



Review article

A review on biodiesel micromixers: Types of micromixers, configurations, and flow patterns

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ABSTRACT

The production of biodiesel conventionally relies on batch reactors for the transesterification of oil and alcohol. However, the inherent limitations of batch-wise biodiesel production, including biphasic oil and alcohol, the establishment of equilibrium during transesterification, and heightened manufacturing costs, underscore the need for intensifying biodiesel synthesis. The integration of microreactors and micromixers presents a promising avenue to achieve these objectives, driving significant interest in the development of continuous biodiesel synthesis within microreactor systems. Continuous microreactors, empowered by micromixers, offer key advantages such as a heightened interfacial area between immiscible reactants and phases, as well as reduced mass transfer resistance, culminating in elevated biodiesel yields. Consequently, the micromixer emerges as a pivotal component in microreactor systems. This review delves into the pivotal role of micromixers in biodiesel production within microreactors, shedding light on micromixer types, channel configurations, reactor dimensions, mixing indices, and the influence of co-solvents in micromixers. The efficiency of various micromixer types is meticulously analyzed using a mixing index and yield of oil. Furthermore, the review addresses the challenges inherent in biodiesel production when employing micromixers and microreactors.

1. Introduction

Research into biofuels is initiated because of environmental pollution. High fossil fuel prices also play a role. There's a need to secure energy and the environment. Uncertainty about future petroleum availability is an issue. This uncertainty stems from rising demand and declining reserves. Biodiesel, made from vegetable oil or animal fats, is rapidly replacing fossil fuels. The process of transesterification involves a chemical interaction between a vegetable oil primarily made up of triacylglycerides and alcohol. This is the most popular approach to producing biodiesel [1–3]. Hence, to address environmental concerns and reduce dependence on finite fossil fuel resources' It's imperative to continue advancing research into renewable biofuels like biodiesel. To further accelerate this transition, efforts should focus on optimizing transesterification processes and implementing sustainable production practices to ensure a viable alternative to petroleum-derived fuels.

The oil-alcohol immiscibility causes the transesterification to proceed in two phases. A biodiesel-glycerol two-phase system is developed during the second phase of transesterification. This is a quick and chemically regulated phase where the reaction is a

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homogeneous medium. This phase is preceded by another one, which is a slow equilibrium regime until the conclusion of the reaction. Mass transfer between the two immiscible phases (oil and alcohol) governs the first phase, which is sluggish. The immiscibility of raw materials to biodiesel slows down mass transfer between two phases and lengthens the reaction times. To achieve the higher reaction rate required, it is necessary to overcome the mass transfer constraint between these reactants. Hence, effective contact between the mixture of oil, alcohol, and catalyst becomes essential. The challenges posed by oil-alcohol immiscibility in transesterification, enhancing mass transfer between these phases is crucial to improve reaction rates. Approaches to achieve this could include optimizing mixing conditions, catalyst selection, and reactor design to facilitate effective contact between oil, alcohol, and catalyst, thereby accelerating biodiesel production while minimizing reaction times and energy consumption [3].

Therefore, a large amount of churning of the mixture is necessary to reduce the resistance of mass transfer and enhance contact surfaces. As a result, Stirred-tank reactors are employed in several industrial facilities. Even though this conventional biodiesel synthesis method is commonly employed on an industrial scale, several significant obstacles still need to be overcome. This may include long reaction time or residence time required to achieve a better yield of biodiesel [1,2], which can range from 60 to 120 min. Also, it induces high capital investment costs, is energy-intensive, has low manufacturing efficiency, and the unprofitable economics of industrial-scale batch reactors and a huge extent of wastewater generation [3]. Also, it necessitates a mechanical stirring tool (external mixers), which uses a lot of energy. Moreover, the size of the batch reactor places a cap on the amount of biodiesel that can be converted [4,5] in the absence of product withdrawal, thus the benefits of continuous operation are not presented [2]. Therefore, research into renewable biofuels, driven by the need for sustainable energy amid environmental concerns is a promising alternative. However, challenges such as sluggish mass transfer in transesterification and drawbacks in conventional stirred-tank reactors hinder efficient production. Overcoming these obstacles is vital for widespread biodiesel adoption, emphasizing the importance of advancing reactor technologies for sustainable and economically viable biofuel production.

Several intensification technologies have been developed to get over the drawbacks of the conventional batch transesterification process [6]. For instance, various heating and mixing technologies, including microwave irradiation, the supercritical-fluid method, hydrodynamic cavitation, reactive distillation, and ultra-sonication were employed to increase manufacturing efficiency [7–14]. The biggest obstacle to increasing the capacity of biodiesel production, however, is the high operational cost and technological complexity of the new approaches compared to the conventional process [15]. Therefore, intensification technologies like microwave irradiation, supercritical-fluid methods, and others aim to improve biodiesel production efficiency. Yet, their high costs and complexity hinder widespread adoption. Future research should focus on optimizing these methods for better cost-effectiveness and scalability to integrate them into commercial production.

Microtechnology application for an effective biodiesel production process is one of the efficient ways to get over these challenges. The term microtechnology refers to a group of procedures, methods, and equipment used to produce miniaturized structures with a size between 0 and 10^3 μm . High surface-to-volume ratio, rapid heat, and mass transmission, less consumption of energy, adaptable capacity and design, safety, and environmental friendliness are just a few benefits of Microtechnology [16,17]. Microtechnology offers advantages like high surface-to-volume ratio and rapid heat and mass transfer. Optimizing microreactor systems through precision engineering could enhance efficiency of biodiesel production processes, fostering broader adoption across industries. Microreactors typically have internal characteristics with dimensions between 0 and 1000 μm . Microchannels' compact dimensions enable the use of small reagent amounts under perfectly controlled operating conditions, allow for the quick screening of reaction operational conditions, and enhance process safety in general. In addition, the microreactor system exhibits superior mass and heat transfer, a short residence time, fewer waste byproducts, laminar flow, efficient mixing, a close molecular diffusion path, improved control of the process, and reduced consumption of energy. Therefore, microreactors are being used in more and more industries, including the chemical industry, biotechnology, pharmaceutical industry, and medicine [18]. The use of this technology in the synthesis of biodiesel may represent the next significant development in both process intensification and economics. Different geometries such as micro size, multiplicity architecture, and materials are what define microreactor systems.

A micromixer for enhancing mixing and a microchannel, also known as a micro tube, to reach the reaction to completion mainly make up the microreactor system. These can be used singly or collectively for biodiesel production [19]. Micromixer-microreactor systems offer compact dimensions and excellent mass and heat transfer, suitable for diverse industries. Utilizing them in biodiesel synthesis can boost efficiency and economics. Since two immiscible phases (oil and alcohol) are involved in biodiesel synthesis, it presents a chance for intensification of the process employing a microreactor system [20]. The use of a microreactor system (microreactor and micromixer) can greatly increase the mixing between the immiscible phases, boosting the transfer rates and, as a result, raising the yields of biodiesel. Therefore, the key step in the biodiesel production process is mixing, which adds residence time and expense [21]. In many microreactor systems, effective mixing is crucial. Mixing is a procedure that makes all of a system's attributes homogeneous within a short time. Typically, it goes through two steps: The process begins by forming a finely distributed structure of a heterogeneous mixture. The second stage results in a homogeneous mixture between the adjacent molecules as a result of diffusion. Hence, mixing equipment relies on rearranging the fluids such that their constituent parts are evenly distributed. Larger contact surface areas are consequently created during efficient mixing, which enhances mass and heat transport and highly encourages fast chemical reactions. The reaction, however, could be slowed down if there is a local scarcity of one of the reactants, an uneven distribution of the catalyst, and temperature non-uniformities if the mixing process is subpar. Due to its impact on several factors, including heat and mass transfer rates, process running time, costs, and ultimately product quality, mixing efficiency in a micromixer is crucial for enhancing the performance of the process [21].

A few $\mu\text{L}/\text{h}$ to a few mL/h are typical flow rates for biodiesel production using a continuous microreactor system. A laminar flow regime results from these quite slow flow velocities. Reynolds numbers in the microreactor are therefore low. It is widely recognized that the laminar regime does not support the creation of high heat and mass exchanges since there is no turbulence in the flow [4,22].

Thus, it is crucial to build micromixers with higher mixing efficiency in tiny channels and at lower Reynolds numbers. Laminar flow is characterized by low Reynolds numbers, which can occasionally make it difficult for different species to mix. This can be justified because molecular diffusion produces the fluid mixture in the majority of micromixers. Most micromixers work well using the diffusional mixing method without any help from turbulence because they almost always have a laminar regime because of the small dimension of the microchannel. This implies that in biodiesel production using continuous microreactors, low flow rates create a laminar flow regime with challenges for heat and mass exchanges. Micromixers with higher efficiency are essential in tiny channels to overcome the limitations of laminar flow. Despite low Reynolds numbers, molecular diffusion predominantly drives fluid mixture creation in micromixers, the importance of their effectiveness in micro channels [23]. Hence, to enhance biodiesel production in continuous microreactors, prioritize developing more efficient micromixers to address challenges posed by low Reynolds numbers and laminar flow. Enhancing mixing efficiency in microchannels, especially at lower Reynolds numbers, is essential for optimizing heat and mass exchanges in biodiesel production.

This review paper on micromixer types, configurations, and flow patterns is crucial for consolidating knowledge in the field, particularly due to the absence of a detailed review on this topic. Exploring various micromixers, their configurations, and flow patterns, the review serves as a key resource for researchers, engineers, industry professionals, and policymakers in sustainable energy and biofuel technologies. Its primary contribution lies in providing a holistic understanding of current biodiesel micromixing technology, offering insights beyond theory and addressing research gaps. The evaluation of methodologies enhances its utility, providing a critical perspective.

2. Types of micromixers

Microfluidic mixing is a technique used in microstructured devices to quickly and thoroughly mix several samples. The main ways these devices produce fluid mixing is by boosting the diffusion process between the various species fluxes and developing chaotic micro mixing. Micromixers are typically categorized as passive or active based on the principles used to induce mixing inside the microscale [24]. Active micromixers use external sources of energy to improve mixing, such as electrical, magnetic, acoustic, or optical energy [25–27]. In contrast, mixing with passive micromixers is achieved through the diffusion of molecules and the fluid's own considerable disorder or chaotic advection. The use of active micromixers is constrained since most of these types of equipment need a complicated setup to construct the platform, which can result in a high mixing index [25]. For the realization of complex functionalities, passive micromixers can be made reasonably easily and are easily integrated with other microfluidic devices. As a result, passive micromixers are utilized more frequently for biodiesel synthesis [28]. It's worth investigating ways to streamline the setup of active micromixers, boosting their usability and competitiveness against passive counterparts. This could potentially expand their utility across diverse domains.

2.1. Active type micromixers

Small appliances called micromixers are used to combine fluids on a microscale. Active-type micromixers can achieve high mixing efficiencies at low Reynolds numbers by enhancing mixing with external energy. According to Cai et al. [29], this makes active micromixers ideal for blending viscous liquids like vegetable oils and biodiesel [29]. To produce biodiesel, numerous active-type micromixers have been developed. Piezoelectric micromixers [30], Acoustic micromixers [31], Electro kinetic micromixers [32], and Magnetic micromixers [33] are some of the active micromixers used in biodiesel production and purification process.



Fig. 1. Acoustic micromixer [29].

2.1.1. Acoustic micromixers

The production of biodiesel is largely dependent on effective micro-scale mixing, or micromixing, which increases the rates at which triglycerides and alcohol react during the transesterification process. Because of laminar flow regimes, conventional stirring techniques are unable to accomplish this in microfluidic channels. An innovative and promising approach to this problem is provided by acoustic micromixers (Fig. 1). These devices induce microstreaming and turbulence in the fluid mixture by sending sound waves into the reaction chamber. As a result, the laminar flow is disturbed, and the reactants are distributed more evenly throughout the microchannels. Acoustic micromixers are a desirable technological option for streamlining the production of biodiesel because of their improved mixing, which results in quicker reaction times and possibly higher biodiesel yields [29,34].

Acoustic micromixers also offer several advantages over conventional methods. Because they are naturally contactless, there is no need for moving parts inside the microreactor, which lowers the possibility of contamination [35]. Furthermore, the sound waves' frequency and intensity can be precisely adjusted, enabling the mixing process to be adjusted for various reaction conditions. This degree of control makes it possible to optimize the production of biodiesel even further and may also make it possible to process other biofuels in microfluidic systems more effectively [36].

2.1.2. Piezoelectric micromixers

Piezoelectric micromixers emerge as a compelling solution to solve the challenge happened micro-scale (micromixing) mixing [37]. These gadgets take advantage of the special qualities of piezoelectric materials, which produce sound waves in the presence of an electric field. Sound waves are introduced into the reaction chamber by carefully integrating these materials into the walls of the microreactor. The laminar flow is disturbed by these sound waves, which causes microstreaming and turbulence in the fluid. This phenomenon encourages a more uniform distribution of reactants throughout the microchannels, greatly increasing mixing efficiency [38].

Piezoelectric micromixers have benefits beyond better mixing. These micromixers operate without physical contact, in contrast to traditional techniques that depend on moving parts. This removes the possibility of contamination brought on by the deterioration of the microreactor's stirring mechanisms. Furthermore, precise control over the electric field corresponds to precise control over the sound waves produced. This makes it possible to carefully tailor the mixing procedure for particular reaction conditions, guaranteeing effective biodiesel production regardless of feedstock or type of alcohol variations [30]. Piezoelectric micromixers also have built-in scalability. It is easy to adapt the design and fabrication principles to microreactors with different dimensions and configurations. They can be used in small-scale laboratory reactors and possibly large-scale industrial settings for the production of biodiesel due to this feature [39].

2.2. Passive micromixers

A typical component of a microreactor is a passive micromixer. It is also referred to as static micromixers [40] that accomplish mixing under the supplied pressure to cause fluid flow. To improve the passive mixing efficiency, these micromixers are made with different channel geometries that enhance the interfacial surface between the various fluids [41]. Due to the predominating laminar flow inside the microfluidic device, mixing in passive micromixers is mostly based on the diffusion of molecules and chaotic advection. Molecular diffusion may be improved by enhancing the contact surface between the various fluids and reducing the path of diffusion between the phases. The laminar flow in the micromixer can be manipulated to achieve chaotic advection. The flow pattern that results decreases the path of diffusion and enhances mixing. Each of the above micromixing techniques has various capacities, speeds of mixing, and operational parameters [24]. As a result, numerous researchers have been researching to create various methods to improve the effectiveness of mixing oil and alcohol throughout the course transesterification reaction [42].

Exploring novel channel geometries and flow manipulation techniques within passive micromixers can further enhance mixing

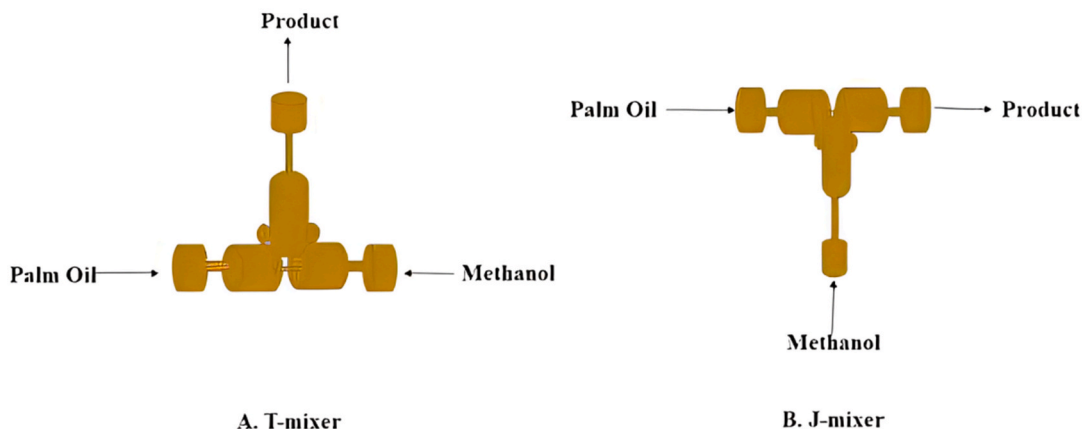


Fig. 2. A) T-mixer, B) J-mixer [44].

efficiency. This is particularly important for applications like the transesterification reaction in biodiesel synthesis.

