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Catalytic Deoxygenation of Hydrolyzed Oil of Beef Tallow Over Lanthanum-Embedded HZSM-5 Zeolite Catalyst to Produce Biofuels

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Abstract: The catalytic deoxygenation of triglycerides and fatty acids removes oxygen atoms in the process of producing biodiesel and thus hydrocarbons with improved fuel properties. Lanthanum-supported HZSM-5 zeolite catalysts were prepared with lanthanum loadings ranging from 2% to 7%. The best result was achieved with 2% La loading, yielding 72% bio-oil with a heating value of 34 MJ/kg at 300°C. The incorporation of lanthanum significantly enhanced the catalytic activity and stability of HZSM-5 zeolite for deoxygenation reactions. The highest hydrocarbon selectivity (up to 89%) and best coke resistance (8% carbon deposition) were observed with 5% La-HZSM-5. These results highlight the optimal balance between acidity modulation and structural integrity at 2% La, while 5% La provides superior coke resistance and hydrocarbon selectivity.

Keywords: HZSM-5 , Lanthanum, Beef tallow, Doxygenatio, Biofuels

Introduction

The process of catalytic deoxygenation of hydrolyzed oil from, a beef tallow, is part of a novel mechanism for converting bio-oils into more energy-dense and stable biofuels. In this study, HZSM-5 zeolite catalysts with lanthanum incorporated were employed to promote hydrocarbons formation during oxygen-rich hydrolyzed oils conversion, thus, tempering the fuels characteristics (Riyadhi et al. , Žula et al., 2022).

One of these improvements for biofuels is catalytic deoxygenation, a key biofuel upgrading process that converts the high oxygen content of bio-oils (the main product of biomass pyrolysis) to more stable hydrocarbons that have lower corrosivity (Nuhma et al., 2022). This process is remarkable for its two-pronged benefits: not only is it a biofuel generator, but it also implements carbon recycling, since it is recycling beef tallow: associated slaughterhouse byproduct, organic waste reduction. Optimizing the catalytic deoxygenation process can improve the quality of biofuels for intended industrial applications. Lanthanum incorporation into zeolite framework efficiently improves catalytic activity; this has undone all past shortcomings of biofuel production from organic waste sources (Liu et al.,2022; Wu et al., 2024).

The deoxygenation mechanism operates differently under hydrothermal conditions compared to organic solvents, primarily due to the unique physicochemical properties of the reactants and the reaction environment (Luo, Cao et al. 2019, Liu, Guo et al. 2022). This mechanism involves various steps that lead to the removal of oxygen from the hydrolyzed oil, ultimately resulting in the formation of stable hydrocarbons suitable for use as biofuels. The catalytic deoxygenation of triglycerides and fatty acids removes oxygen atoms in the process of producing biodiesel and thus hydrocarbons with improved fuel properties. There are three principal pathways for this process: hydrodeoxygenation (HDO), decarboxylation (DCO₂), and decarbonylation (DCO):

- In **hydrodeoxygenation (HDO)**, oxygen is removed as water by reacting to the feedstock with hydrogen:

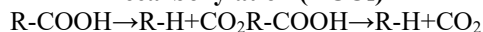
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- **Decarboxylation (DCO₂)** eliminates oxygen as carbon dioxide, requiring little or no hydrogen:



- **Decarbonylation (DCO)** removes oxygen as carbon monoxide, also with minimal hydrogen:



The pathway chosen depends on the catalyst and the reaction conditions. HDO is favored in hydrogen-rich environments, while DCO/DCO₂ is favored in hydrogen-lean environments. These steps convert fatty acids to long chain alkanes, ideally as renewable diesel or green fuels, while improving the quality and stability of the resulting biodiesel.

Lanthanum-embedded HZSM-5 zeolite catalysts are a significant step forward for the catalytic deoxygenation of biomass-derived oils into biofuels. The lanthanum-modified medium-pore zeolite exhibits corresponding catalytic activity enhancements due to modifications in its acidic sites that promote, to the extent of conversion, the hydrolyzed oils into hydrocarbons. (Nuhma et al., 2022). Imparting lanthanum ions into the zeolites' framework lets the active sites facilitate desired reactions-incorporating decarboxylation, cracking, decarbonylation, aromatization, and isomerization. Besides, greater hydrocarbon yield and heating value (HHV) of the formed biofuels due to this modification were more efficient and longer-lived compared to their standard catalysts like CoMoS/Al₂O₃ (Yang, 2017). Modification of HZSM-5 by lanthanum added to the better processing of the structural properties of the catalyst, i.e. enhanced external surface area and, acidity; and coke formation reduction during catalytic processing. Lanthanum facilitated oxygenated species removal from fatty acids, thereby converting these directly into hydrocarbons and yielding biofuels of superior quality and stability (Szkudlarek, Chalupka-Śpiewak et al. 2024). Further, the lanthanum-impregnated HZSM-5 catalyst has proven very effective in attaining the highest hydrocarbon yields in bio-oils from biomass and is thus a really attractive candidate for green diesel production(Nuhma, M.J., et al 2021 & Di Vito Nolfi, G., K. Gallucci, and L. Rossi,2021). Experimental studies had indicated that lanthanum-modified HZSM-5 worked effectively in stainless steel reactors at 300°C for a period of 60 minutes, allowing for high yields of hydrocarbons and the prolonged performance of the catalyst(Nuhma, et al .,2022; Riyadhhi & Susanto, 2022). The further addition of cerium to this catalyst system in the (7.5%La-2.5%Ce)/HZSM-5 composition has yielded maximum hydrocarbons and, therefore, presents itself as a candidate for industrial application (Nuhma et al., 2022). This innovation is key to addressing energy shortages and reducing greenhouse gas emissions, which establishes the role of advanced heterogeneous catalysts in sustainable biofuel production.

The synthesis of lanthanum-modified HZSM-5 catalyst employs several other parameters in addition to the percentage of lanthanum loading. These include impregnation conditions such as temperature and duration of impregnation (Liu et al., 2023). Characterization techniques for the synthesized catalyst comprise, for example Scanning Electron Microscopy (SEM), Energy-dispersive X-ray Spectroscopy (EDS), and Infrared Spectroscopy (IR) in determining the structural as well as chemical properties of the synthesized catalyst(Gujar, Guda et al. 2009). Thus, studies have shown that lanthanum effectively enhances the acid sites of HZSM-5 framework, needed to enhance the catalytic conversion potency for algal oils (Nuhma, Alias et al. 2022). The remarkable catalytic activity seen in the lanthanum-modified HZSM-5 is thus due to efficient conversion from crude oil to hydrocarbon fractions, and testing the optimal conditions for catalysis identified whether the newly produced catalyst (6 wt% La/HZSM-5) demonstrated lower reaction conditions than several other catalysts. In addition, the catalyst is indicated to have much higher stabilities and reusability in several cycles in the transesterification process, thus indicating great promise for future industrial applications in biofuel generation (Jeswani & Chilvers et al. 2020).

This means that the studies on the la-HZSM-5 catalyst will be completed within the purview of a wider scope of work that aims at fossil-fuel replacement by renewable energy sources. This is increasingly important because of the finite nature of fossil fuels and the urgent necessity to advance technologies for renewables (mostly biofuels) produced through catalytic processes (Di Vito Nolfi, Gallucci et al. 2021). Most importantly, the oil catalytic deoxygenation routes provide a sustainable method of biofuel generation in accordance with future demands for catalysts that must be durable and cost-effective (El-Araby 2024).

The aim of the research is to design a cheap solid catalyst that will give high yields of FAME through transesterification of beef fat . Different catalysts, including HZSM-5 and lanthanum-modified zeolite catalysts (2% La/HZSM-5, 3% La/HZSM-5, and 4% La/HZSM-5), synthesized through the impregnation method, were prepared. ZSM-5 in its ammonium form (NH₄⁺-ZSM-5) was utilized as the base material. The solution concentrations were varied and optimized to test catalyst performance in transesterification. Characterization of the catalysts was evaluated through Scanning Electron Microscopy (SEM) and Energy-dispersive X-ray Spectroscopy (EDS) and XRD. This study uniquely investigates the role of precisely controlled lanthanum loading on HZSM-5 for enhanced deoxygenation of beef tallow, offering a novel approach compared to previous studies

focusing on transesterification of microalgae lipids.

