PS and HK-M supervised the progress of manuscript. SC wrote the first draft of manuscript. MO, KC and KK helped review and editing the manuscript. SN provided funding for the manuscript. All the authors contributed to manuscript revision, read, and approved the submitted version.

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