Commonly, passive micromixing techniques that have been always employed for transesterification are multi-lamination based, and chaotic advection. Therefore, the next part of this review gives a full overview of two of the most prevalent forms of alcohol-oil micromixers, namely lamination-based and chaotic advection-based micromixers for biodiesel production.

2.2.1. Lamination-based passive micromixers

More efficient mixing via diffusion will be obtained by the production of tiny layers by dividing a major stream into numerous small streams or reducing the microchannel width in the flow direction for a single channel i.e. lamination. Thus, higher contact surface areas and tiny diffusional routes are created. Raising the contact surface areas or interfacial areas and reducing the diffusion route between them can improve molecular diffusion and enhance species mixing. Multilamination can be accomplished in practice by supplying the target samples through distinct holes, cantilever plate valves, or multiple channels in the micromixer, or by designing the various configurations on microchannel so that the target species are folded many times as they move along the micromixing channel. Such methods lengthen the time that the species are in contact with one another as well as the contact surface area between them. As a result, it is possible to undertake efficient species mixing typically within 55–300 ms [43]. As a result, micromixers have a big impact on many microreactor systems.

It is difficult to mix two immiscible phases in microreactors, such as alcohol and oil. T and J-type micromixers are most frequently utilized for injecting transesterification reactants. Fig. 2A and B shows how these mixers differ from one another. Oil and alcohol collide at a 180° impact angle in the T-micromixer, producing minute droplets as a result of the high pressure. J-micromixer, on the other hand, has a 90° impact angle. The bigger droplets are produced by the head-on contact at low pressure. Experiments were done to determine how geometric patterns, like hydraulic diameter and distribution of droplet size, affected the production of FAME. When compared to the J-type micromixer, the T-type micromixer demonstrated better mixing efficiency [44]. Seldom-used coaxial fluid mixing results in the circular movement of the droplets inside micromixers and has a 0° impact angle both of which contribute to effective mixing. Fig. 3 depicts a visual representation of co-axial fluidic mixing. particularly, it uses less energy than T and J-mixers to operate [45].

With the rise of methanol-to-oil molar ratio and temperature, the microchannel reactor having a T-micromixer demonstrated a greater oil yield. Moreover, due to intensive tiny droplet aggregation at the microreactor's exit, the segmented oil droplets flow and methanol at the inlet showed a quasi-homogeneous phenomenon. The results of a similar investigation using a T-micromixer with three various internal diameters, including 1.27 mm, 2.286 mm, and 0.33 mm, showed that the yield of fatty acid ethyl ester was highly influenced by the rates of mass transfer and the micromixer's internal diameter [46]. Also, comparisons and reports on the performance of micromixers such as T, cross, and Double-T-micromixer were made. Oil and alcohol move through two inlets in a T-micromixer, whereas oil flows through the middle inlet and alcohol through the other two inlets in a cross-micromixer. In a Double-T micromixer, alcohol flows via the other two inlets while oil moves through the other two alternate inlets. As shown in Fig. 4, three types of mixers were compared. Fig. 4a indicates the T-mixer, Fig. 4b illustrates the Cross T-mixer, and Fig. 4c shows the Double Cross T-mixer. The cross micromixer demonstrated the highest yield and mixing efficiency, which were determined to rise with residence time [23].

Gholami et al. [47] used a Plexiglas microreactor with a horizontal spiral-shaped microtube with an internal diameter of 0.8 mm, where the transesterification of Norouzak oil took place and a T-shaped joint micromixer for oil and methanol mixing. Fig. 5 displays the T-shaped joint micromixer with a horizontal spiral-shaped microtube. A biodiesel yield of 96.8 % was achieved at the operating conditions of 6.11%w/w CHOH catalyst, 1:0.37 methanol to oil volume ratio, and 12.48 min of residence time [47].

In conclusion, lamination-based passive micromixers significantly boost microreactor mixing efficiency, creating tiny layers and increasing contact surface areas. T-micromixers excel in mixing efficiency over J-type micromixers, and coaxial fluidic mixing offers an energy-efficient alternative. T-micromixers in microchannel reactors demonstrate enhanced oil yield with higher methanol-to-oil ratios and temperatures. Comparative evaluations highlight the superior yield and mixing efficiency of the cross-micromixer.

Rahimi et al. [48], on the other hand, for the transesterification of soybean oil with methanol using KOH homogeneous base

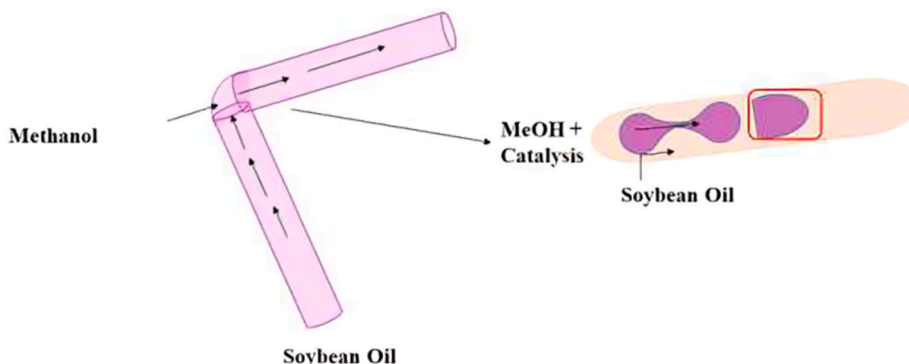


Fig. 3. Co-axial micromixer [45].

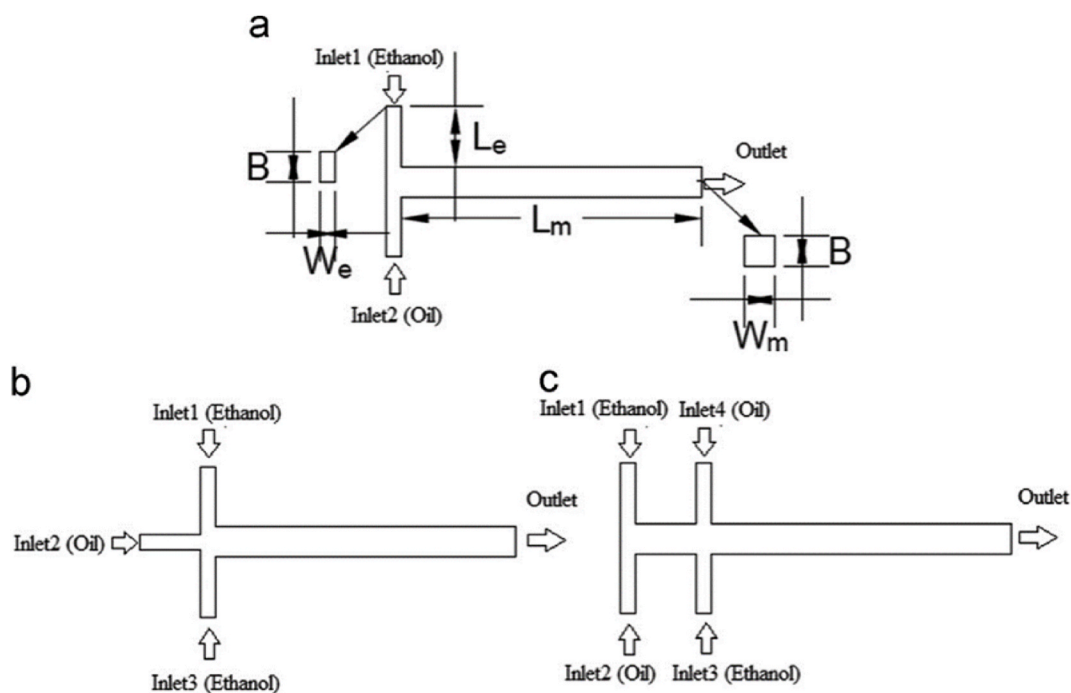


Fig. 4. a) T-mixer, b) Cross T-mixer, c) Double cross T-mixer [23].

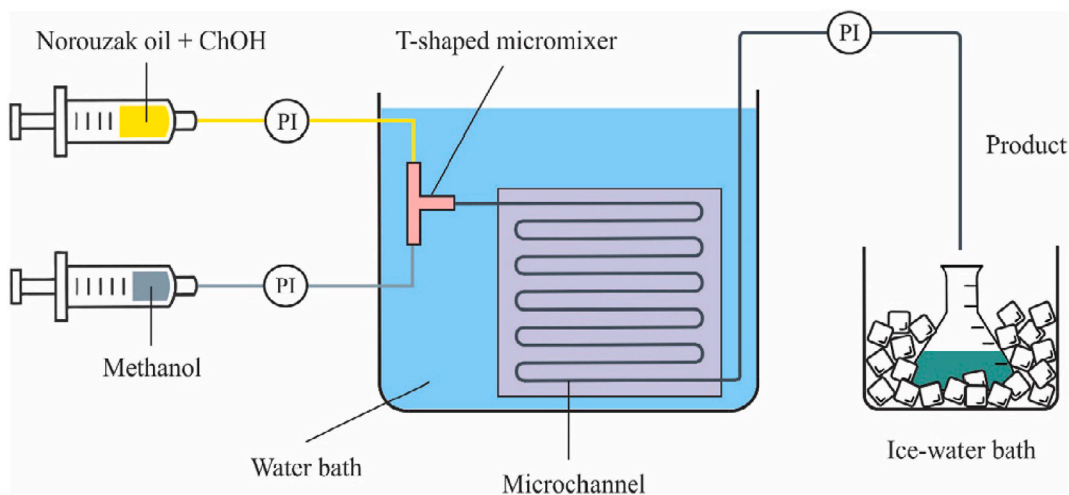


Fig. 5. T-shaped joint micromixer with a horizontal spiral-shaped microtube [47].

catalyst, a continuous flow microreactor that was made up of two components: A T-shaped joint micromixer and circular vertical spiral-shaped tubes with equivalent hydraulic diameters of 0.8 mm was used. Fig. 6 displays the micromixer and microreactor. The FAME yield reached 98 %, at the optimal operating conditions of methanol to oil molar ratio of 9:1, 1.2 wt% catalyst concentration, a temperature of 60 °C, and 3min residence time [48].

For the synthesis of biodiesel from sunflower oil and methanol, Guan et al. [51] employed a right angle to the horizontal spiral-shaped 300 mm long microtube reactor with a T-shaped junction micromixer for their study, which had an inner diameter of 0.8 mm as shown in Fig. 7. A FAME yield of 100 % was achieved under the following operating conditions: a methanol to oil molar ratio of 23.9:1, 4.5 wt% catalyst concentration, a temperature of 60 °C, a residence duration of 100 s, and an overall flow rate of 8.2 mL/min. The formation of the quasi-homogeneous phase is responsible for the higher yield which was formed in the oil and alcohol. In comparison, the microtube reactor outperforms the lab-scale batch reactor significantly better in oil yield [51].

The T-micromixer, J-micromixer, rectangular interdigital micromixer (RIMM) (Mikroglas, Germany), and slit interdigital micromixer (SIMM-V2) (IMM, Mainz, Germany) were the four different types of micromixers employed in biodiesel production micro

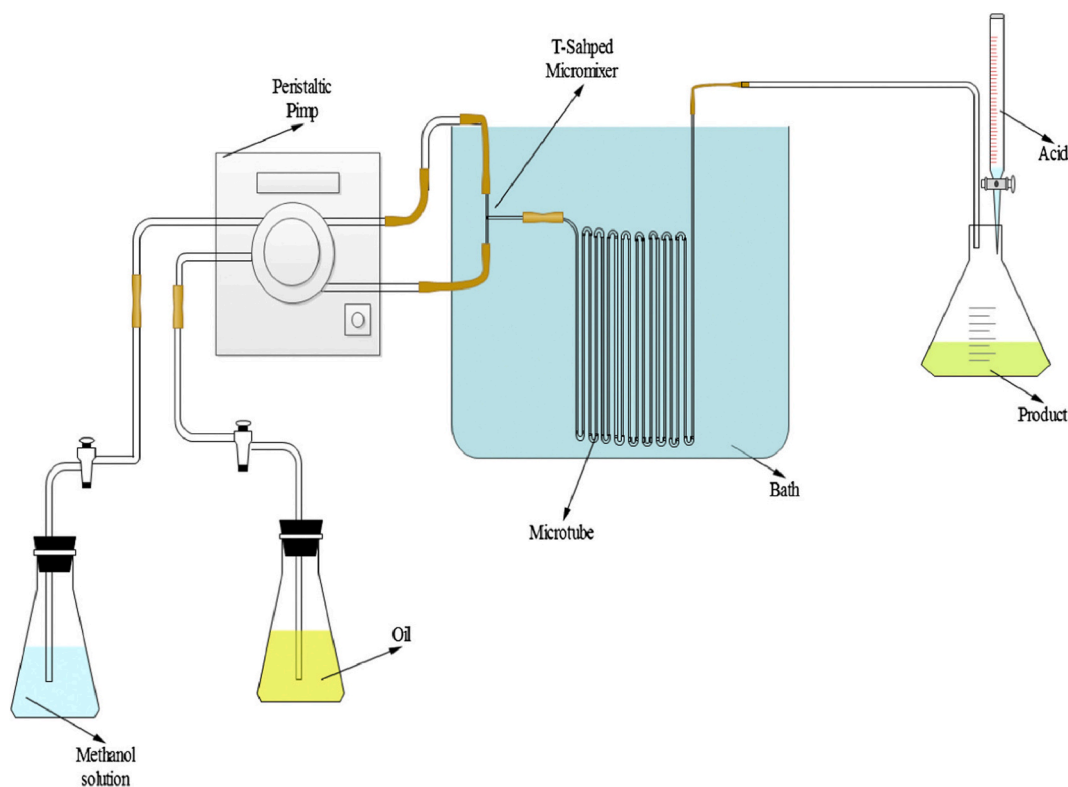


Fig. 6. Micromixer-microreactor system [48].

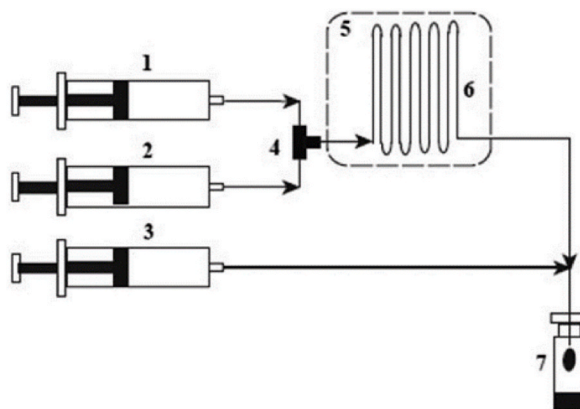


Fig. 7. micromixer and microreactor for transesterification: (1) methanol pump; (2) Sunflower oil pump; (3) Acetic acid pump; (4) T-mixer; (5) oil bath; (6) microtube reactor; (7) Biodiesel collector [51].

reaction system by Sun et al. [49]. Fig. 8 depicts a visual representation of these micromixers. Fig. 8a, b, 8c, and 8d show the configurations of T-mixer, J-mixer, RIMM, and SIMM-V2 mixers respectively. The rectangular micromixer used a rectangular sheet attached to the reactor to guide fluid through a large number of interconnected pores with a size of $60\ \mu\text{m}$ and into a microtube. The transesterification of cottonseed oil with methanol was done in a capillary delay loop constructed of stainless steel with an inside diameter of $0.6\ \text{mm}$ attached at the end of the micromixer. Here, the same reaction conditions: 8:1 methanol to oil molar ratio, 1 wt% catalyst concentration of KOH, the temperature of $60\ ^\circ\text{C}$, 44 s residence time, and an overall flow rate of $1\ \text{mL}/\text{min}$ in a 2.5 m long capillary delay loop were used with each of these micromixers. According to the findings, multi-lamination micromixers were more effective at producing biodiesel than straightforward T- and J-type micromixers. The yields of FAME produced by the T-micromixer and J-micromixer were comparable and less than half of those produced by the rectangular and slit interdigital micromixer. It appears that using the rectangular and slit interdigital micromixer significantly enhanced the surface area of contact between the methanol and

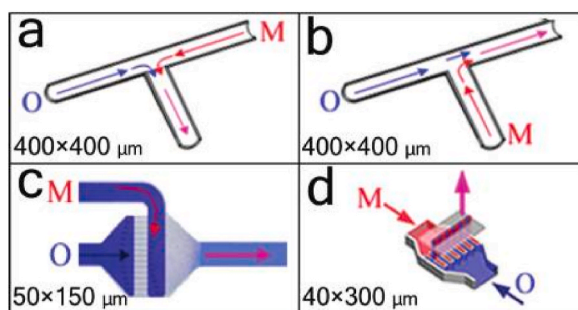


Fig. 8. Micromixers channel dimensions (width × height): a) T-mixer, b) J-mixer, c) RIMM, and d) SIMM-V2. M, methanol inlet; O, oil inlet [49].

oil, leading to larger FAME yields. At the optimum reaction conditions of methanol to oil ratio of 8:1, reaction temperature of 70 °C, and residence duration of 17 s, the multi-lamination type offered a biodiesel yield of 99.5 % [49]. When a multi-lamination flow channel is created several narrow diffusion channels are developed. More oil and alcohol diffuse when the diffusion channel is smaller, producing more biodiesel than other processes.