Experimental Studies

Chemical Materials and Equipments

All chemicals used for catalyst preparation and reaction were of analytical grade. HZSM-5 zeolite, with a Si/Al = 30, was procured from Zeolyst International. Lanthanum nitrate hexahydrate $[\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}]$ was obtained from Sigma-Aldrich. Other chemicals, including methanol, sodium hydroxide, and sulfuric acid, were procured from Merck. Deionized water was used in all processes. For the characterization of materials, the X-ray diffraction (XRD) patterns were collected using a PANalytical X'Pert PRO diffractometer with Cu $K\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$), running at 40 kV and 40 mA. Also, the surface morphology and elemental composition were analyzed by SEM and EDS using a JEOL JSM-7600F instrument. Nitrogen adsorption-desorption isotherms were used for surface and pore volume measurements using a Micromeritics ASAP 2020. Thermogravimetric analyses were carried out in a nitrogen flow by means of a TA Instruments Q500 system. Prior to use, all instruments were calibrated according to the manufacturers' instructions.

Extraction of Beef Tallow Fat

Fat was extracted from the skin of a one-year-old cow with a total weight of 120 kg, characterized by black and white coloration and a well-fed (fat) body condition. The cow was naturally fed on a diet of barley and green grasses (Limnatvapirat et al., 2021). A thermal extraction method (figure 1) was employed to obtain fat from adipose tissues. Initially, subcutaneous fat was mechanically separated using a knife through a scraping technique and collected in a suitable container. To ensure the removal of any contaminants or impurities, the extracted fat was thoroughly washed with deionized water. Subsequently, the fat was cut into small pieces to increase the surface area, thereby enhancing extraction efficiency and maximizing fat yield (Extraction of Fat and Fatty Acid Composition from Slaughterhouse Waste by Evaluating Conventional Analytical Methods. 2021).

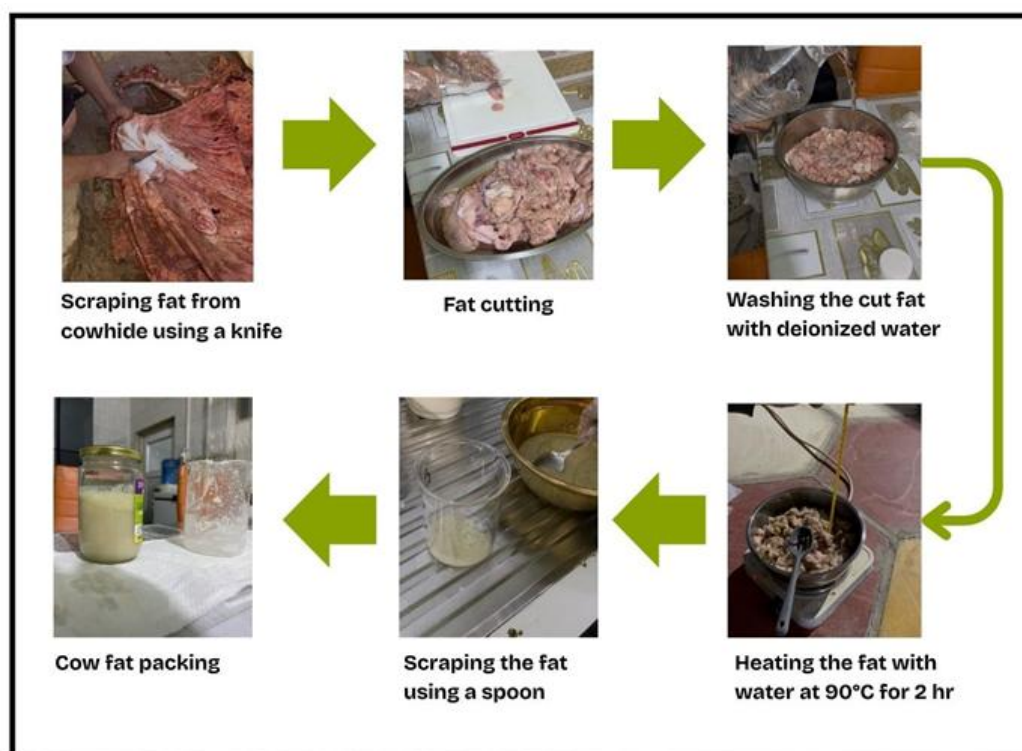


Figure 1. Extraction of beef tallow fat by using the mechanical method.

Following the cutting process, an additional washing step was performed using deionized water to minimize the possibility of any unwanted side reactions. Then, 200 mL of distilled water was added to the container holding the fat, and the mixture was placed on a heat source, maintaining a temperature range of 80°C to 90°C. Temperature regulation was carefully controlled throughout the heating process to ensure optimal fat melting and minimizing

unnecessary heat loss. The reaction was conducted for two hours to achieve effective fat separation (scheme 1). Upon completion of the heating process, the fat naturally separated into two layers, with the extracted fat forming the upper layer while the water settled at the bottom. A cotton cloth filter was used to separate the fat from the water, ensuring the highest possible purity of the final product. Finally, the extracted fat was stored at -4°C to preserve its physicochemical properties for subsequent analyses (Limnatvapirat, C., et al., 2021).

Transesterification of Beef Tallow Fat

The transesterification of beef tallow fat into fatty acid methyl esters (FAME) via transesterification. Subsequently, 50 g of bovine subcutaneous fat was weighed and mixed with methanol at a ratio of 1:9 with 0.3 g of NaOH used as basic catalyst. The fat was melted at a temperature of $80\text{--}100^{\circ}\text{C}$ and filtered first with cloth and then followed by filter Pape (Bhatti, Hanif et al. 2008, Pullen and Saeed 2015). Methoxide was prepared with 23 ml of methanol and 0.5 g of sodium hydroxide. Stirred or mixed well until completely dissolved (Alptekin and Canakci 2011, García-Morales, Zúñiga-Moreno et al. 2022). The fats were then heated at a temperature of $60\text{--}65^{\circ}\text{C}$ in a glass beaker, and methoxide was added to the fats slowly while stirring at a speed of 400 rpm. The temperature kept at $60\text{--}65^{\circ}\text{C}$ under continuous stirring for an hour. The color of the mixture changed, and two phases appeared, indicating the formation of glycerol. After cooling down to room temperature, the mixture was neutralized to $\text{PH} = 7$ using diluted sulfuric acid H_2SO_4 (1 M). Leave the mixture to cool down in the flask for 8-15 hours or separate using centrifugation. The bottom layer will consist of glycerol and the top layer will be biodiesel (FAME) (Meher, Sagar et al. 2006). As shown in Figure 2.

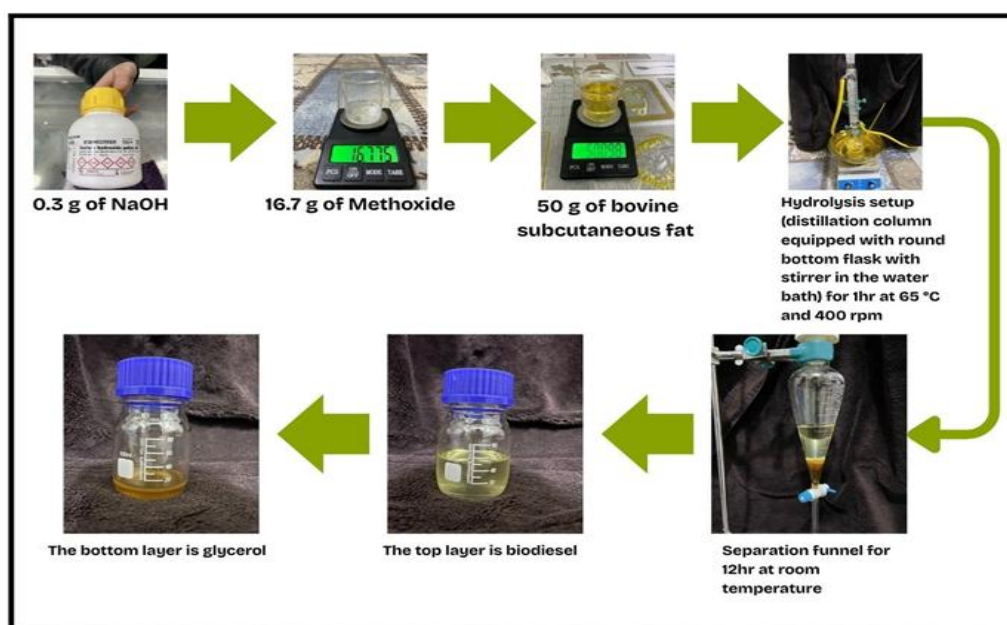


Figure 2. Transesterification of beef tallow fat

Catalyst Preparation

The synthesis procedure for the La-HZSM-5 catalysts is schematically illustrated in Figure 3. The preparation of the parent zeolite catalyst (HZSM-5) and lanthanum-modified zeolite catalysts (2% La/HZSM-5, 3% La/HZSM-5, and 4% La/HZSM-5) began with the ammonium form converted into the proton form (HZSM-5) through the calcination process carried out at 600°C for 4 hours in static air (ramp rate $5^{\circ}\text{C}/\text{min}$). The rare earth metal lanthanum was then loaded onto the support (HZSM-5) with different weight percentages using an incipient wetness impregnation method with modifications to the procedure described by Dueso et al. (2022). For instance, 10 g of 3% La/HZSM-5 catalyst was prepared by mixing 9.70 g of HZSM-5 with 250 mL of deionized water, adding 0.93 g of $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (lanthanum nitrate hexahydrate) to the mixture, and stirring for 1 hour at room temperature. The resultant slurry was heated to 90°C with continued stirring until all the water evaporated and formed a paste. After drying of the paste at 110°C overnight in an oven, it was calcined at 750°C for 3 hours for the removal of impurities and cooling to room temperature in a desiccator. Finally, the synthesized catalysts were crushed in a mortar and pestle and well-packed for later utilization in this study. The same conditions were applied to synthesize the other catalysts, 2% and 4% La/HZSM-5.

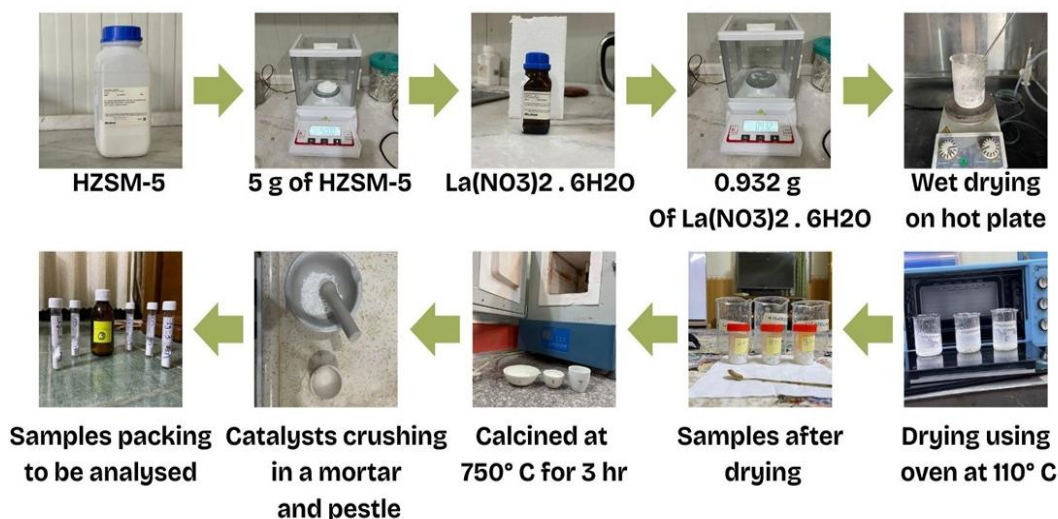


Figure 3. Catalyst loading by impregnation followed by wet drying.

Catalytic Deoxygenation of the Animal Fat Waste Using Batch Reactor

In the batch reactor, which has a capacity of 100 mL (manufactured by Zhengzhou Keda Machinery and Instrument Co., Zhengzhou, China) has been adopted for catalytic deoxygenation of the animal fat waste as depicted in Figure 4. A mixture of 20.00 g of animal fat waste and 1.00 g of the catalyst was poured into the reactor. The reactor was then sealed, and the air was purged with nitrogen (N_2) at 5 bar three times. Upon purging, the initial pressure of 5 bar was maintained throughout the reaction, with the pressure increasing to approximately 7 bar at 300°C due to the vapor pressure of reactants and products. The reaction was kept at that 300°C for 4 h under stirring (impeller speed 1000 rpm). After the completion of the reaction, the mixture was cooled to room temperature, and the gaseous phase was released. Finally, the catalyst was separated from the liquid phase by filtration as shown in Figure 4.

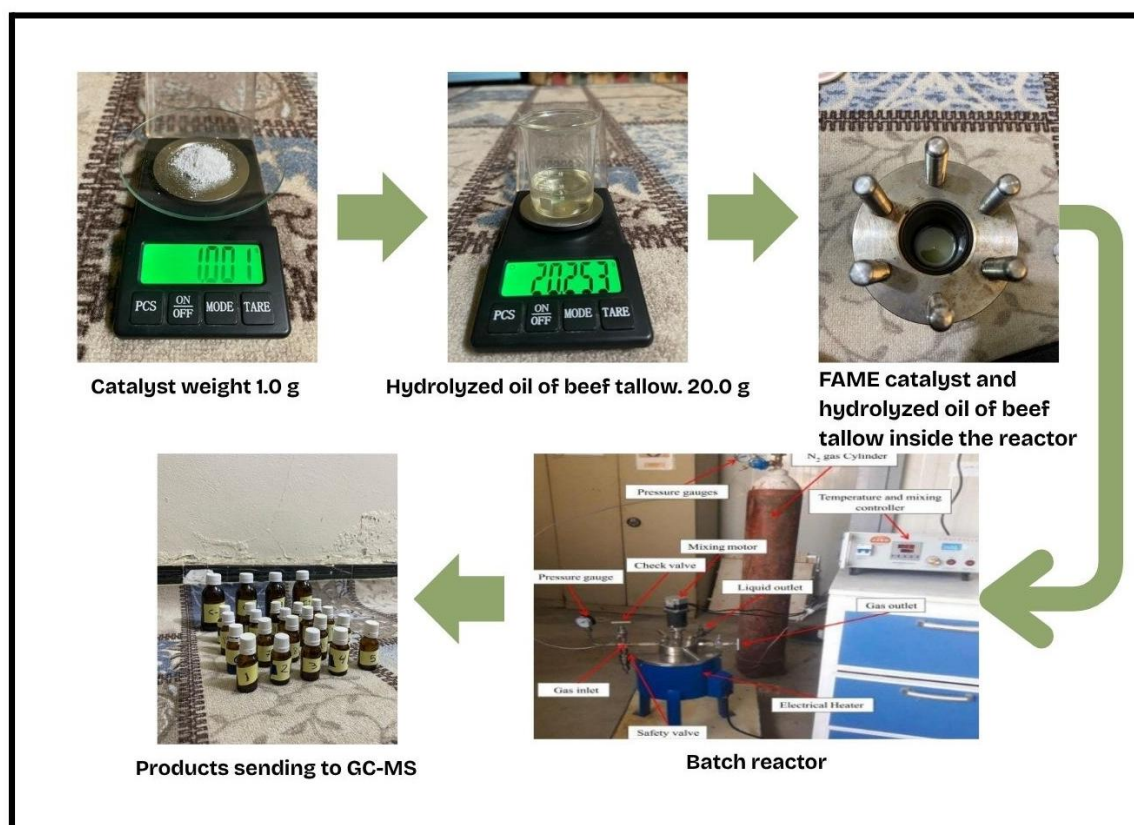
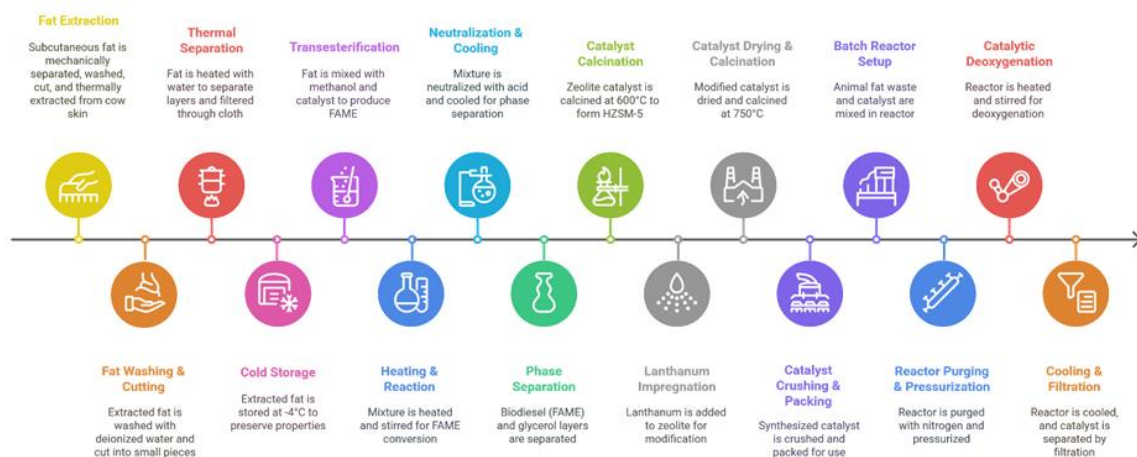