The split-and-recombine (SAR) micromixer, which is based on lamination, is a passive device that can effectively mix reactants at low Reynolds numbers (below 10) with a sluggish flow rate [52]. By dividing the fluids into several sub-channels and then merging the channels to create a single channel, this kind of micromixer is intended to carry out passive mixing. One type of lamination-based SAR micromixer that is frequently used is the Y-shaped micromixer [53,54]. A lamination-based SAR passive micromixer with a Y-shaped intake, as shown in Fig. 10, was studied by Subramaniam et al. [55]. Two distinct reactants are received in separate flows by the Y-shaped intake, which mixes them into a single flow. The reactor is made up of an output module (OM) at the bottom, an input module (IM) layer at the top, and stacked microchannel layers termed enhancement modules (EMs) in the middle [34]. The goal of the stackable module EM layer is to enhance reactant mixing while having a compact reactor design. Furthermore, because of its various stackable layers, the EM module is a great way to improve mixing by facilitating greater molecular diffusion of reactants [55].

The angle at which the micromixer impacts the surface determines the extent of mixing and the size of the droplets that are generated. The Y-junction micromixer has an impact angle of less than 90°. A crucial element regulating droplet size in microreactors is the residence time, denoting the mean duration of reactant in the microchannel from the start of the micromixer to the output. The length of the channel and the combined flow rates of alcohol and oil are often adjusted to manage the residence time. The mixture flow will experience a pressure reduction as the channel length grows, hence lowering the flow rates and mixing potential [55].

Three different microchannel reactor designs were examined by Yusuf et al. [56] for the possibility of producing biodiesel from vegetable oil. Three different designs of polymethyl methacrylate acrylic sheets were laser cut to create the microchannel reactors: (1) T-mixer, (2) Y-mixer, and (3) Tesla-shaped. A water bath was utilized to maintain a reaction temperature of 60 °C for the transesterification reaction, which was conducted with a sodium hydroxide catalyst loading of 1.0 wt percent. The Tesla-shaped microchannel design had the highest biodiesel yield of 97.9 % at 4.85 s. The biodiesel yields of the T-mixer and Y-mixer designs were 93.3 % and 94.3 %, respectively. The superior mixing efficiency of reactants was facilitated by the design geometry of the Tesla-shaped reactor, which had grooves and valves [56].

A microchannel reactor made of zig-zag mixers was used for alkali-catalyzed transesterification. Continuous multi-lamination mixing in the microreactor channel significantly enhances biodiesel yield compared to a single mixer. The process can be ramped up by adding more parallel couples of micromixer and reaction sheets because the resistance in the microreactor sheet is larger than in

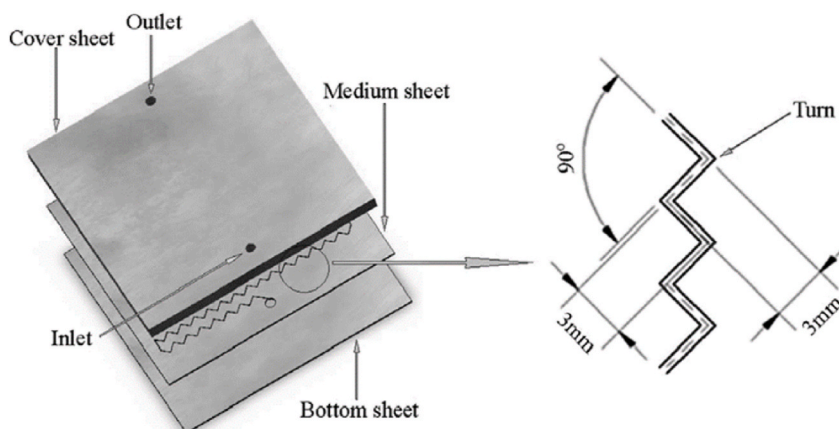


Fig. 9. Zigzag micromixer and microreactor [50].

the mixer sheet. Splitting the flow into the micromixer units can lower the maldistribution of the flow. Due to the increased rate of mixing in the interior of zig-zag channels, the zig-zag microreactor significantly minimizes the maldistribution of the flow and performs exceptionally well for transesterification [57].

For the production of biodiesel from soybean oil and methanol, electric-spark-made stainless steel, zigzag microchannel reactors with 10, 50, 100, 200, and 350 periodic numbers of turn and a hydraulic diameter of 240–900 μm were used. As seen in Fig. 9, this mixer's right-angled point where pumping oil and methanol collide functions like a multi-lamination mixer. All of the microchannels were identical in length (1.07 m), but they varied in their diameter or zigzag turns. The microchannel with the most turns (350) for the identical diameter, length, and operating conditions was determined to function best. The zigzag microchannel with the shortest diameter was determined to perform best for the same number of turns and identical operating conditions. At 56 °C reaction temperature, 9:1 alcohol to oil molar ratio, 1.2 wt% NaOH, and 28s residence time, a yield of 99.5 % was attained in a reactor with 240 μm hydraulic diameter. This happened, because, more zigzag patterns increase the amount of time that molecules of alcohol and oil are in contact with each other, and smaller equivalent hydraulic diameters decrease diffusion path diameters [50]. This implies that the microchannel with the shortest diameter proved most efficient for the same number of turns and conditions. Increased zigzag patterns enhance contact time between alcohol and oil molecules, improving biodiesel yield. Smaller hydraulic diameters reduce diffusion path lengths, further enhancing reaction efficiency.

2.2.2. Chaotic advection (internal) passive micromixers

The transport of molecules within a flowing fluid is called advection. Since it often happens in the flow direction, it has no impact on the substance's transverse transit. A transverse flow component can, however, be produced by advection in other directions called chaotic advection. The contact surface areas grow exponentially as a result of the created transverse components flow, and the striation thickness decreases as a result, which can greatly enhance mixing [24]. At small Reynold numbers, chaotic advection inside the micromixer is typically produced by altering the microchannel geometry for stretching, folding, splitting, and flow breaking [43]. These alter or reorganize the fluid flow to shorten the diffusion length and increase the contact surface area between the fluids [58].

The numerical investigation of two configurations of micromixers by Martinez et al. [2] presented tesla and omega chaotic advection micromixers with varied geometry in microchannel reactors, to enhance the mixing within the channels. The internal structures of different micromixer-microreactors were examined in Fig. 11a, b, and 11c. To achieve effective mixing, they were organized progressively. Due to the split and redirection of fluids, Tesla-shaped channel inclined surfaces developed effective collision between fluids. Due to the unstable fluid flow velocities in omega-shaped channels, the fluid forms vortices, which separate the flow and cause it to converge in the next channels [2].

Circular obstacles were used to promote micromixing, and the results were reported by Santana Harrson S et al. [42]. The numerical investigation of two configurations of micromixer design, MCO with three obstructions and MWO with seven obstructions (configurations: 35 mm in length, 1500 mm in width, and 200 mm in height), was conducted. Fig. 12a, b, and 12c depict a visual representation of these micromixers. Because of stream splitting, recombination, and vortices development, simulation outputs demonstrated that the MCO configuration performs better than MWO configurations in terms of fluid mixing degree, ultimately leading to higher vegetable oil yield in the production process of biodiesel. With a maximum extent of 99.99 % biodiesel yield, experimentally determined results demonstrated high ethyl esters yield for both MCO and MWO micromixer designs. With the MCO setup, FAEE yield increased as the ethanol-to-oil molar ratio increased from 6:1 to 9:1, but FAEE declined as the molar ratio further increased. Production of biodiesel was decreased at catalyst concentrations higher than 1 %. As a result, the yield of biodiesel synthesis is increased by the employment of micromixers with circular obstacles [42].

To perform continuous transesterification of soybean oil with methanol at a fixed molar ratio of methanol to oil of 6:1 and reaction temperature of 60 °C, Santacesaria et al. [59] developed a tubular reactor filled with stainless steel spheres as static micromixers with different diameters. An intense local mixing was achieved in microchannels within the 300–1000 μm range by varying the spherical diameter of micromixers. According to the research, static mixers that produced strong local micromixing promoted the growth of a large interfacial area, resulting in very high biodiesel conversion at mild temperatures of 60 °C and very short residence times [59]. Bishoge et al. [60] investigated the production of biodiesel from soybean oil using a continuous microflow system (MFS) (slit plate

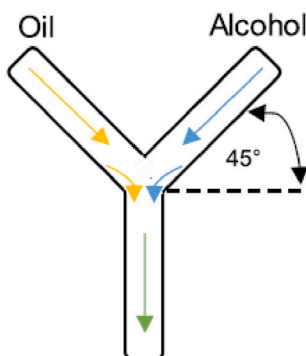


Fig. 10. Y-junction type micro mixer [55].

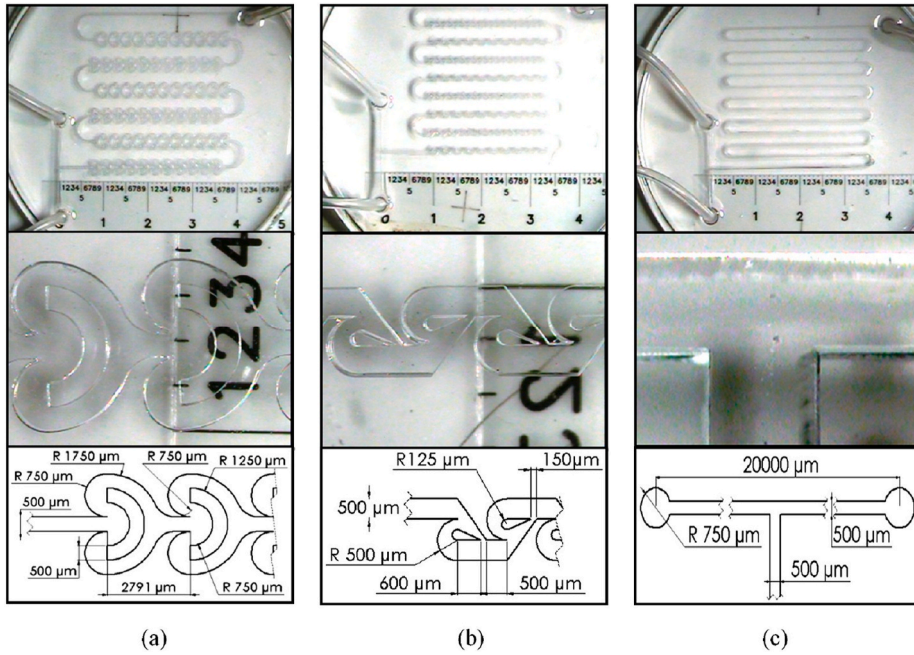


Fig. 11. Micromixer-microreactor internal structure: a) omega-shaped, b) Tesla-shaped, and; c) T-shaped [2].

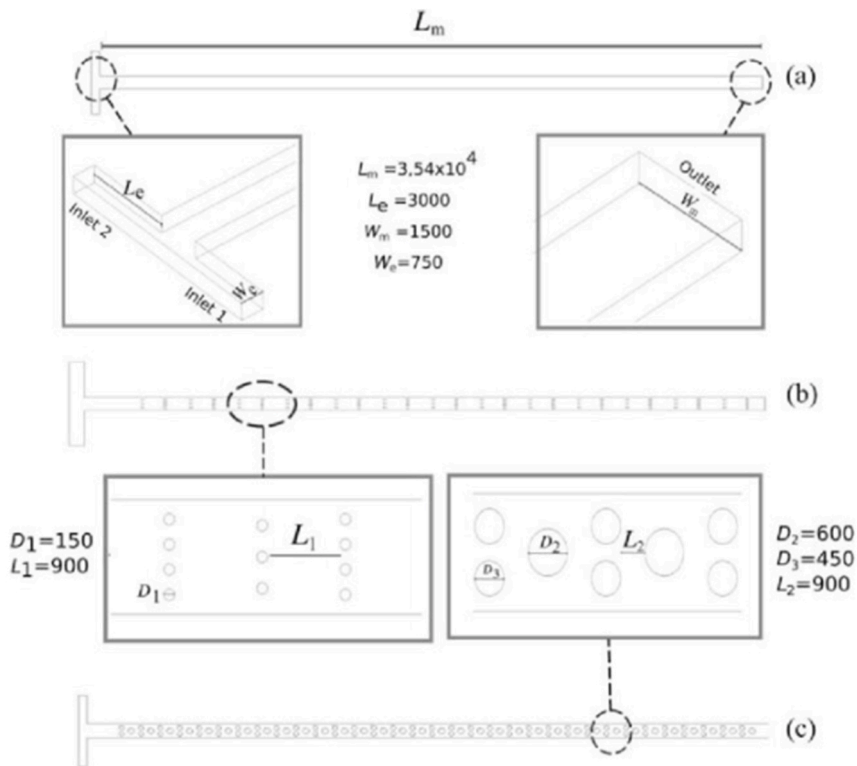


Fig. 12. Micromixer configurations: a) T-shape; b) MWO; c) MCO [42].

Table 1
Comparisons of different micromixers.

Types of micromixer	Micro mixer dimension	Types of passive micromixer	Feedstock	Co-solvent	Catalyst	Methanol-to-oil molar ratio	Temperature °C	Flow rate	Residence time (minute)	Yield %	reference
T-shaped	–	Lamination	–	–	6.11%w/w CHOH	1:0.37	–	–	12.48	96.8	[47]
T-shaped	0.8 mm	Lamination	–	–	1.25 wt% KOH	9:1	60	–	3	98	[48]
T-shaped	0.8 mm	Lamination	Sunflower oil	–	4.5 wt% KOH	23.9:1	60	–	1.67	100	[51]
zigzag	240 µm	Lamination	Soybean oil	–	1.2 wt% NaOH	9:1	60	–	0.47	99.5	[50]
†-shaped	–	lamination	Sunflower oil	–	KOH	10.3:1	60	0.05 ml/ min	5	98.48	[20]
SIMM-V2	–	Chaotic advection	cottonseed oil	–	1 wt%	8:1	70	–	0.283	99.5	[49]
Wire coil	L = 0.3 m, i.d = 0.5 mm	Chaotic advection	–	–	1.2 wt%KOH	9:1	60	–	3	99	[21]
Ribbon wool	–	Chaotic advection	–	–	2wtKOH%	6:1	60	8.33 ml/ min	3	97.05	[63]
Dixon rings	–	Chaotic advection	–	–	–	8:1	70	10 ml/ min	0.283	99.5	[49]
T-shaped + CaO packing	–	Chaotic advection	Palm oil	–	30 mg CaO	24:1	65	–	8.9	99	[62]
T-shaped + CaO packing	–	Chaotic advection	Palm oil	14.5 wt%THF	30 mgCaO	20:1	65	–	6.5	99	[62]
confluence angles	–	Chaotic advection	Soybean oil	0.45v/v Hexane to methanol	1 wt% KOH	3:1v/v	57.2	–	9.05	98.8	[64]

micromixer) and methanol catalyzed by sodium methoxide (CH_3ONa). Five factors influencing biodiesel yields were examined: temperature, methanol to oil molar ratio, aperture plate size, CH_3ONa dosage, and residence period. In comparison to batch studies, it has been found that the biodiesel yield might achieve a greater value with a substantially shorter residence period [60]. Furthermore, the trend for residence duration and the effect of CH_3ONa concentration on biodiesel yield was comparable. Furthermore, raising the methanol-to-oil molar ratio may boost the output of biodiesel; nevertheless, an excess of methanol resulted in a drop in biodiesel yield. Table 1 displays a comparison of biodiesel yields obtained from various types of micromixers, along with the dimensions of the micromixers.

The chaotic micro mixer type was created and was built of PVC by Boukhalkhal et al. [61]. It had ten periods of the C shape geometry (S-3D base element) and 10 mm equivalent hydraulic mean diameter with a square channel cross and 870 mm unfolded length (Fig. 13). The study setup for continuous biodiesel production and the chaotic micromixer-reactor topography are illustrated in Fig. 13a and b respectively. A pipe reactor (stainless steel pipe) was connected to the C-shaped geometry to ensure adequate residence time. The greatest ester content of the biodiesel produced at the operating conditions: 6:1 methanol to oil molar ratio, 1 wt% KOH, a temperature of 50 °C, and 3 g/s optimum mass flow rate of rapeseed oil was 99 % [61]. The yield of biodiesel was 99.5 % when the methanol to oil molar ratio was 8:1, the reaction temperature was 70 °C, the flow rate was 10 mL/min, and the residence period was 17 s, and 0.7 MPa pressure drop using a PTFE microtube filled with Dixon rings as the delay loop. Consequently, packed bed micro-structured reactors can produce biodiesel at a high throughput rate relatively quickly [49].

The utilization of a packed bed microchannel reactor for biodiesel production (configuration of $60 \times 1 \times 0.5$ mm) filled with an activated CaO catalyst was investigated by Chueluecha et al. [62]. The T-micromixer was used to feed the reactants, and 30 mg of catalyst powder was put into the channel. A schematic representation of the micromixer and reactor setup, including dimensions, is provided in Fig. 14. With different residence times, the activated CaO produced more biodiesel (90.5 % of biodiesel yield for activated CaO against only 82.7 % for non-activated CaO). Throughout 24 h of continuous operation, the catalyst continuously produced better yield. At the optimum residence time of 8.9 min, at a reaction temperature of 65 °C and 24:1 methanol to oil molar ratio, a yield of 99 % was achieved [62]. According to the results, producing biodiesel more efficiently can be achieved by using a packed bed microchannel reactor that is filled with an activated CaO catalyst.

To synthesize biodiesel, Santacesaria et al. [63] used a stainless steel cylindrical tube with a 20 cm length, and 10 mm interior diameter using a KOH catalyst. Fig. 15 illustrates a micromixers-reactor with a packed bed configuration, utilizing stainless steel ribbon wool as the chosen packing material. The internal void volume of the bare cylindrical tube reactor at first was 15.7 cm³. The reactor was filled with stainless steel ribbon wool that had been as tightly compressed as possible. With a molar ratio of methanol to oil of 6:1, 2 wt% catalyst weight, 60 °C reaction temperature, and a residence time of 180s, the yield was 97.05 %. Transesterification was performed without ribbon wool at an overall flow rate of 8.33 ml/min to investigate the impact of the packed bed's presence, and the amount of FAME produced was only 3 % [63]. This study underlines the significant impact of a packed bed microchannel reactor in enhancing biodiesel synthesis. The result indicates the substantial positive influence of employing a packed bed micro channel reactor on the process of biodiesel synthesis.

Soybean oil transesterification using ethanol and KOH catalyst in various micromixers was investigated by Pontes et al. [60]. The T (2.3 mm i.d. stainless steel channels), LTF-MS, and LTF-MX designs were the three that were examined. The inner diameter variation

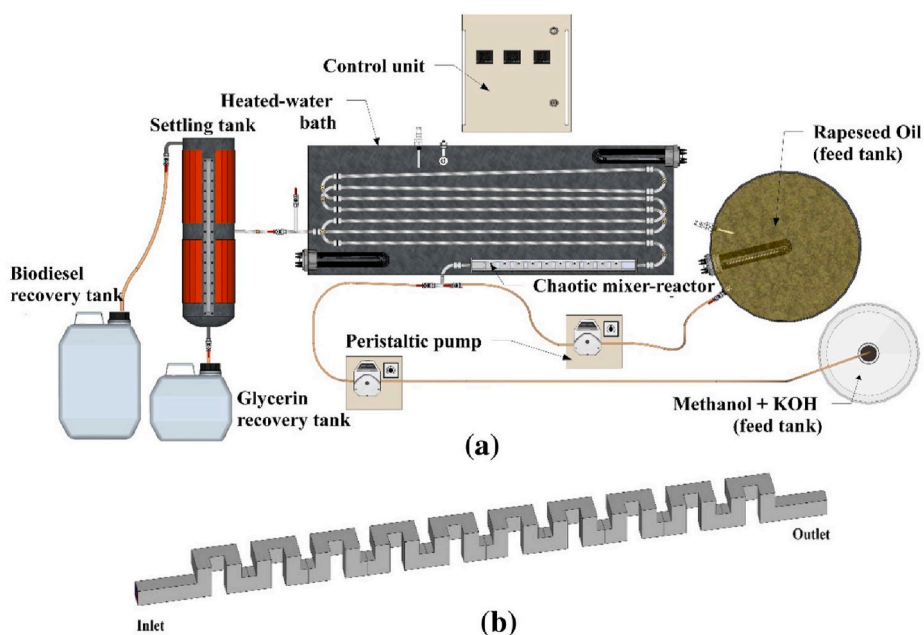


Fig. 13. a) Continuous biodiesel production experimental setup, b) Chaotic micromixer-reactor topography [61].

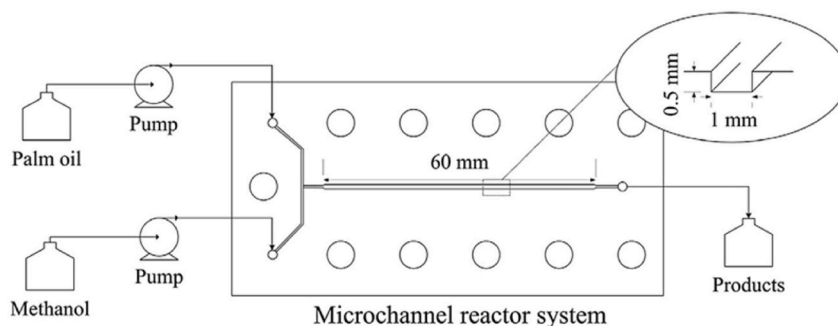


Fig. 14. Schematic picture of micromixer and reactor setup and dimensions [62].

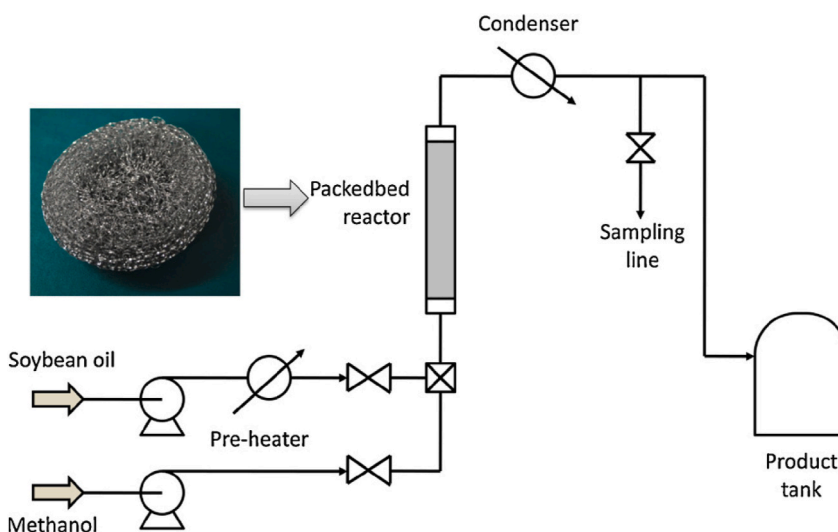


Fig. 15. Packed bed micromixer-reactor made of stainless steel ribbon wool as a packing material [63].

brought on by internal barriers generated by the LTF-MS arrangement encouraged fluid mixing. Fluid inlet stream splitting and recombination were imposed by the LTF-MX architecture. Pontes et al. [60] looked into the relationship between reaction yield and mixing efficiency, in contrast to the previously described studies. A modified Villermaux-Dushman method was used to calculate the mixing efficiency [65]. The LTF-MX micro-mixer demonstrated exceptional reaction yield and mixing efficiency. The authors elucidated the relationship between mixing and reaction yield, emphasizing the crucial function that mixing plays in the transesterification process due to the substantial influence of the mass transfer process.

3. Application of micromixer in biodiesel production

Micromixers facilitate quick and effective fluid mixing at the microscale. They have drawn a lot of attention in the field of biodiesel processing because of their distinct advantages over conventional mixing techniques, especially for purification purposes [39]. Micromixers facilitate chaotic advection and simplify mixing through the use of microchannels with precisely engineered geometries. As a result, the mixture becomes more homogeneous, which is essential for efficient purification procedures [66]. Regarding residence times, micromixers are smaller than traditional mixing tanks. As a result, the purification process proceeds more quickly and efficiently [2,31]. Micromixers' high surface-to-volume ratio allows better mass transfer between the purification agents and reactants. This enhances the elimination of contaminants and impurities [67]. Regarding energy consumption, micromixers are far less than traditional mixing techniques. This is because microfluidics allows for efficient mixing at lower fluid volumes [68]. Micromixers are easily scalable, allowing them to be adjusted to different process requirements. Additionally, they can be incorporated into bigger biodiesel production systems for automatic and continuous purification [69].

Micromixers are miniature devices that play a critical role in microfluidic systems by efficiently combining two or more fluids. To increase transesterification efficiency and improve reactant mixing (such as oil, alcohol, and catalyst), biodiesel production has used both passive and active micromixers [29]. Because of their affordability and ease of use, passive micromixers are frequently used, but active micromixers might be chosen in more difficult mixing situations involving reactants with high viscosities or strict mixing specifications. For the production of biodiesel, active-type micromixers have several advantages over passive-type micromixers.

Enhanced mixing efficiency, quicker mixing, and controlled mixing are some of these benefits. Active-type micromixers also face many challenges for biodiesel production such as being more expensive to fabricate, typically consuming more power, and generating heat, which can affect the temperature of the fluids being mixed [70].

Some types of passive and active type micromixers' advantages and disadvantages are summarized in Table 2.

4. Hydrodynamics of biodiesel micromixers

Interfacial forces between oil and alcohol, reaction temperature, flow rate, and the size and type of the micromixer-microreactor system are all important factors in the transesterification process's hydrodynamics. Different flow regimes can be developed depending on how the components vary. Parallel flow, slug flow, annular flow, intermittent slug flow, drop flow, and dispersed drop flow are the most prevalent forms of flow patterns [74,75]. According to Guajardo et al. [74], the slug flow forms when the interfacial force is greater than the system's inertial forces for the micromixer and microreactor. When inertial and interfacial forces are equal, parallel flow occurs. An annular flow forms when the inertial force dominates the forces at the oil-alcohol interface [74].

It is found that the types of micromixers have a significant impact on the biodiesel yield in microreactors. Researchers investigated the flow regimes of the T, J, RIMM, and SIMM-V2 micromixers. While the T and J micromixers in Fig. 16a and b showed a uniform slug at their outlet, the RIMM and SIMM-V2 micromixers created a slug and methanol droplets distributed in oil at their output as depicted in Fig. 16c and d respectively [49]. Due to the development of smaller slugs, the RIMM and SIMM-V2 micromixers produced a better biodiesel output by creating a larger surface area to volume ratio [49] which increases the rate of the reaction.

Rahimi et al. [48] looked at how oil yield during the transesterification reaction in the T-shaped micromixer was affected by flow rate and flow pattern. The mixing of oil and methanol at 1 ml/min, as illustrated in Fig. 17a, was ineffective because the two fluids create independent streamlines. However, it was effectively mixed at a flow rate of 11 ml/min, and a greater yield was attained (Fig. 17b) [48].

W. Shaaban et al. [76] looked into how a T-micromixer with three distinct dimensions (Table 3) affected slug size. The smaller the micromixer, or higher the surface-to-volume ratio, the smaller the slugs were. At 1.2 min of residence time under ideal reaction circumstances, the T1 (T-micromixer = 1.27 mm id) achieved 97 % oil yield. This shows that the FAME is influenced by inner diameter and shape [76].

In a zigzag micro mixer drop flow pattern, according to Wen et al. [50], 97 % FAME was produced in 28s of reaction time. The size of the droplet had a significant impact on how quickly the transesterification reaction occurred. The rate of the reaction increases with decreasing droplet size. However, other researchers investigated the oil-methanol mixture flow pattern in a T-shaped micromixer-connected capillary microreactor and discovered slug forms of flow, achieving 97 % in 10 min of residence time [50]. In addition, Kashid et al. [77] claimed that the slug-type flow was generated while using a T-micromixer. The delay loop's recirculating slugs accelerated the pace of mass transfer [77]. Using computational fluid dynamics simulation, Guajardo et al. examined the flow pattern of transesterification reactants in a T-micromixer connected microchannel reactor with an inner diameter of 710 μm and a length of 5 m. Slug flow was the predominant type of flow created under the operating parameters of 60 °C reaction temperature, 6:1 methanol-to-oil molar ratio, total volumetric flow rates of 0.5–4 ml/min, and 0.7%wt% NaOH catalyst. Over 99 % yield was attained in 4 min of residence time thanks to the slug's enhanced mass transfer rate during recirculation [74].

Variations in the methanol-to-oil molar ratio led to the development of various flow patterns in the T-shaped micromixer, as shown in Fig. 18. Micrographs of two-phase flow inside the microchannel at various methanol-to-oil molar ratios are shown in Fig. 18a and b

Table 2
Advantages and disadvantages type of passive and active type micromixers.

Micro mixer types	Advantages	Disadvantages	References	
Passive micromixer	Lamination-based	<ul style="list-style-type: none"> ✓ Simple Fabrication ✓ High mixing efficiency ✓ Design Flexibility ✓ Low-pressure drops 	<ul style="list-style-type: none"> ✓ Limited Mixing Control ✓ Bonding Challenges ✓ Limited Reusability 	[67]
	Chaotic advection	<ul style="list-style-type: none"> ✓ High mixing efficiency ✓ Low-pressure drops ✓ Lower energy consumption ✓ Allows for scalability of the mixing process 	<ul style="list-style-type: none"> ✓ Complex design ✓ Higher pressure drops ✓ Risk of clogging by impurities 	[33]
	Diffusive Micromixers	<ul style="list-style-type: none"> ✓ Simple design and fabrication ✓ Low-pressure drops ✓ Reliable and maintenance-free 	<ul style="list-style-type: none"> ✓ Limited Characterization ✓ Not suitable for high-viscosity fluids ✓ Limited mixing efficiency and slow mixing 	[71]
Active micromixer	Acoustic Micromixers	<ul style="list-style-type: none"> ✓ Moderate mixing efficiency and pressure drops ✓ Fast mixing ✓ Controllable mixing 	<ul style="list-style-type: none"> ✓ Moderate fabrication complexity ✓ Power consumption ✓ Not suitable for all fluids 	[34,35]
	Piezoelectric Micromixers	<ul style="list-style-type: none"> ✓ Very simple and miniature ✓ High mixing efficiency ✓ Low power consumption 	<ul style="list-style-type: none"> ✓ Expensive to fabricate ✓ Fragile and susceptible to damage ✓ Limited mixing volume 	[30]
	Electro kinetic Micromixers	<ul style="list-style-type: none"> ✓ Simple design and fabrication ✓ Fast and Efficient Mixing ✓ Precise Control 	<ul style="list-style-type: none"> ✓ External Power Source ✓ Electrode Compatibility ✓ Conductivity Dependence 	[32,72,73]

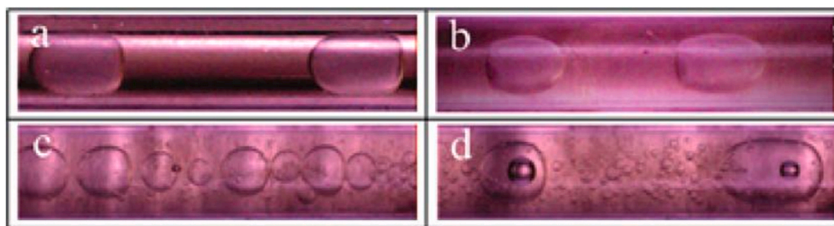


Fig. 16. Flow patterns in the transparent PVC tubes (inner diameter) 1.2 mm) at the outlets of different micromixers: a) T-mixer, b) J-mixer, c) RIMM mixer, d) SIMM-V2 mixer [49].