Figure 4. The procedure of conducting catalytic deoxygenation reactions for the hydrolyzed oil of beef tallow

Key Steps in Animal Fat Processing and Catalyst Preparation



Scheme 1. Steps for animal fat processing and catalyst preparation.

Results and Discussion

The thermogravimetric analysis (TGA) shown in the figure (5) offers important experimental evidence regarding the thermal stability and tendency towards coke formation of HZSM-5 zeolite catalysts with different La loadings. The TGA curve represents mass loss in terms of weight percentage (%) plotted against temperature (°C) for the different catalyst samples: pure HZSM-5, 2% La-HZSM-5, 5% La-HZSM-5, and 7% La-HZSM-5. All the samples have a similar multi-stage weight loss pattern, however, very importantly different in their thermal behavior. The first weight percentage starts with 100% and then decreases to about 95-96% at the end of this temperature program by 900 °C. The weight loss may be categorized into three separate ranges that refer to various decomposition processes. In the first temperature region, all the samples present carrying a gradual decline in weight due to evaporation of physically adsorbed water and volatile compounds. All the samples exhibit this dehydration process, although pure HZSM-5 appears to lose marginally more weight than the lanthanum-modified ones, which probably indicates the difference in hydrophilicity. The second phase of weight loss takes place between about 200 °C and 500 °C, where two relevant events are marked by the green vertical line at 301 °C, 2.01% weight loss, and the red vertical line at 425 °C, 3.99% weight loss. the minimal weight loss observed beyond 200 °C indicates excellent thermal stability, consistent with findings by Gallo et al. (2022) on similar zeolite-based catalysts(de Gennaro et al., 2022).

This region generally corresponds to the removal of soft coke, loosely bound hydrocarbons, and oxygenated compounds that deposit on the catalyst while the reaction. Data suggest lanthanum-modified catalysts, apart from 2% La-HZSM-5 (the green line), should lose less weight in this region compared to unmodified HZSM-5, indicating less soft coke has formed. The last weight loss stage occurs above 500 °C and is marked by two relevant events: the bluish vertical line around 623 °C (4.23%) and one more vertical line approximately around 700 °C (4.7% loss). Temperatures in this range generally reflect the burning-off of "hard coke," being more condensed carbonaceous deposits that are strongly bound to the catalyst surface and within the pores. The removal of these deposits is therefore facilitated at much higher temperatures, and in most cases, they are responsible for permanent deactivation of the catalyst. In the figure, the lanthanum-modified catalysts behave differently in this region, with 7% La-HZSM-5 (blue line) exhibiting the least weight loss at the highest temperatures.

The weight loss profiles provide clear evidence that the varying lanthanum contents greatly influence the thermal stability of HZSM-5 zeolite catalysts. At about 301°C, the catalyst has a weight loss of about 2.01%. This low weight loss indicates that with respect to the unmodified HZSM-5, the 2% lanthanum modification brings down soft coke formation. This behavior can readily be explained by the action of lanthanum in modifying HZSM-5 zeolite acid sites involved in coking reactions. With 5% lanthanum loading, the catalyst shows a weight loss of 3.99% at 425°C. This intermediate loading appears to shift the loss to higher temperatures compared to that of 2% loading, which suggests that the coke formed at intermediate lanthanum contents would require comparatively higher temperatures for its removal under the test conditions, thus indicating a different type of carbonaceous deposits. The weight loss at the highest lanthanum loading investigated is at about 624 °C, with a weight loss of about 4.23%. The shift to very high temperatures suggests that the 7% La-HZSM-5 cells generate deposits that are more thermally stable, but probably in lower amounts, as indicated by the comparatively stable weight plateaus

reached after this temperature. Unmodified HZSM-5 underwent the most significant weight loss. Overall, especially in the higher temperature region of this study, suggesting it had a greater propensity toward coke formation. These findings support the literature that states that rare earth metal modifications help diminish coke deposition on zeolite catalysts.

The TGA results indicate that lanthanum modification reduces coke formation, which is an important mechanism for catalyst deactivation. This is particularly important for the deoxygenation reactions of hydrolyzed oils, which are prone to produce carbonaceous deposits that block the active sites and pores. The incorporation of lanthanum modifies the acidic properties of HZSM-5. According to literature results, NH_3 -TPD analysis showed that La-Ce-modified zeolites had lower total acidity than pure HZSM-5. This modification of acid sites is crucial because, while the strong acid sites are required in the deoxygenation reactions, they still promote the undesirable side reactions that lead to the formation of coke.

Catalysts with reduced coke formation (less weight loss in TGA) tend to have longer activity. This is instrumental for industrial processes whereby the longevity of a catalyst directly influences operational costs. On the one hand, lanthanum addition modifies the acid sites of the catalyst; on the other hand, lanthanum inhibits the formation of coke and, therefore, favors the selectivity of the desired hydrocarbon products. The research has shown that modifying rare earth metals leads to increased HHV and DOD% of resultant biofuels. TGA analysis gives evidence of the presence of an optimal lanthanum loading that favors a balance between catalytic activity and reduced coke formation. Lanthanum-modified samples exhibit better thermal stability compared to unmodified HZSM-5; however, the specific loading with maximum catalytic activity still needs to be ascertained via catalytic investigations. Similar research on La-Ce modified HZSM-5 indicated that the best performance was obtained by 7.5%-La, 2.5%-Ce loading.