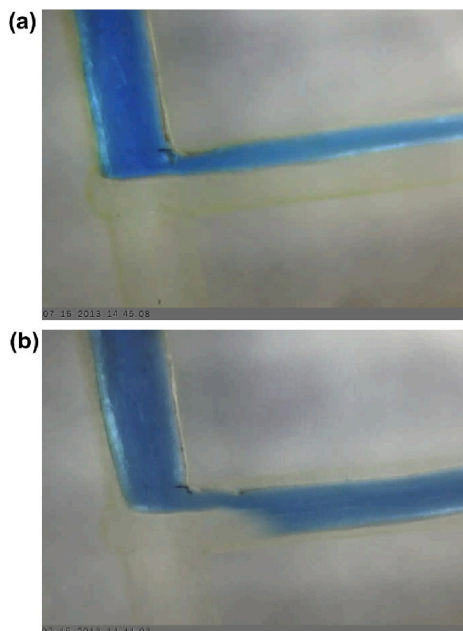

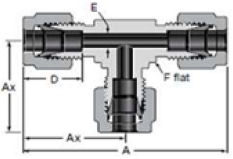
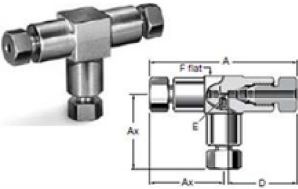


Fig. 17. The fluid flow pattern inside a T-shaped micromixer with two different flows [48].

18c, 18d 18e, 18f, and 18g. The lower molar ratio of methanol to oil (3.3–3.7) was where the droplet flow regime was produced. The droplet's size is smaller than the microreactor's inner diameter. However, a slug flow pattern evolved at larger methanol-to-oil molar ratios. Its length exceeds the microchannel's diameter. Due to the increasing volumetric flow rate of methanol, as seen in Fig. 19, the size of the slug grew as the methanol to oil molar ratio rose. Slug formation speed was influenced by the volumetric flow rates of methanol and longer intervals between successive slugs are caused by a bigger volume of oil present in a continuous phase, as shown in Fig. 20. With an increase in the methanol to oil molar flow rate, more slugs formed. This is because more slugs can form in the oil phase the more methanol there is in the continuous oil phase. As can be seen in Fig. 21, as the number of slugs increased, so did the extent of conversion since the slug's surface-to-volume ratio increased [75]. From this finding, it can be concluded that lower ratios yield a droplet flow, while higher ratios result in longer slugs, impacting conversion efficiency. Volumetric methanol flow correlates with slug size, affecting slug formation frequency and overall conversion. Increasing slugs enhance yield due to their higher surface-to-volume ratio.

In their work, Verma and Ghosh [78] discovered four different flow regimes in a T-micromixer-connected serpentine delay loop with an inner diameter of 2 mm and a length of 0.5 m. Similar, slender-shaped slugs were generated at a reduced surface flow rate of the dispersed phase (methanol) (Fig. 22 (i)). The slug flow pattern gradually changed to intermittent when the surface velocity of the dispersed phase (methanol) increased (Fig. 22 (ii)). Intermittent Slug Flow" develops at relatively higher superficial gas velocities compared to superficial liquid velocities. In this regime, the slugs are larger in length, creating an annular structure with a small bridge of the continuous phase between them. Drops of the dispersed phase follow the long slug (Fig. 22 (iii)). A different type of flow pattern called a droplet-dispersed flow developed at a greater superficial methanol velocity (Fig. 22 (iv)) [78]. Due to the large surface area to volume ratio, the slug flow pattern amplifies mass transfer and effectively speeds up the reaction [78]. This implies that, as the surface velocity of the dispersed phase increases, the flow pattern transitions to intermittent, and at higher velocities, a droplet-dispersed flow emerges. This information suggests that controlling the flow conditions in micromixers can be crucial for optimizing mass transfer and reaction efficiency in microreactor systems.

Table 3
Specifications of the micro mixers used [76].

No.	Scheme	Tube OD	Dimensions				
			A	Ax	D	E	F
Dimensions, in							
T1		1/6	1.4	0.7	0.34	0.05	3/8
T2		1/8	1.76	0.88	0.5	0.09	3/8
T3		1/16	1.32	0.66	0.61	0.013	3/8

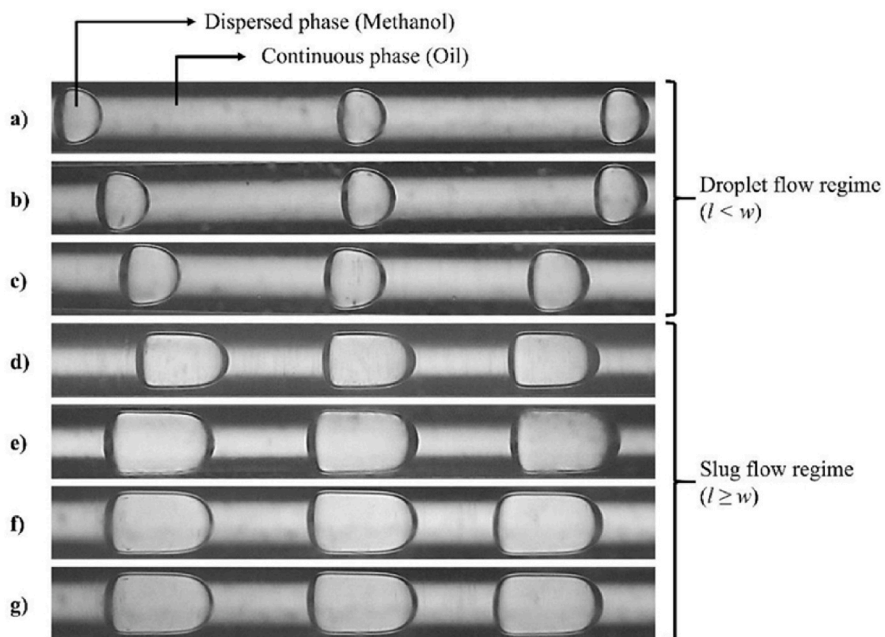


Fig. 18. Micrograph of two-phase flow inside microchannel at methanol-to-oil the molar ratio of a) 3.3, b) 4.6, c) 7.6, d) 10, e) 15, f) 20, and g) 22.9 [75].

5. Co-solvent in micromixers

The reactant fluids of the transesterification reaction are immiscible (oil and alcohol) to one other, which has a significant effect on the mass transfer resistance. This can be minimized by employing the co-solvent that has to be solubilized by both oil and alcohol to form a homogeneous phase during the transesterification reaction. Therefore, utilizing the right co-solvent can reduce the

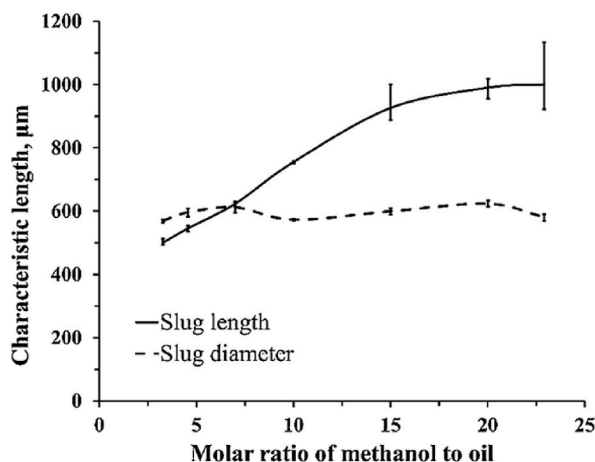


Fig. 19. The characteristic length of slugs at different methanol-to-oil molar ratio [75].

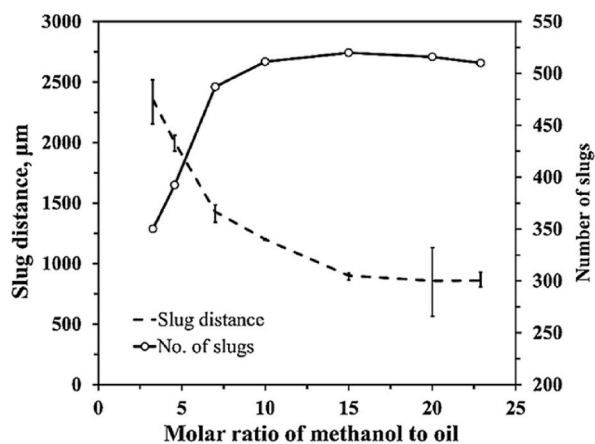


Fig. 20. The slug distance and the calculated number of slugs at different methanol-to-oil molar ratios [75].

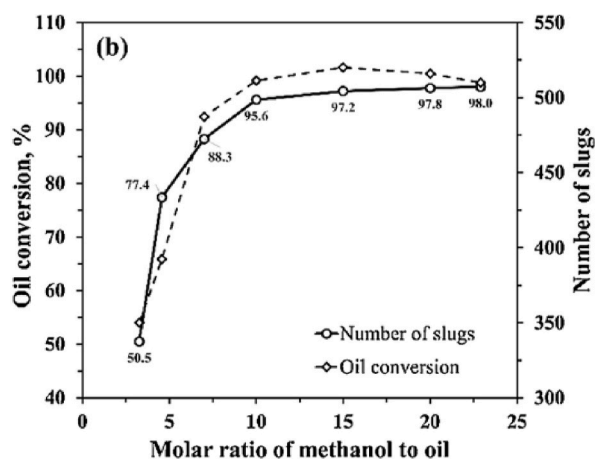


Fig. 21. Comparison between the effect of several slugs on the oil yield [75].

immiscibility of these reactants [79]. Several researchers have reported that co-solvent reduces effectively the reaction temperature, oil-to-alcohol molar ratio, operating pressure, and lowers the induction period, and improves the rate of the reaction due to the reduction of initial mass transfer resistance between oil and alcohol [80–82]. To utilize their advantages more efficiently some

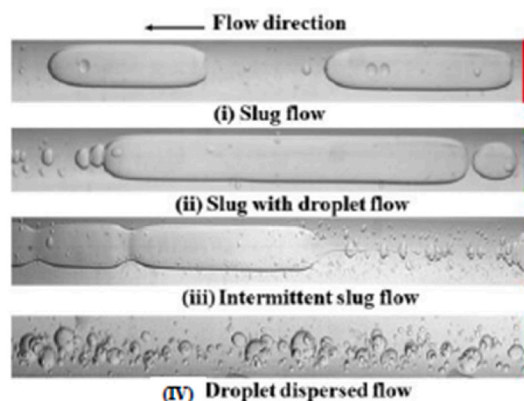


Fig. 22. Representative photographs of flow patterns [78].

researchers have employed co-solvents together with micromixers on microreactor systems for biodiesel production.

Verma & Ghosh [78] examined the effects of several co-solvents on the rate of reaction and the level of conversion using a microtube reactor with a T-shaped micromixer. Fig. 23 shows that oil yield reached its maximum value faster in the presence of co-solvent than it did in the absence of it. Co-solvents reduce the interphase mass transfer resistance, which is why this happens. The conversion in a microreactor with a T-shaped micromixer was also shown in Fig. 24 in both the absence and presence of several co-solvents. All types of co-solvent in the microreactor provided better conversion compared to the conversion attained in the absence of co-solvent. Furthermore, the effects of the co-solvent diethyl ether at 25 °C on the flow patterns of the combination of oil and methanol were investigated (Fig. 25a) and contrasted with those in the presence of the co-solvent. According to Fig. 25a, due to the presence of co-solvents in i & ii, a homogenous phase evolved at the exit of the T-shaped micromixer as a result of the reduced oil-methanol mass transfer resistance. Oil and methanol were less immiscible because of the co-solvent. As a result, at the third length of the microtube, biodiesel oil yield was attained at a rate of 63.4 %, and at the 36 cm length and 1.33 min residence time, it was at a rate of 92.8 %. The oil was not properly converted to biodiesel due to the formation of clear, stable, and segregated (Fig. 25b–i) up to 60 cm from the exit of the T-micromixer and bigger red methanol segregates (Fig. 25b–ii) at the exit of the microreactor in the absence of co-solvent. i.e., only 35 % yield was attained when the microreactor was shut off. Fig. 25c shows the location of the T-shaped micromixer in the homogeneous oil-methanol phase, with numerical values indicating oil yield [78].

Generally, the study demonstrated superior conversions with various co-solvent types compared to scenarios without co-solvent. Particularly, diethyl ether, operating at 25 °C, played a pivotal role in creating a homogeneous oil-methanol phase, resulting in an impressive 92.8 % conversion. Conversely, the absence of co-solvent led to incomplete conversion and segregated phases, demonstrating the critical role of co-solvents in optimizing biodiesel production efficiency within microreactors.

The effects of the co-solvent and T-micromixer were investigated by Aghel et al. [83] in the serpentine micro tube (Fig. 26; inner diameter = 0.8 mm). Tetrahydrofuran and n-hexane were both employed to lessen the oil-methanol mixture's immiscibility during the transesterification reaction, which was catalyzed by limescale. The biodiesel yield was 97.03 % at the operating conditions of 62 °C

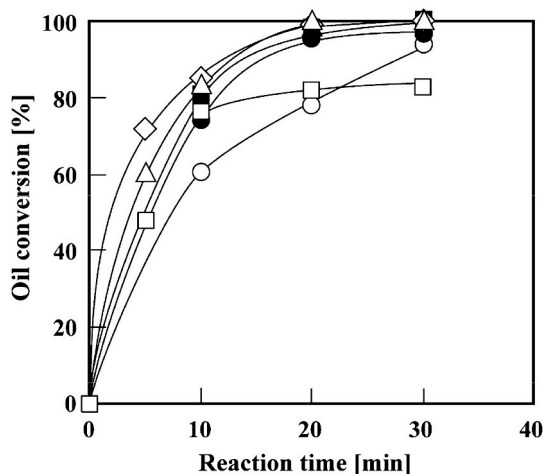


Fig. 23. Oil yield as a function of reaction time in the presence of various co-solvents. Reaction conditions: T = 25 °C, methanol to oil molar ratio (co-solvent): □,4(DME); △,6(DME); ◇,8(DME); ■,6(DME); ▲,6(TBME); ●,6(TBME); ○,6(no cosolvent),0.5 wt%KOH [78].