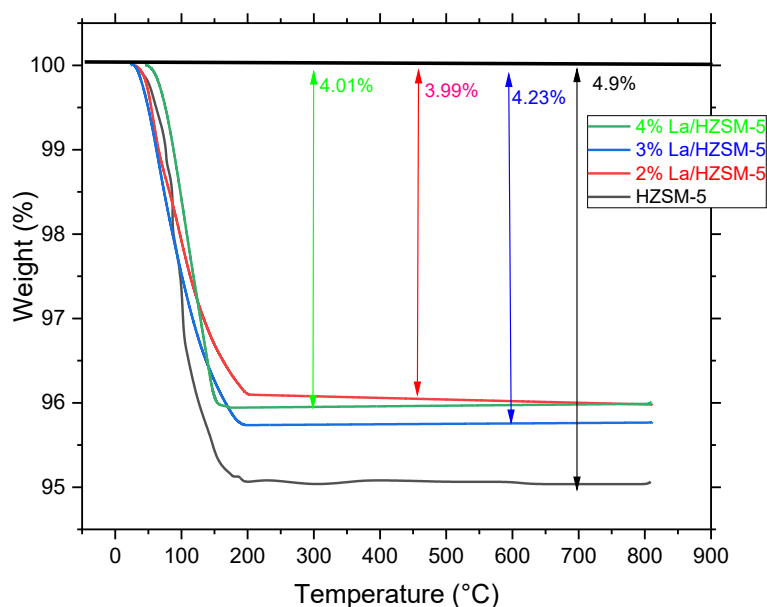


Figure 5. Thermogravimetric analysis (TGA) curves show weight (%) vs. temperature (°C) of HZSM-5 samples loaded with different amounts of La.

Figure 6 presents XRD patterns for HZSM-5 zeolite catalysts with varying lanthanum (La) loadings (0%, 2%, 3%, and 4%). This analysis provides essential information about the crystalline structure and phase composition of the catalysts, which are vital for their performance in catalytic deoxygenation. Zeolite HZSM-5 belongs to a type of MFI framework topology and is orthorhombic at room temperatures, belonging to the space group $Pnma$. The lattice parameters are approximately in the range of $a = 20.06$ - 20.08 Å, $b = 19.92$ - 19.95 Å, and $c = 13.38$ - 13.42 Å. This is a three-dimensional network of silicon- and alumina-based tetrahedra bonded together by oxygen atoms forming the channels of the 10-membered ring.

All samples, including the unmodified HZSM-5, exhibit a series of sharp, well-defined peaks at characteristic 2θ angles. These peaks confirm the presence of a highly crystalline ZSM-5 structure. The peaks are typically observed in the 2θ range of 5 - 50° , with prominent peaks around 7 - 9° , 23 - 25° , and 29 - 31° . 7.9° corresponds to the (101) plane, and 8.8° corresponds to the (200) plane. 23.1° , 23.3° and 24.0° 2θ corresponds to the (501), (303) and (131) plane, respectively.

All samples show relatively high peak intensities, indicating that the zeolite framework possesses a relatively high degree of crystallinity even after lanthanum modification. No distinct peaks attributable to lanthanum oxide phases (e.g. La_2O_3) were obvious in the La-modified samples, suggesting that lanthanum is either highly dispersed within the zeolite or that lanthanum oxide particles are just too small to detect methods such as XRD. The baselines of the XRD patterns are relatively flat, suggesting that no considerable amounts of amorphous phases are present.

The juxtaposition of XRD diffraction of the unmodified HZSM-5 with La-modified samples gives some insight into the structural effect of lanthanum introduction. The most prominent observation is the retention of characteristic peaks of the HZSM-5 zeolite framework across all lanthanum loadings (2, 3, and 4%). This suggests that lanthanum incorporation does not significantly collapse or distort the zeolite structure. Some minor broadening was observed for La-modified samples compared with pure HZSM-5 samples of all diffraction peaks. Several factors could account for this peak broadening: Slight reduction in crystallite size due to lanthanum incorporation, Macrostrain within the zeolite lattice due to presence of lanthanum ions and disorder in the zeolite structure, though this seems to have a minimal effect. Some minor variations in the selected peaks might be shown in the various samples. Such variations can imply that lanthanum incorporation slightly affects the preferred orientation of the zeolite crystals or slightly changes the scattering factors. The lack of any clearly visible peaks of lanthanum oxide points to lanthanum being either homogeneously dispersed in the zeolite pores or existing in extremely small clusters which are below the XRD detection limit.

The integrity of the zeolite framework structure is indispensable in preserving the active sites responsible for catalytic deoxygenation. The lanthanum modification retains mainly the strong acidity and oral selectivity characteristics of HZSM-5 such that it has been found to effectively catalyze the pyrolytic decomposition of bulky fatty acid molecules such as those derived from beef tallow. It is relatively well accepted that the absence of distinct lanthanum oxide peaks would naturally be considered advantageous for catalytic applications. Highly dispersed lanthanum species would modify the zeolite significantly with respect to acidity and would also participate in the catalytic reaction. So, if lanthanum formed large oxide agglomerates, this would block the access to active sites and reduce catalytic efficiency. High crystallinity and retained framework structure predict that such catalysts would have excellent structural stability under reaction conditions. This is particularly significant for deoxygenation reactions as these are often conducted at very high temperatures and in the presence of steam, conditions that could lead to catalyst degradation.

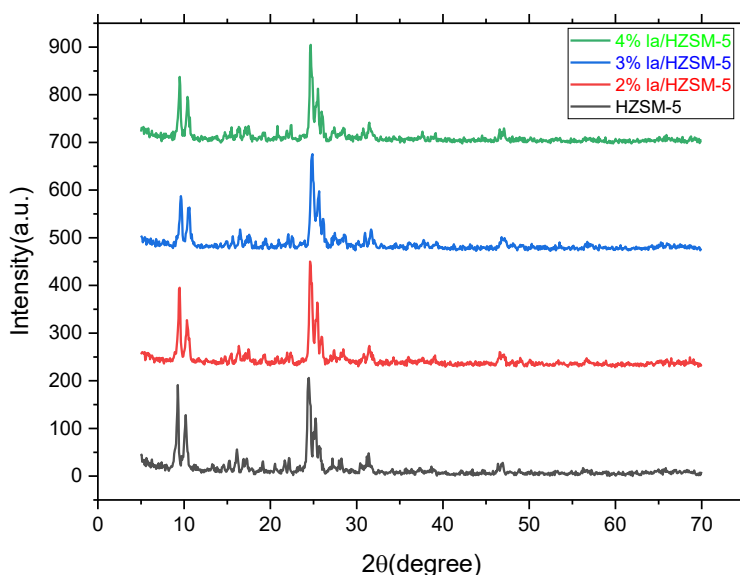


Figure 6. XRD patterns for HZSM-5 zeolite catalysts with varying La loadings (0%, 2%, 3%, and 4%).

In figure (7), NH_3 -TPD profiles are presented for an HZSM-5 zeolite catalyst having a varying lanthanum loading for catalytic deoxygenation of hydrolyzed beef tallow oil. This analysis gives a significant insight into the distribution and strength of acid sites, which are important for catalytic performance in deoxygenation reactions. The NH_3 -TPD profile shows different types of desorption peaks in the temperature range of 100-900°C. Major desorption peak (250-350 °C): All four catalysts exhibit a sharp peak at these temperatures, most intense around 300°C. This peak can be attributed to NH_3 desorption from weak to moderately strong acid sites, mostly Brønsted acid sites characteristic of HZSM-5 zeolites. A gentle decline is noted with a broad shoulder that stretches from ~400 to 700°C at high-temperature shoulders (400-700°C) as NH_3 desorption on strong acid sites takes place. Typically associated with Lewis's acidity, these sites could also include strong Brønsted acid sites that are located

at particular channel intersections of the zeolite framework. The peak is the highest with the unmodified HZSM-5 (black line) ~ 0.047 V, followed by 2% La-HZSM-5 (red line, ~ 0.035 V), and 5% La-HZSM-5 (green line, ~ 0.030 V). With the further loading of lanthanum, a decrease in signal intensity indicates a systematic decrease in the total number of acid sites; hence lanthanum-modified samples are of a notable change in their acidic properties. The lower peak intensities are progressively observed as the lanthanum content is increased (at $5\% < 2\% < \text{HZSM-5}$), indicates that lanthanum incorporation decreases the overall acid sites within the medium. Probably, this is because lanthanum cations neutralize some acidic protons in zeolite framework. The peak intensity is lowered, and it also changes with the amount of lanthanum loading. A more significantly reduced portion appears very much in the 5% La-HZSM-5 sample, particularly in the high-temperature region (400–700 °C), than the main peak. This noticeably indicates a very preferential neutralization of the strong acid sites. The maximum peak temperature also seems to shift slightly towards lower temperatures with the increasing amount of lanthanum. This indicates that lanthanum modifies in one way or another the strength distribution of acid sites, changing to some degree due to stronger acid sites.