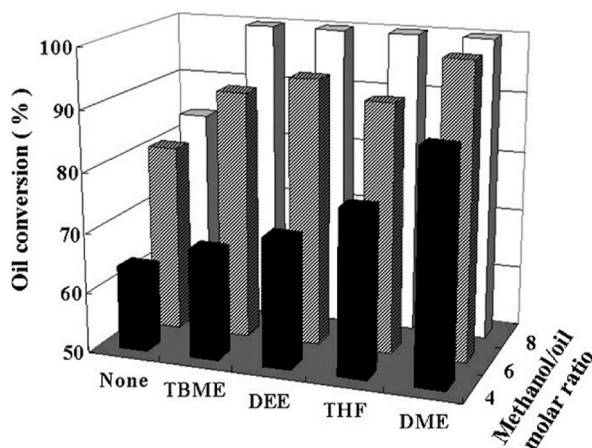


Fig. 24. Comparison of oil yield in the presence of different co-solvents.

Reaction conditions: shaking frequency = 2.6 Hz; reaction temperature = 25 °C; reaction time = 10 min; co-solvent/methanol molar ratio = minimum solvent/methanol molar ratio required for miscibility, 1 wt% KOH based on oil weight [78].

reaction temperature, 8.17 wt% limescale, 1:1.95 v/v methanol to oil, and 0.58:1 n-hexane to oil v/v in the presence of n-hexane. However, at operating parameters of 64.5 °C reaction temperature, 8.13 wt% limescale catalyst, 1:1.97 v/v methanol to oil, and 0.62:1 v/v tetrahydrofuran to oil, a biodiesel yield of 95.21 % was found after a 10-min reaction. The findings demonstrated the ability of n-hexane and tetrahydrofuran to shorten residence time and quicken reaction rate [83].

In a microtube reactor with an inner diameter of 1.5 mm, Basiri et al. [84] investigated the effects of hexane co-solvent and Y-micromixer (inner diameter = 0.8 mm) on the synthesis of biodiesel. According to Fig. 27, the investigations on the three different types of micromixers were carried out under the best possible conditions for reactions (I = no co-solvent in the Y micromixer, II = co-solvent present in the Y micromixer, III = co-solvent present on the three inlet micromixer). The introduction of hexane as a co-solvent created a homogenous phase of the oil-methanol combination, which decreased the resistance to mass transfer. The hydrophobicity of hexane to move over oil weakened oil-oil interaction [84]. Hexane facilitates the formation of a pseudo-homogeneous reaction mixture, which makes it simpler for methanol to enter the oil phase. Hexane serves as a carrier for the oil's dispersion rather than taking part in the reaction itself. Hexane can also move between oil molecules with ease because it is hydrophobic, which reduces the cohesive forces between oil molecules. Hence, hexane boosted biodiesel yield by 13 wt% when added to the three-micromixer intake and by 23 wt% when added to the Y-micromixer because it reduced mass transfer resistance and had a short diffusion distance [84]. This improvement was attributed to reduced mass transfer resistance and a shorter diffusion distance facilitated by hexane. Overall, the study illustrates hexane's positive impact on enhancing biodiesel production efficiency in microreactors.

The oil's immiscibility in alcohol limits the mass transfer during the transesterification event. The use of a co-solvent may reduce mass transfer resistance and speed up the process [80,85]. The T-shaped micromixer coupled microtube reactor, which had an 800 m diameter and a 5 m length, used acetone as a co-solvent. FAME increased when acetone's weight percentage rose from 10 % to 14 %. However, a further rise led to a decrease in FAME because of the dilution effect, which restricts access to methanol and oil. The operating conditions of a 12.5-min reaction time, a temperature of 60 °C, a methanol to oil volume ratio of 2.15:5, and a ratio of 13.95 wt percent of acetone to methanol resulted in the production of 97.16 % FAME. As shown in Fig. 28, a greater FAME content was created in less time when a co-solvent (hexane) was present. This demonstrates how the transesterification reaction can be improved by a co-solvent [64].

By increasing the miscibility of the phases, the reaction rate increases due to the disappearance of mass transfer resistance between the phases [80,85]. By employing micromixer and co-solvents, mass transfer resistance can be reduced. Short diffusion distance in micromixer-microreactor makes oil and alcohol molecules diffuse fast. The transesterification reaction conversion increases during this situation. Moreover, the use of a micromixer and co-solvent in the microtube reactor leads to an increase in the rate of the reaction [64].

The utilization of a packed bed microreactor with a configuration of 60 × 1 × 0.5 mm packed with an activated CaO heterogeneous solid catalyst was investigated by Chueluecha et al. [86]. A catalyst powder weighing 30 mg was dumped into the channel before the reactants were delivered. For all different residence times, the activated CaO produced more biodiesel yield (90.5 % biodiesel yield for activated catalyst against only 82.7 % for non-activated catalyst). Throughout 24 h of continuous operation, the catalyst continuously produced better yield. At the optimum operating condition of: 8.9 min reaction time, 65 °C reaction temperature, and 24:1 methanol to oil molar ratio, an oil yield of 99 % was achieved. The effects of several co-solvents, such as tetrahydrofuran (THF), ethyl acetate, and iso-propanol, were investigated using the same microreactor configuration i.e. packed bed microchannel reactor. The maximum biodiesel yield was obtained at a residence period of 7.1 min when 40 wt% THF was fed to the reactor; at the same residence time, 20 wt% iso-propanol produced the highest FAME yield.

In contrast, ethyl acetate was used with a residence duration of 8.9 min and a maximum biodiesel production of 20 wt%. Results showed that, compared to other co-solvents, Iso-propanol was the most effective co-solvent, providing a higher degree of mixing

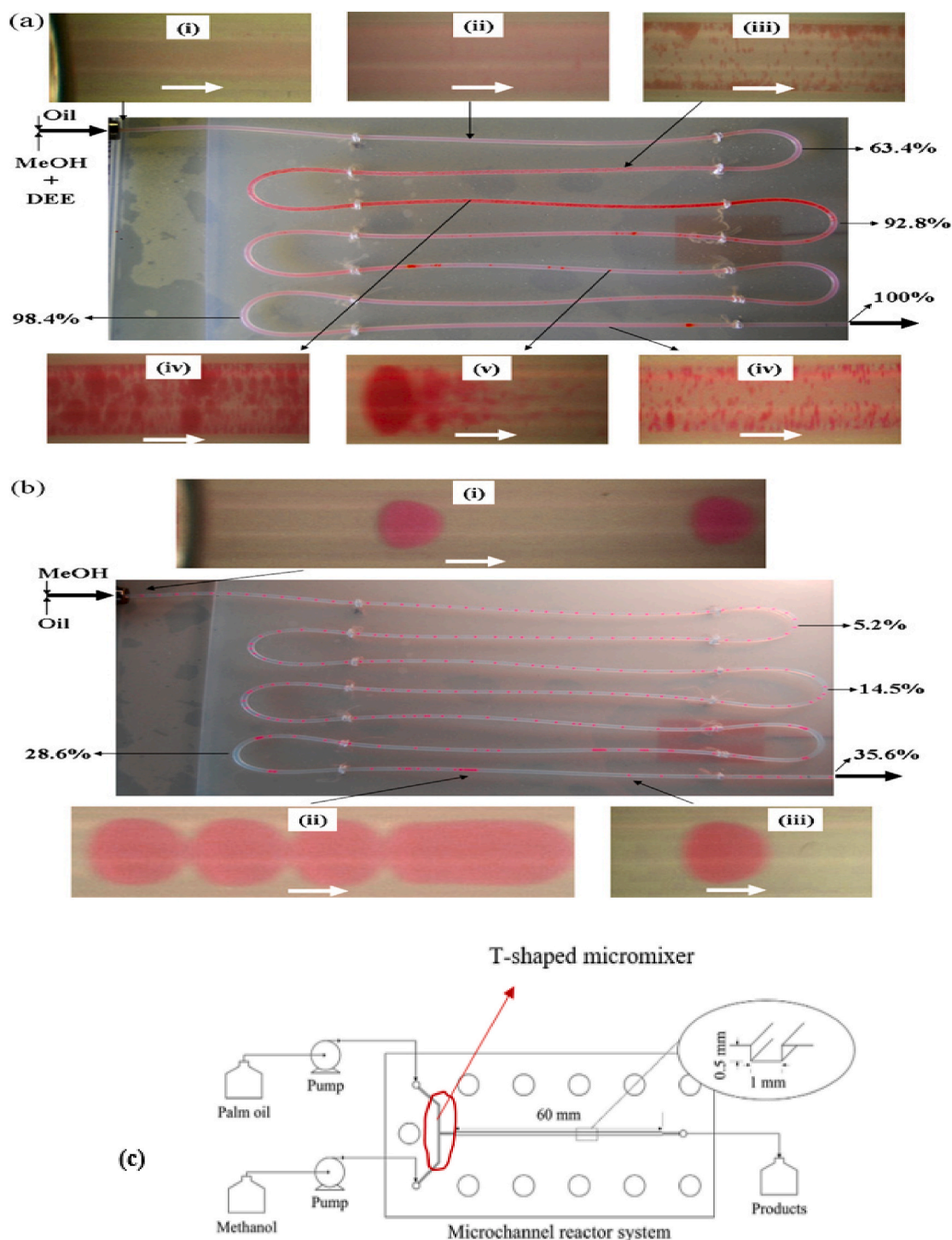


Fig. 25. Microscopic images of flow behavior in a transparent FEP microtube reactor in the absence (a) and presence (b) of DEE. DEE/methanol molar ratio = 0.73; methanol/oil = 8; total flow rate = $10 \text{ cm}^3/\text{h}$; reaction temperature = $25 \text{ }^\circ\text{C}$; (c) location of T-shaped micromixer in homogeneous oil-methanol phase. The numerical value indicates oil yield [78].

efficiency. Moreover, when the reaction was run at shorter residence durations, unfavorable effects on FAME yield were seen when co-solvents such as THF and ethyl acetate were used. While the reaction efficiency improved due to shorter residence times and a higher methanol-to-oil ratio, the yield was improved by premixing the oil and co-solvent. 99 % pure fatty acid methyl esters were produced at the optimal process parameters of 6.5 min residence time, methanol to oil molar ratio of 20:1, and co-solvent of 14.5 wt% [86]. Hence,

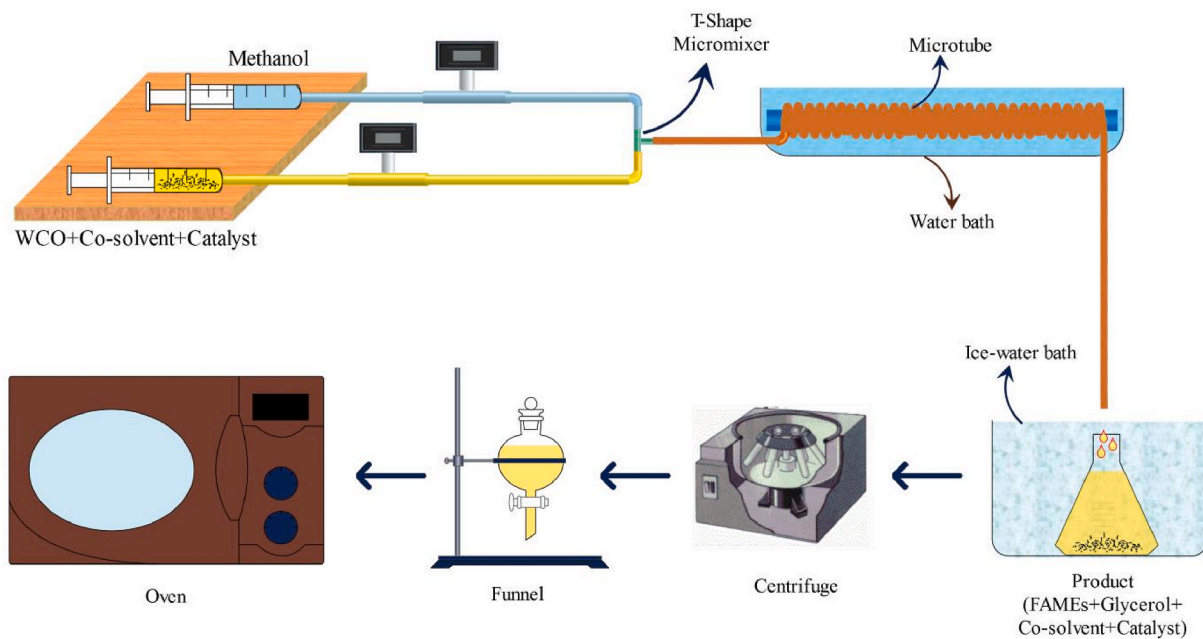


Fig. 26. Schematic diagram of biodiesel production [83].

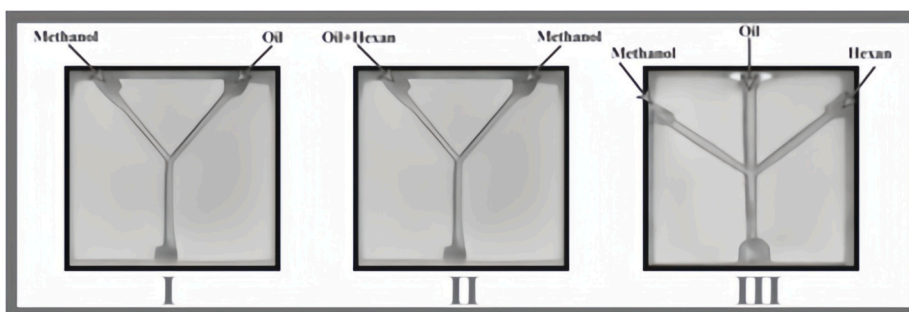


Fig. 27. Mixer configurations and mixing forms of the reactants in different experiments [84].

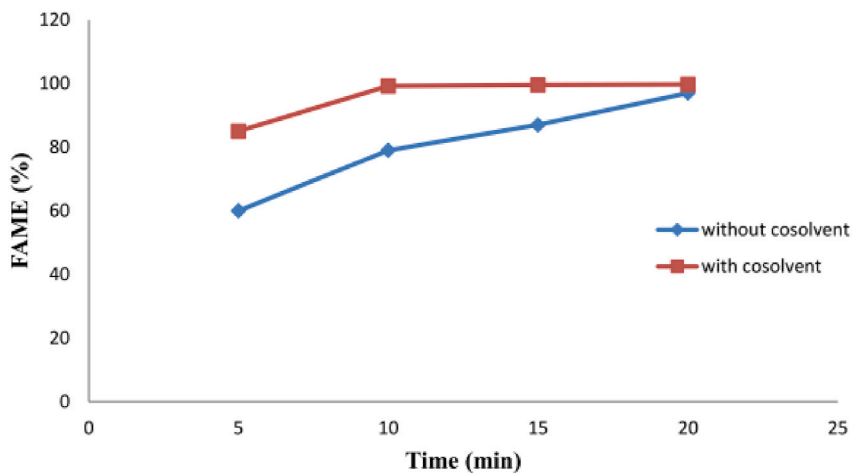


Fig. 28. Effects of co-solvent on biodiesel yield [64].

shorter residence times and higher methanol-to-oil ratios improved reaction efficiency, and premixing oil with co-solvent further enhanced yield, producing 99 % pure FAME at optimal parameters.

Hexane was used as a co-solvent in three different types of micromixers with confluence angles of 135°, 90°, and 45° to increase mixing effectiveness. The different confluence angle is shown in Fig. 29. Three kinds of micromixers, made on a flat plate by poly-methyl methacrylate, were studied. The configurations involved varying the angles at which three fluid inlets: methanol, oil, and hexane met. The confluence angles of 45° (E1 configuration), 90° (E2 configuration), and 135° (E3 configuration) were employed [64].

Methanolysis of soybean oil was carried out in the presence of a homogeneous KOH catalyst adding hexane as a co-solvent. The maximum yield of 98.8 % was obtained, provided by confluence angles of 45° at the reaction temperature of 57.2 °C, 1 wt% KOH concentration, residence time of 9.05 s, oil to methanol ratio of 3:1v/v and hexane to methanol ratio of 0.45v/v. The hydrophobic property of hexane affects the interaction between the molecules to keep the fluids' homogeneity, as evidenced by the improved performance at 45°, followed by 135° and 90°. The addition of co-solvent can improve oil-methanol miscibility which can help micromixers perform better in mixing within a short residence time [64]. This reveals that The addition of hexane as a co-solvent improved oil-methanol miscibility, leading to enhanced micromixer performance within a short residence time. This study underscores the significance of co-solvents in optimizing micromixing processes for biodiesel synthesis.