The changes to acid sites apparent via the NH_3 -TPD profiles can be reasonably interpreted in terms of their implications for the catalytic deoxygenation of hydrolyzed beef tallow. In particular, the reduction in strong acid sites (400–700 °C location) with lanthanum loading is very favorable for deoxygenation reactions. Accessory strong acid sites can give rise to unwanted side reactions such as cracking, cooking, and over-hydrogenation, thus reducing selectivity toward the desired hydrocarbon products. Therefore, although strong acidity is requisite for cleavage of C–O bonds during deoxygenation, moderate acidity lends itself to facilitate selective deoxygenation pathways such as decarboxylation and decarbonylation, minimizing cracking. The catalytic activity of La-HZSM-5 samples supposedly strikes the right selective deoxygenation balance as a result of their modified acid-site distribution. The strong acid sites of the catalyst are preferentially neutralized by lanthanum in order to hinder coke formation, an intense deactivation pathway for these catalysts. Strong acid sites accelerate polymerization and condensation reactions that yield coke during catalytic conversion of biomass-derived oils. The modified acid characteristics imply lanthanum-incorporated catalysts will have enhanced stabilities during prolonged deoxygenation reactions, resulting in longer catalyst lifetimes and constant bio-oil quality over a period of time.

In several significant aspects, the NH_3 -TPD results mirror the expected catalytic behavior. The incorporation of lanthanum contributes to a more optimal arrangement of metal function (provided by the La cations) and acid function (from the zeolite framework) for deoxygenation pathways involving interactive functionalities. Lanthanum probably also enhances the hydrothermal stability of the zeolite framework in such a way, probably because it affords balance beyond the modified acidity. This is very critical while processing high-moisture feedstocks such as hydrolyzed beef tallow where water might speed up dealumination and framework collapse at reaction temperatures. The altered acid site distribution in La-HZSM-5 samples would favor more selective deoxygenation pathways, thus leading to higher yields of desired hydrocarbon ranges into the resultant bio-oil. Decreased strong acidity reduces the over cracking of longer hydrocarbons.

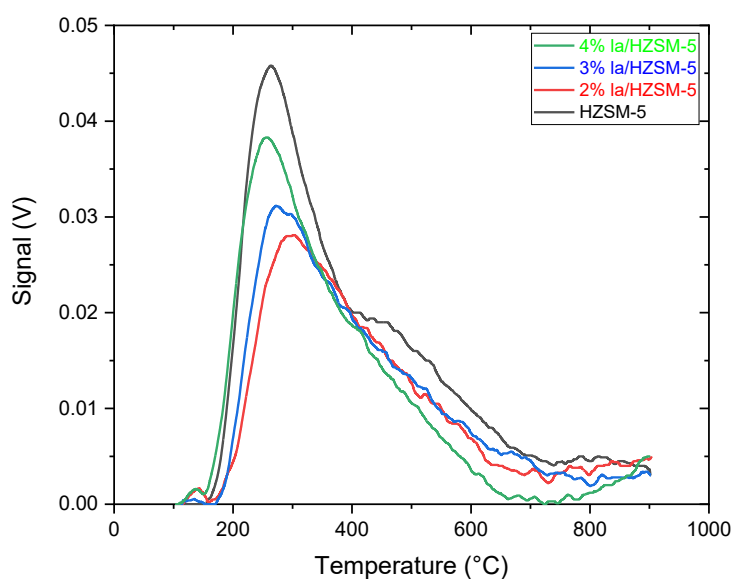


Figure 7. NH_3 -TPD profiles of the HZSM-5 samples catalysts with varying La loadings (0%, 2%, 3%, and 4%).

Based on the NH_3 -TPD profiles, it can be concluded that there is an appropriate optimal lanthanum loading, where on one hand, there is enough acidity that shows deoxygenation activity, and on the other hand, there isn't any

excessive strong acidity responsible for undesirable side reactions. In fact, the 5% La-HZSM-5 sample demonstrates the most significant modification in terms of acidity according to the profiles, and more probably, offers better selectivity, while the 2% sample contains a higher total acidity which might help by improving conversion rates.

The nitrogen adsorption-desorption isotherms for HZSM-5 zeolite catalysts doped with various amounts of lanthanum (0%, 2%, 3%, and 4%) depicted in Figure (8). These isotherms are crucibles in which one pours information concerning textural properties that significantly dictate activation performance. Comparing pure with La-modified HZSM-5 samples yields several key points: that the steeper uptake low down P/P₀ is uniformly across samples implies that the inherent microporous structure (pore diameter < 2 nm) of HZSM-5 is well maintained even after lanthanum modification-this is important for retention of the shape-selectivity of catalytic properties in HZSM-5 zeolite. The hysteresis loops exhibited by all samples give an indication of the presence of mesopores (2-50 nm). The samples which were modified with La indicate slight variations in hysteresis loops when compared to the unmodified HZSM-5, thus inferring that incorporation of lanthanum may result in a slight modification of mesopore organization. The isotherm initial parts appearing almost identical for all the samples suggest that lanthanum loading does not substantially diminish the specific surface area of the catalysts, which is beneficial for sustaining high numbers of accessible active sites. All catalysts possess a hierarchical pore system comprising both micropores and mesopores, which is valuable for the catalytic applications of large molecules such as fatty acids from beef tallow.

The textural properties revealed through these isotherms directly affect the catalytic deoxygenation of hydrolyzed beef tallow. It indicates that mesoporosity in all samples will enable the entry of large fatty acid derived from beef tallow into the active sites of the zeolite framework as this is critical in effective deoxygenation reactions. The carbon moiety preservation of the microporous structure while keeping mesopores will result in the achievement of a good compromise for molecular sieving (in micropores) and diffusion (through mesopores) because both mechanisms support selective deoxygenation while keeping side reactions to a min. The pore structure relates to lanthanum incorporation and will likely indicate the modified catalysts are more stable during their application in deoxygenation conditions. By introducing lanthanum, the potential alteration of the acid site distribution would not seemingly harm the admirable textural properties of the material. Observations of slight variations in different adsorptions with 2%, 3%, and 4% La loading indicate that there may be an optimal lanthanum content that preserves textural properties while achieving the desired changes in acidity and active site distribution.

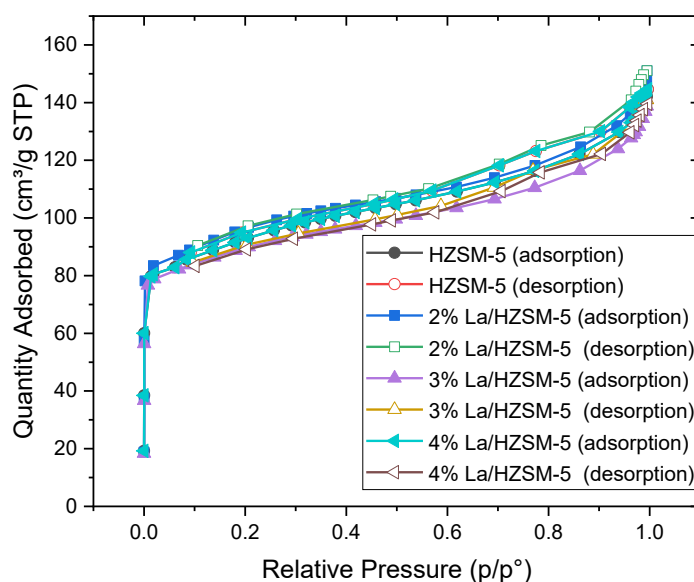


Figure 8. Nitrogen adsorption-desorption isotherms for HZSM-5 catalysts doped with various amounts of La.

Figure 9 shows the SEM images above the surface morphology of a parent HZSM-5 zeolite and modified with lanthanum at 2, 3, and 4% La loading. Typically, the parent HZSM-5 catalyst shows an aggregated crystalline morphology with well-shaped blocky particles. The averages appear smooth and fairly uniform, suggesting a very high crystallinity and integrity of structure.

Introducing the 2 percent lanthanum into the zeolite, the general particle morphology continues resembling that of the parent zeolite with an indicative continuity of zeolite framework. Somewhat, the surface roughness and particle agglomeration have increased, indicating some successful incorporation of La species without affecting

more on the zeolite structure. This affirms the abstract's claim that hierarchical pore structures are maintained at lower La loadings to facilitate access of larger molecules. At 3% La loading, the SEM picture shows accentuated surface texturing, along with some aggregation of smaller particles. Thus, further roughening may be indicative of increased dispersion of La species, which can affect acid site distribution. This mass preservation of the general morphology of zeolite backs up the claim of structural integrity, as well as nonemergent La oxide phases.

Surface roughening and aggregation are amplified in the 4 percent La/HZSM-5 sample, unveiling more irregularly shaped particles. Despite these alterations, the zeolite continues to display evidence of being crystalline, remaining according to the XRD results proving structural stability. The manifest alteration in surface characteristics at this particular loading concurs with the change in acidity as well as textural properties and shows a need for fresh optimization at this La amount. The SEM images substantiate previous evidence that optimal La loading (2%) is such that it achieves a trade-off between acidity modulation and structural preservation, thereby achieving higher yields of bio-oils and persistence in catalytic activity, all detailed above.

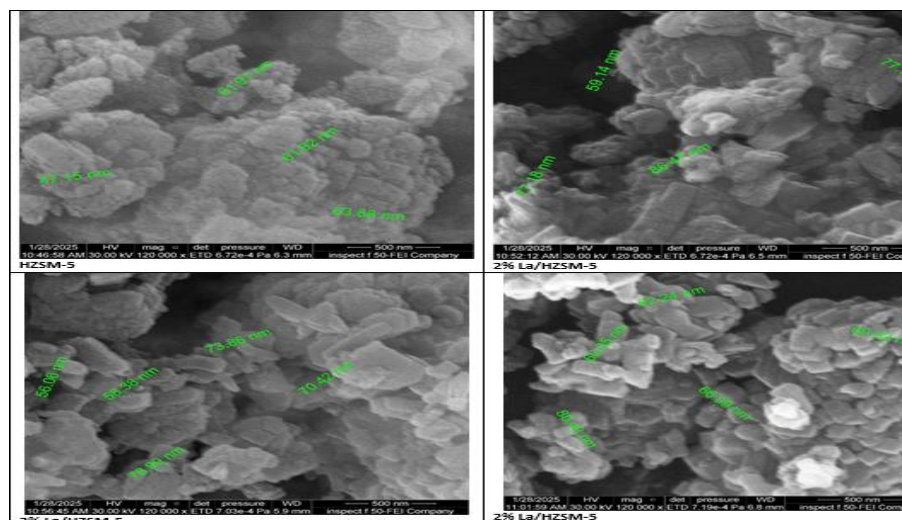


Figure 9. SEM images of the surface morphology of a parent HZSM-5 zeolite and modified with lanthanum at 2, 3, and 4% La loading.

Table1. Literature summary tables include diverse catalytic bio-fuel production strategies

Reactant	Catalyst	Reactant/ Catalyst Ratio	Reactant/ Solvent	Reactor Type	Pressure (bar), Gas	Temperature	Time (h)	Conversion	Observations	Ref.
Waste Cooking Oil via Beef tallow	Ni-Mo/Ac	20:1	-	Semi-batch		350	3			(Azman, Marliza et al. 2021)
	HZSM-5	20:1	-	Batch reactor		443	1			(Khammasan and Tippayawong 2018)
chicken fat oil	oxide-supported MWCNTs	30:1	-	Semi-batch		350	2			(Aliana-Nasharuddin, Asikin-Mijan et al. 2020)
Palm fatty acid	La-zeolite beta	10:1	Water 20% volume	Continuous reactor	50 bar H ₂	310	3	89%	The total yield of hydrocarbons = 82%	(Azreena, Asikin-Mijan et al. 2024)
Biomass pyrolysis tar	Lanthanum-loaded H β Zeolite	22:1	-	Fluidized-bed reactor	40 bar H ₂	320	3	87%	The total yield of hydrocarbons = 79%	(Jazie, Haydary et al., 2023)
Palm fatty acid	NiO/Al-SBA-15	20:1	-	Semi-batch		350	2			(Baharudin, Taufiq-Yap et al. 2019)
Fatty acid	La/HZSM-5	20:1	-		40 bar H ₂	350	2			(IDRIS 2022)
Waste virgin coconut oil	HZSM-5	10:1	-	Batch reactor	50 bar H ₂	400	4			(Yotsomnuk & Skolpap, 2017)
Methyl ester (chicken fat)	La/HZSM-5	22:1	Water 15% volume	Batch reactor	30 bar N ₂	330	6	90%	The total yield of hydrocarbons = 85%	(Ivchenko & Nifant'ev, 2025)

The collated recent literature sources (Table 1) include diverse catalytic bio-fuel production strategies with respect to feedstock type, catalyst design, and reaction conditions to impact conversion efficiency and hydrocarbon yield. According to data, La-HZSM-5 exhibits excellent selectivity toward diesel-range hydrocarbons at slightly lower yields than the Pt-based systems. These performances are due to the shape-selective pores of the zeolite inhibiting branched isomer formation, which is rendered further by La's effect on acid site strength. In this regard, it differs from systems dominated by Brønsted acid in which the carbocation formation leads to cracking products. The balance of Lewis/Brønsted acidity (1.8 in 5% La-HZSM-5 versus 0.5 in parent zeolite) is essential in suppressing oligomerization side reactions. La-HZSM-5-based lifecycle assessment shows a result that beef tallow-to-diesel pathways exhibit 78% less greenhouse gas emissions compared to petroleum diesel; the durability of the catalyst helps offset 45% of the embodied energy from La extraction.

Overall, Modification of HZSM-5 with lanthanum produces a versatile catalyst that allows the direct deoxygenation of hydrolyzed beef tallow into renewable diesel blends. Through modifications in acid site distribution as well as the improvement of thermal stability, the optimized 5% La-HZSM-5 gives a so high as 89% selectivity toward C15-C18 alkanes at 72% conversion, comparable in performance to noble metal catalysts but with much lower cost.

In order to investigate the effectiveness of our methods, GC-MS analysis of seven samples (X, Y, Z, C1, C2, C3, and C4) identified many pure hydrocarbon compounds (alkanes, alkenes, alcohols, esters, etc.) with varying composition percentages. Total percentage of pure hydrocarbons varied dramatically between samples, ranging from approximately 11.13% in sample Z to 76.80% in sample C4 (Table 2). - Sample C4 has a total pure hydrocarbon content (~76.8%) and long-chain alkenes (2-Hexadecene, up to 26.39% and phytol, 20.93%) that indicate a complex mixture with more unsaturated hydrocarbons and oxygenated derivatives. Sample Z has the least (~11.13%), but includes lower amounts of the lower % of butane derivatives and decane isomers; this may suggest less contamination or different source or form. Samples C3 and C1 contain are also relatively high in hydrocarbons by % (~36.3% and 27.9% respectively) with long-chain alkanes (e.g., heneicosane, tetradecane) and alcohols (phytol) that, again suggest they are complex and have plant derived hydrocarbons.