Also, the study investigated by Santana Harrison S et al. [87] the use of solid acids (SAC-13) as a catalyst for triglyceride transesterification with compressed methanol. The addition of a co-solvent, supercritical carbon dioxide, accelerated the reaction rate, enabling high biodiesel yields at mild temperatures. Continuous experiments in a fixed bed reactor were conducted at 150–205 °C, 6–24 ml/min flow rate, and 25 MPa pressure. Optimal conditions of 200 °C temperature and 2 min' reaction time achieved 99.4 % triglyceride conversion and 88 % methyl ester yield. The reaction rate was 20 times faster than conventional methods, attributed to reduced transport resistance facilitated by the co-solvent [87]. This study underscores the effectiveness of co-solvents in enhancing transesterification efficiency and accelerating reaction rates under mild conditions, offering promising advancements for biodiesel production.

Carlucci [88] investigated the impact of co-solvents on biodiesel purity using refined palm oil, ethanol, and three different co-solvents (THF, acetone, and isopropanol). The study examined various operating conditions, including temperatures between 220 and 350 °C, a residence time of 3.5 min, and a co-solvent percentage of 20 % (w/w). Results showed that alkyl ester percentage increased with the addition of any co-solvent, reaching over 90 % at 8.5 min' residence time. Acetone yielded the highest percentage due to its higher polarity, enhancing mixture homogeneity. Varying co-solvent amounts and alcohol/oil ratios also influenced alkyl ester percentage. In supercritical conditions, all three co-solvents were crucial for achieving homogeneous mixtures and accelerating reaction rates. However, co-solvent type's influence diminished at temperatures exceeding 350 °C [88]. This study highlights the importance of co-solvents in enhancing biodiesel production efficiency, particularly in achieving homogeneous mixtures and accelerating reaction rates under various conditions.

To learn more about the ethyl ester conversion process, Likozar et al. [89] looked into the transesterification of triglycerides utilizing supercritical ethanol catalyzed by an ion-exchange resin catalyst (Nafion_ SAC-13) [89]. Reducing the operating conditions while keeping a high reaction rate would be possible by adding carbon dioxide as a co-solvent at a supercritical state. Therefore, a 1:3 M ratio of ethanol to carbon dioxide was used in the combination. The experiments were carried out in a fixed bed titanium reactor packed with Nafion SAC-13 (10 nm pore diameter, 0.6 ml g⁻¹ pore volume and stable to 200 °C) at temperatures between 150 and 200 °C, pressures between 15 MPa and 25 MPa, reaction times between 2 and 10 min, and molar ratios of ethanol to vegetable oil between 20 and 45. The optimum parameters are 200 °C, 20 MPa, and a molar ratio of 25 ethanol to oil for 4 min, during which time the biodiesel yield was assessed to be 80 % [59].

6. Mixing index

Recently, significant progress has been achieved in the field of miniaturizing systems to create microreactors. The synthesis of biodiesel was accomplished using these microreactors. The mixing of the reactive components, which will determine the yield and

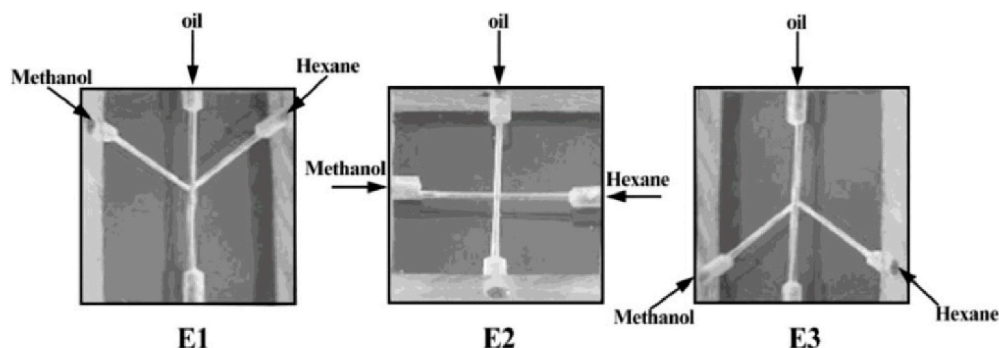


Fig. 29. Comparison of various micromixers that are in use [64].

conversion of the reaction within a short residence time, is one of the most crucial features of these microdevices [23].

An essential factor that considerably influences biodiesel yield is mixing within the microchannel. For effective mixing, the flow behavior within the microchannel should essentially be laminar. The flow is transformed from laminar to turbulent as the Reynolds number rises, causing vortices that impede the diffusion of the methanol-oil mixture [90]. Turbulent flow would have a shorter residence period, which has a substantial impact on mixing and results in reduced conversion. Because a lack of one of the reactants or an uneven distribution of the catalyst will result from poor mixing [58]. Equation (1) can be used to get the mass fraction variance using the mass fraction of oil's standard deviation in a normal cross-section of flow direction [91]:

$$\sigma = \sqrt{\frac{\sum (Y_i - \bar{Y})^2}{N}} \quad (1)$$

Where σ is the variance of the mass fraction, Y_i is the mass fraction at point i , \bar{Y} is the mean mass fraction and N is the number of sample points over the cross-section. The variation is maximum for fluids completely unmixed and minimal for perfectly mixed fluids. The parameter provided can be calculated using Equation (2) the mixing index of the mixture of vegetable oil and alcohol [92]:

$$M = 1 - \sqrt{\frac{\sigma^2}{\sigma_{\max}^2}} \quad (2)$$

Where,

M = the mixing index

σ_{\max} = the maximum variation over the range of data.

σ_{\max}^2 represents the inlet's highest variance across the data range (the mass fractions vary from 0 to 1). The mixing index ranges from zero to one (total fluid segregation to complete fluid mixing).

Using a mixing index, the effectiveness of three different micromixer types: T-micromixer, Cross-micromixer, and Double-T-micromixer was evaluated during the transesterification reaction of *Jatropha curcas* oil with ethanol to produce biodiesel. Several Reynold numbers were used to assess how the fluids mixed. With mean values of 0.99, the Cross-micromixer displayed the greatest mixing index. Whereas the Double-T-micromixer displayed the reverse trend, that is, the mixing reduced with the rise of fluid velocity, the T-micromixer's mixing index increased directly with Reynolds numbers. Significantly, the Double-T micromixer outperformed the T-micromixer at low Reynolds numbers ($Re < 50$) for fluid mixing. As the flow rate rises, advection mixing mechanisms begin to take place. Advection disturbs the flow, resulting in more species mixing. This may help to explain why, inside the T-micromixer, the mixing index rose as the Reynolds number grew. If the quality of the mixture were solely determined by diffusion between oil and ethanol then raising the velocity may lower the mixing index because higher velocity encourages a shorter time of residence inside the micromixer, which would mean that the oil molecules would cross to the other side in a shorter time of residence. Moreover, the advection phenomena enhanced the fluid mixing in microchannels [93].

From Santana Harrison S. et al. [90] findings, the breadth of the flow stream was decreased by expanding the inflow stream from $n = 1$ to 3 or 4 sub-streams. This increased contact surfaces and created many thin mixing paths which were responsible for the rise in the mixing index. The oil moving across the alcohol flow streams in the case of $n = 3$ permitted the oil molecule to diffuse in two directions rather than just one because of the decreased width of the flow stream. Moreover, this technique speeds up the mixing of the species. The T-micromixer will only achieve its maximum index for mixing (0.67) at the channel's end, while the Cross-Micromixer utilizes just a 10 mm channel length to attain its maximum index of mixing (0.99) for a Reynolds number of 10. Although the Double-T-micromixer also has a higher contact surface area, when Reynold's number is 10, the index of mixing was 0.91 compared to 0.67 for the T-micromixer, a reduced fluid mixture was seen as the velocity increased. This might be the result of less oil coming into touch with alcohol in the micromixer with the higher velocity [90].

Santana Harrison S et al. [42] found that adding a wire coil to the microchannel enhanced mixing and accelerated the attainment of equilibrium. The spirally wound microchannel, due to its curved trajectory configurations, induced centrifugal movement, maximizing the mixing index to 0.99 through advection at increased flow rates. Comparative studies on mixing patterns in T microchannels with circular and alternate circular obstacles at various Reynolds values demonstrated improved mixing indices and maximal yield. A numerical and empirical examination of transesterification in microchannels with circular obstacles showed enhanced mixing efficiency for the MCO micromixer (0.80 mixing index) compared to T-shape (0.31) and MWO (0.32) configurations. The MCO micromixer displayed a Reynolds number-independent mixing index for the T-shape, and a significant increase from 0.30 ($Re = 0.1$) to 0.40 ($Re = 100$) [42].

The improvement was attributed to phenomena like split and recombination of streams and vortex production induced by circular barriers. The MWO configuration had minimal impact on flow direction, resulting in a mixing index similar to the T-shape. Vortex formation in MCO significantly influenced fluid mixing, rising from 0.40 ($Re = 0.1$) to 0.80 ($Re = 100$). In contrast, the MWO barrier showed limited influence, maintaining a nearly identical mixing index to the T-shape [93]. The key to achieving optimal biodiesel yield and conversion lies in effective mixing within microchannels. However, increasing Reynolds numbers shift flow from laminar to turbulent, creating vortices that impede methanol-oil diffusion and reduce conversion. The introduction of equations to quantify mass fraction variance and a mixing index assesses the efficacy of the vegetable oil and alcohol mixture. Evaluation of micromixers highlights the significance of convective effects, particularly in low Reynolds microchannels. Improving mixing efficiency through the expansion of the inflow stream and the introduction of obstacles, such as the spirally wound microchannel and circular obstacles in the MCO micromixer, underscores the critical role of effective mixing in microreactors for successful biodiesel synthesis [93].

7. Heterogeneous catalysts

Due to their potential for greater sustainability than homogeneous catalysts, heterogeneous catalysts are a viable option to replace homogeneous catalysts. Nevertheless, because catalyst removal is required during the purification stage, the post-processing of homogeneous catalyst transesterification/esterification requires a significant amount of energy [94]. Economic studies showed that, in comparison to homogeneous catalysts, the use of heterogeneous catalysts produced higher biodiesel yields, high-purity glycerin as a by-product, and cheaper catalyst and maintenance costs [59]. However, the processing costs have increased overall due to the generated wastewater disposal [55]. Furthermore, the catalysts frequently diffuse into the glycerol layer, which lowers the glycerol's quality and limits the catalyst's capacity to be reused.

Hence, generally, using a heterogeneous catalyst will simplify the separation process, reduce waste water creation, and be more environmentally friendly [95]. In a microreactor, homogeneous catalyst is preferable over heterogeneous catalysts. This is because the slug flow patterns in the micromixer-microreactor system are disturbed by this catalyst, increasing mass transfer resistances [19]. A few studies are only available on the use of heterogeneous catalysts in micromixer-microreactor systems for biodiesel production. In the presence of a CaO/MgO heterogeneous catalyst, Mohadesi et al. [96] produced biodiesel from rapeseed oil using a T-shaped micromixer-connected serpentine microchannel. The biodiesel production was 99.31 % when the ideal reaction parameters of 7.875 wt% catalyst, 1.75:3 methanol to oil volume ratio, 0.575:1 n-hexane to oil volume ratio, 70 °C reaction temperature, and 10 min of residence time were used [96]. This implies that, despite the advantages of simplifying separation processes and reducing environmental impact, homogeneous catalysts are favored over heterogeneous catalysts in microreactors for biodiesel production. This preference arises from the potential disruption caused by heterogeneous catalysts to slug flow patterns in micromixer-microreactor systems, leading to increased mass transfer resistances.

Chueluecha et al. [62] utilized a packed bed microreactor with a configuration of $60 \times 1 \times 0.5$ mm packed with an activated CaO heterogeneous solid catalyst. At the optimum operating condition of: 8.9 min reaction time, 65 °C reaction temperature, and 24:1 methanol to oil molar ratio, an oil yield of 99 % was achieved [62]. The implication of the study by Chueluecha et al. [62] is that utilizing a packed bed microreactor with an activated CaO heterogeneous solid catalyst can lead to highly efficient biodiesel production. This finding contributes to the exploration of efficient and sustainable processes for biodiesel production, emphasizing the importance of reaction time, temperature, and molar ratio in optimizing performance.

However, except for packed bed types of micromixers the usage of heterogeneous catalysts is not preferred due to the blockage of the micromixer-microreactor system [97]. In other words, in micromixer-microreactor systems, the preference for using heterogeneous catalysts is limited to packed bed micromixers. This limitation suggests that while packed bed micromixers are designed to

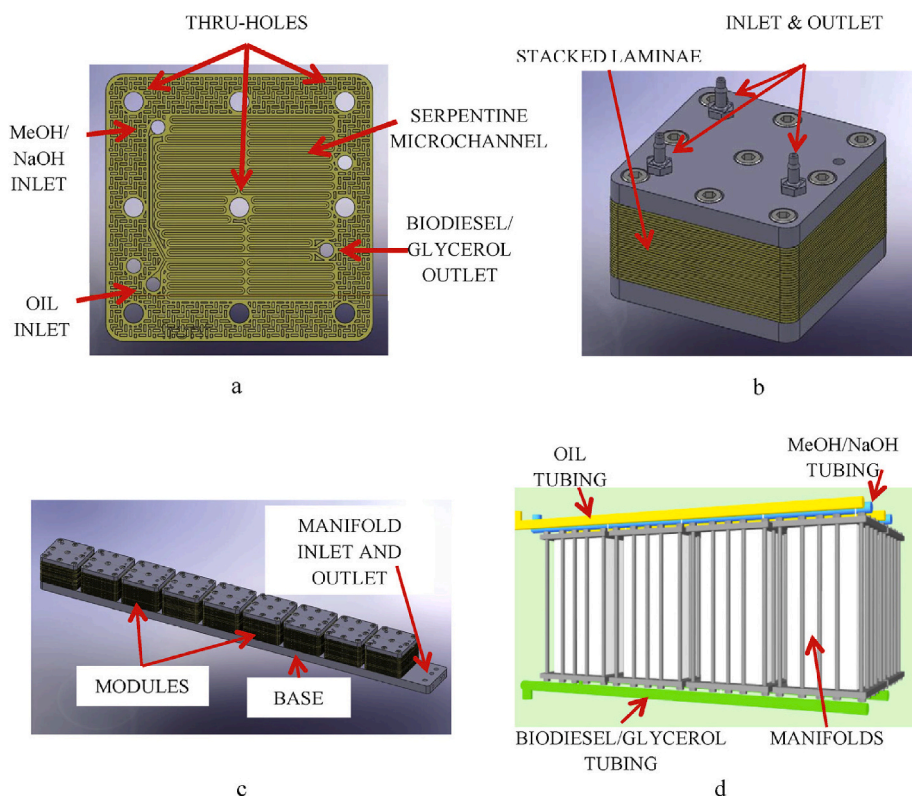


Fig. 30. Modular microreactor design: a) lamina; b) module; c) manifold; and d) full-scale microreactor [102].

effectively accommodate and utilize heterogeneous catalysts, other types of micromixers may encounter challenges. Therefore, we can say that heterogeneous catalysts are more sustainable and economical than homogeneous catalysts in biodiesel production, but there are still challenges to address. To mitigate higher energy consumption and wastewater disposal costs associated with heterogeneous catalyst processes, further research could focus on optimizing the catalyst. This optimization would enhance both selectivity and recyclability.

8. Industrial application of micromixer-microreactor systems

The micromixer-microreactor system has various benefits over the traditional biodiesel manufacturing process, including a reduced residence time, a smaller reaction volume, a smaller footprint, low operating temperatures and pressures, and lower energy costs [98]. As a result, the continual application of micro technology to the biodiesel business can be done at lower capital and operating costs. With less energy and a shorter residence time in the smaller microreactor plant volume, a higher biodiesel yield could be achieved [21, 48,99,100]. Therefore, from an economic and technical standpoint, the use of a micromixer-microreactor system may have a potential industrial application [19].