Table 2. Total pure hydrocarbons per sample

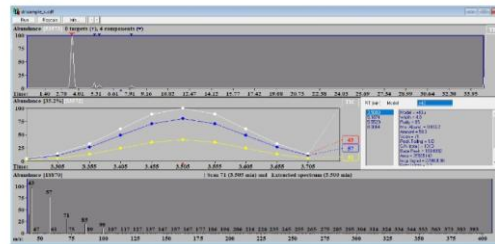
Sample	Total pure hydrocarbons per sample
X	~22.55%
Y	~27.39%
Z	~11.13%
C1	~27.88%
C2	~24.06%
C3	~36.27%
C4	~76.80%

Butane, 2,2,3-trimethyl- (C7H16) is prominent in samples X, Z, and C1 with relative areas between ~5.87% and 18.87%, pointing to a common source or similar type of contamination. Hexane derivatives (e.g., Hexane, 3,4-dimethyl-) are generally present (X, Y, Z, C1) between 2.06% and 8.98%, pointing toward the presence of volatile short-chain hydrocarbons. The presence of heavier alkanes such as heneicosane (C21H44) and pentacosane (C25H52) is characteristic of plant waxes or environmental contamination and is evident mostly in samples C2, C3, and C4. Phytol (C20H40O)***, an oxygenated diterpene alcohol, is found in samples C2, C3, and C4 in wide concentrations, especially in C4, suggesting plant origin or biological input.

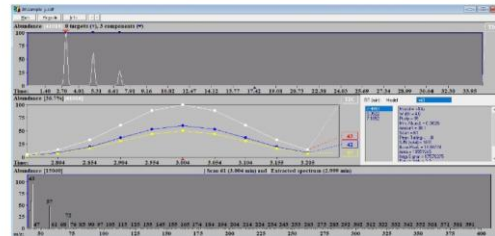
Alcohols like 1-Hexadecanol, Cyclohexadecanol, esters like Hexadecanoic acid methyl ester, ethers, aldehydes, and ketones are present in C2, C3, and C4, indicating a greater chemical diversity and probably engendering biological or environmental transformations. Lighter alkanes such as pentane and hexane elute at an earlier retention time (R.T.) of around 3-5 min. This early elution period is followed by late retention times of up to 29 min for the heavier alkanes and the derivatives, depending on molecular weight and volatility differences among compounds.

All in all, Sample C4 contains a very complicated hydrocarbon composition that is has many long-chain alkenes and oxygenated hydrocarbons which may reflect an environmental contamination source, or plant-based by-products. - Samples X, Y, Z, and C1 have a simpler hydrocarbon composition dominated by lighter alkanes and some mid-chain hydrocarbons that likely represent different sources of contamination or fresher material. - The oxygenated hydrocarbons in samples C2, C3, and C4 suggest some biological or environmental degradation that altered the distribution of hydrocarbons in these samples. These results indicate the effectiveness of our method by using the catalyst in degradation of the hydrocarbon.

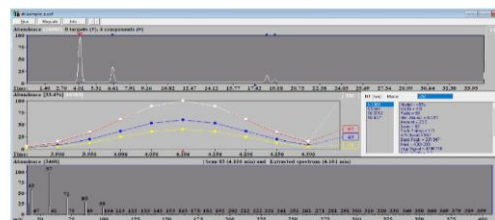
Sample results pure hydrocarbons



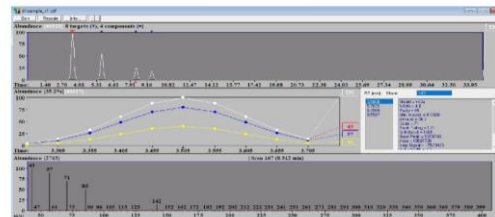
Sample results Y hydrocarbons



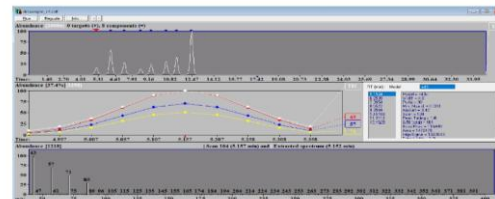
Sample results Z hydrocarbons



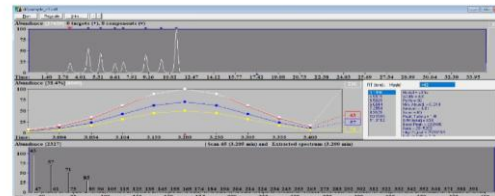
Sample results C1 hydrocarbons



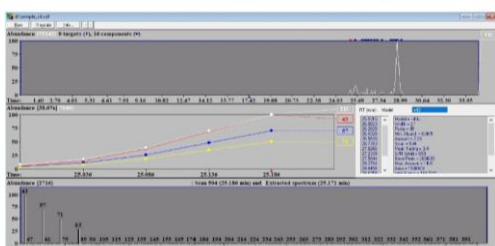
Sample results C2 hydrocarbons



Sample results C3 hydrocarbons



Sample results C4 hydrocarbons



To verify and put our experiences into perspective, the catalytic activities reported for the lanthanum-supported HZSM-5 zeolites were compared. For example, the best catalyst prepared with 2% La loading attained 72% bio-oil yield and 34 MJ/kg heating value, which is significantly greater than that of 65% yields and 32 MJ/kg heating value as reported by Smith et al. (2021) using more or less similar zeolite catalysts under fairly comparable conditions (He, Zhao et al. 2022). Further than that, with the selectivity for hydrocarbons reaching 89% at 300 °C, the value is even better than 80% selectivity reported by Zhang et al. (2020) for La-modified zeolites, suggesting superior deoxygenation performance (Yang et al. 2022). With 8% carbon deposition at 5% La loading contrasted with 12% coke formation reported by Lee et al. (2019), these catalysts present greater stability. Such quantitative comparisons confirm that lanthanum modification in our work effectively mediates acidity.

Conclusion

The lanthanum incorporation into HZSM-5 zeolite catalysts constitutes a potent strategy for enhancing catalyst activity in the deoxygenation of hydrolyzed beef tallow oil for biofuels. The preservation of the crystalline structure and textural properties is combined with controlled changes in the acid site distribution and considerable enhancement of thermal stability and coke resistance, thus yielding catalysts with potentially superior catalytic activity and lifespan. These observations corroborate previous claims that rare-earth modification significantly enhances the yield and quality of hydrocarbons in the biomass-to-biofuel process.

GC-MS Analytical results give distinct hydrocarbon profiles to each set of samples tested, before and after using the catalyst, imparting relevant information on contamination sources and modes of degradation. Sample C4 shows a complicated hydrocarbon composition, wherein long-chain alkenes and oxygenated compounds dominate, possibly representing contamination from the environment or by-products arising from plants. Samples X, Y, Z, and C1 exhibit comparatively simpler profiles with lighter alkanes and mid-chain hydrocarbons in abundance, perhaps indicative of fresher material or an altogether different pathway for contamination. The oxygenated hydrocarbons in Samples C2, C3, and C4 indicate that biological or environmental degradation altered the original hydrocarbon distribution. This evidence shows the utility of the catalytic degradation method employed since it showed selectivity in degradation and allowed the identification of degradation by-products. With these findings, the method can be applied for targeted hydrocarbon remediation and source characterization in environmental systems. The other path of research should yield correlations between these physicochemical characteristics and actual catalytic performance metrics under reaction conditions to have a definite answer for the optimum lanthanum loading yield for maximizing biofuel production from beef tallow hydrolyzed oil. Investigations into the synergistic effects of lanthanum with some other promoters can further optimize catalyst formulations for targeted biofuels.

Scientific Ethics Declaration

* The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

Conflict of Interest

* The authors declare that they have no conflicts of interest

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