However, numerous types of research reported using microtechnology at a low flow rate, which is a significant hurdle for commercial biodiesel synthesis applications [87]. The scaling up of the individual micromixer-microreactor unit is significant in the development of these issues. The generation of commercial biodiesel, however, requires an effective scale-up technique that involves stacking several micromixer-microreactor laminae [101]. The manufacture and assembly procedures of micromixer microreactor-based biodiesel plant employment were reported. A full-scale plant was developed through numbering of individual laminae (Fig. 30a). The laminae were stacked into modules (Fig. 30b) and the modules were stacked into manifolds (Fig. 30c) and finally assembled into a full-scale micromixer-microreactor system (Fig. 30d) biodiesel production capacity 1.2×10^6 L of biodiesel per annum. To produce this amount of biodiesel 14000 laminae were joined together to make 35 individual manifolds which have several inlets and outlets. Each manifold had 8 modules. Each module was made of 50 micromixer-microreactor system [102]. Hence it can be concluded that the industrial use of micromixer-microreactor systems in biodiesel production offers significant advantages, including reduced residence time, smaller reaction volume, compact footprint, and lower energy costs. This leads to potential cost savings in capital and operations. Despite challenges at low flow rates, effective scale-up of individual units is crucial for commercial biodiesel synthesis. The development of a commercial plant involves stacking multiple micromixer-microreactor laminae, enabling large-scale biodiesel production.

New studies on scale-up techniques have revealed interesting ways to increase production rates using bigger micromixers. These methods include scale-out techniques using appropriate dimension expansion, which assert that channel dimensions can be increased to a size that is adequate to achieve the required performance. On the other hand, the increase in channel length affects species dispersion and mixing efficiency, which in turn results in a decrease in fluid mixing and chemical reaction efficiency [67]. Information regarding reaction kinetics behavior and fluid dynamics appears to be helpful patterns for the optimization of the development and application of micromixer-microreactor in industrial processes [101]. Micromixers-microreactors implement the numbering-up technique to increase reaction output without having to increase the size of the reactor. The numbering-up technique for micromixer-microreactors involves running a cluster of several reactors simultaneously inside a single module. This characteristic makes it possible to apply all of the optimized parameters of a single micromixer regardless of scaling up the output scale, including reaction temperature, reaction duration, and flow rate. Optimizing heat and mass transport of chemical reactants is the main goal of intensifying reaction yield in microchannel reactors [34]. This is because the reaction yield is influenced by effective mass transfer caused by improved molecular mobility between chemical reactants.

Once every possibility for chemical intensification and smart scale out to the mesoscale has been explored, the process known as "numbering-up" can be used to enhance capacity even more. In contrast to conventional reactors, where scaling-up happens with an increase in each specific reactor unit size, this indicates an increase in the number of structural units. The two stages of the numbering-up idea are capacity selection and route scaling, as elaborated by Gaikwad & Gogate [103]. The flow rate per channel can be used to identify the three ranges: 10, 100, and 1000 L/h. There are three main steps in the scaling route. First, one channel is enhanced (dimensions are increased while maintaining the heat and mass transfer characteristics of the micromixer-microreactors); second, an internal numbering-up process creates a modular unit by joining several channels (mixer, reaction channels, heat exchanger, and separator) into one unit; and third, an external numbering-up process arranges up to five or six modular units [103]. There are significant disadvantages to both of these methods: external numbering-up requires more space and is more expensive than internal numbering-up, which is restricted by the number of units on a single chip and results in a pressure drop that grows as the number of units increases [59].

For the continuous manufacture of chemicals, a continuous-flow micromixer-microreactor system is an interesting option [104–109]. Compared to traditional batch or semi-batch reactors, this system provides exact control over the reaction parameters, rapid heat and mass transfer, and inherent safety [110–113]. Even while reaction yield and efficiency have improved on a small scale, the industry has only successfully adopted a small number of these methods. The significant difference in size between laboratory and production-site micromixer-microreactor systems is a primary cause of the current findings. On a production scale, a throughput of kg/h or more is necessary, although a lab microreactor typically has a throughput in the range of g/h or g/min [111,114]. As a result, a scale factor of 100–1000 is typical, which is difficult to achieve without losing the benefits that the micromixer-microreactor combination offers naturally. The micromixer-microreactor system's narrow channel size, which results in short transfer distances and a high surface-to-volume ratio, is the primary source of most of its benefits. Nevertheless, these micro-channel properties would stop right away when the channel's dimensions increased during scale-up. Therefore, it was determined that the numbering up technique

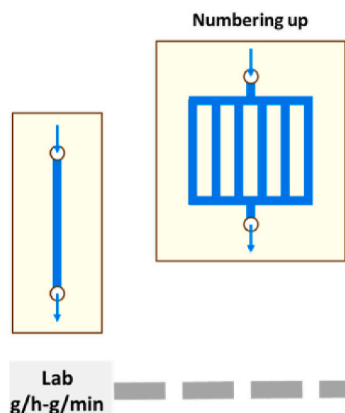


Fig. 31. Scale-up approaches of microreactors from lab scale (a single channel reactor) to production scale [18].

(Fig. 31) was the best approach to action for scaling up the micromixer and microreactor system for the manufacture of biodiesel. This technique allows for the design of compact micro-plants and the achievement of higher capacity by linking micromixers and microreactors with the same verified dimensions to function simultaneously.

The main advantage of this approach is that it maintains the microenvironment's hydrodynamics and transfer properties, enabling reactions to be carried out under settings that are exact duplicates of the microreactor. As a result, to meet the throughput requirements of the industry, hundreds or even thousands of micromixer and microreactor system units may be needed [18]. For example, a single micromixer-microreactor system can produce 50 tonnes of biodiesel per year, while a batch-wise biodiesel factory can produce 50,000 tonnes annually. Thus, 1000 micromixer-microreactor units would be needed to produce 50,000 tonnes of biodiesel annually, or the same amount as a batch biodiesel plant (i.e., 50,000 tons/year divided by 50 tons/year). In general, a micromixer-microreactor system's volume can be determined using Equation (3) [18].

$$V = \frac{\pi}{4} N L D^2 \quad (3)$$

where N is the number of channels connected in parallel, L is the length of the channel and D is the hydrodynamic channel diameter.

Fluid distribution, which is necessary for providing accurate and consistent flow in each reactor unit and, consequently, optimal and consistent performance in each reactor, is still a major difficulty. Therefore, some of the drawbacks of the numbering up technique include the need for sophisticated flow distribution management, the need for highly technological setups, and sensitivity to the distributor's design and construction. To reduce the fluid distribution limitation, Dong et al. [18] designed two types of fluid distributors: internal and exterior micromixer-microreactor system fluid distributors. The reactant flow was allocated for the external fluid distributor before the reactants entered the parallel microreactors (Fig. 32a). Conversely, concerning the internal flow distributor, the distributing unit (distributing channels) and the parallel micromixer-microreactor system were put together simultaneously and

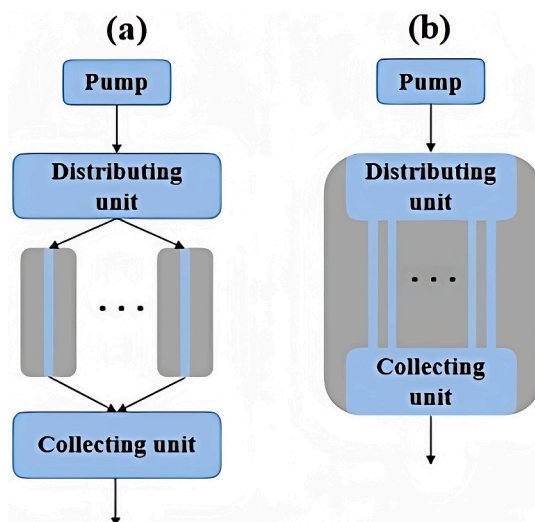


Fig. 32. Micromixer-microreactor system flow distributors. (a) External flow distributor. (b) Internal flow distributor [18].

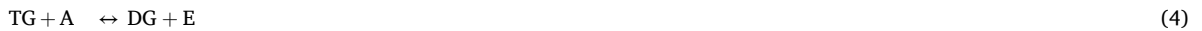
housed in the same frame (Fig. 32b) [18].

Furthermore, to determine the factors that result in a better transfer of experimental flow chemistry results into the pilot-scale processing under specific environmental and financial conditions, Budzaki et al. [115] carried out three case studies: one on catalyst reuse options and two on process concepts for intensified processing of epoxidation and transesterification of natural feedstocks. The findings demonstrated that micromixer-microreactors outperformed conventional industrial fed-batch systems. In their review article, Budzaki et al. [115] concluded that the most promising industrial-scale biodiesel production technologies are the packed-bed reactors (PBR), which precede the continuous-stirred tank reactors [115]. They made the point that when using PBR, the residence time of the reaction mixture in the reactor and its interaction with the immobilized enzymes' active sites is crucial. Low flow rates can be used to regulate that, which will lengthen the oil's residence period and increase its conversion to biodiesel.

9. Reaction kinetics of micromixers

The expense of the micromixers and the search for substitutes for used catalysts are two issues that frequently impede the advancement of microreactor technology. A catalytic micromixer-microreactor needs to be precisely designed by producing both the catalyst and the reactor at the same time, while also accounting for basic reaction kinetics and transport issues [116]. To scale up a micromixer-microreactor system for use in industrial processes, reaction kinetics dynamics are crucial [117–119]. The transesterification of oil and alcohol may be impacted by elements such as the types of feedstock oil, alcohol, and catalyst, the extent of temperature, residence duration, alcohol-to-oil molar ratio, and catalyst concentration even though kinetics is independent of the types of reactor used [119].

According to equations (4)–(6), the transesterification reaction passes through three sequential reversible reactions. In the presence of a basic catalyst, triglyceride (TG) first reacts with alcohol (A) to produce di-glyceride (DG) and ester (E), followed by a reaction between di-glyceride (DG) to make mono-glyceride (MG) and ester, and finally a reaction between mono-glyceride and alcohol to produce ester and glycerol (GL) [120].



The overall rate of transesterification reaction is represented by equation (7) [121]:

$$-r_{\text{TG}} = k_1 C_{\text{TG}}^\alpha C_{\text{A}}^\beta + k_2 C_{\text{E}}^\delta + C_{\text{GL}}^\gamma \quad (7)$$

where, $-r_{\text{TG}}$ = reaction rate of TG (mol/(m³ × s))

$\alpha, \beta, \delta \& \gamma$ = the order of the reaction to TG, A, E, and GL respectively.

k_1 = forward reaction rate constant, k_2 = reverse reaction rate constant

C_{TG} = concentration of TG, C_{A} = concentration of A, C_{E} = concentration of E, C_{GL} = concentration of GL

Using ANSYS CFX 14.0, Santana et al. [90] assessed the overall reaction rate constant that had taken place in a T-shaped micromixer coupled microchannel (w = 1500 mm, h = 200 mm height, and L = 411 mm of longitudinal length) reactor. The results are shown in Table 4 [90].

Han et al. [117] used ANSYS and Matlab to analyze kinetic and transport equations using a two-dimensional reaction model. It was assessed how residence duration affected the biodiesel yield. A significant effect on the yield was caused by the highest yield being attained at a slightly longer residence duration. However, the straightforward kinetic model they used prevented them from doing a numerical investigation of the process [117]. The kinetics of the transesterification reaction carried out with a T-shaped micromixer coupled microchannel delay loop were examined numerically by Richard et al. [122] as depicted in Fig. 33. The concentration of ethyl ester (EE) increased and the concentration of triglyceride (TG) declined as the residence time in the micromixer-microreactor system increased until equilibrium was reached [122].

GITT (Generalized Integral Transform Technique) was used to investigate the transesterification of soybean oil in a T-shaped micromixer-coupled microchannel at various residence times and reaction temperatures. Convective and diffusive mass transfer, as well as reaction kinetics, was modeled in three dimensions. The process employed was second-order reversible transesterification.

Table 4
Evaluated kinetic data at various reaction conditions.

Temperature (°C)	Ethanol/oil molar ratio	NaOH concentration (wt.%)	$\vec{k} * 10^6$ (m ³ mol ⁻¹ s ⁻¹)	$\tilde{k} * 10^8$ (m ³ mol ⁻¹ s ⁻¹)
25	9:01	0.75	1.52	3.15
50	6:01	0.75	8.1	6.61
50	9:01	0.75	8.3	6.07
50	9:01	1	15.68	9.42
50	9:01	1.25	19.38	11.72
50	12:01	0.75	8.73	30.13
75	9:01	0.75	2.37	9.53

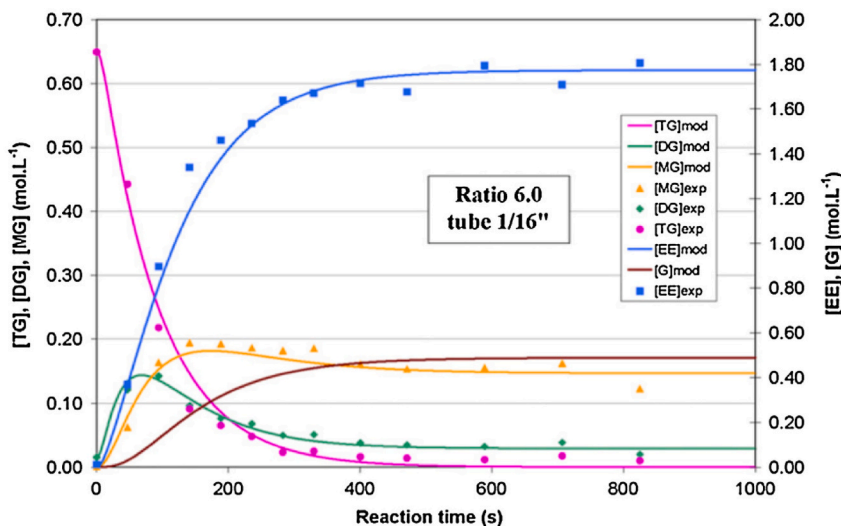


Fig. 33. Time variation of species concentration in biodiesel synthesis [122].

Because there was enough time for mass transfer by diffusion over the prolonged residence time, a greater yield was produced. The rate of the reaction was shown to be influenced by temperature [123]. To scale up a micromixer-micro reactor system for industrial biodiesel synthesis, careful consideration of reaction kinetics is essential. Factors including feedstock type, temperature, residence duration, and catalyst concentration play a crucial role in transesterification. Numerical simulations, such as those conducted by Santana and Han, assess reaction rate constants and kinetics, highlighting the significance of residence duration in optimizing biodiesel production. Richard’s study focuses on transesterification kinetics in a microchannel, revealing concentration variations over time. GITT analysis of soybean oil transesterification underscores the impact of temperature on reaction rates. Overall, these findings underscore the necessity of understanding kinetics for efficient and scalable micromixer-micro reactor biodiesel production.

10. Economic aspects of micromixer-microreactors systems in biodiesel synthesis

Biodiesel is mainly produced by two types of reactors: batch and continuous-flow reactors. Biodiesel produced in batch processes has drawbacks such as the requirement of a high amount of alcohol to move the transesterification reaction forward, separation cost of fatty ester products from the reaction mixture, more water consumption for purification, and high reactor cost [30]. Continuous biodiesel production using different feedstocks and processing techniques overcame such drawbacks [19].

The economic analysis of enzymatic biodiesel production from refined sunflower using immobilized lipase with a capacity of 10,000 tons per annum employing a packed-bed microreactor was done by Budžaki et al. [115]. To determine key bottlenecks, that are causing the negative balance, share of each parameter in total production was plotted and presented in Fig. 34. The figure shows depreciation (immobilization) as the second highest cost share in total production costs (38.5 %). The effects of activity retention (Fig. 35) and utilization of the oxirane groups on Eupergit CM (Fig. 36.) on the total profit were examined. The utilization of the oxirane groups on Eupergit CM shows a higher increase in the total plant profit than an improvement in activity retention. Besides, the analysis of SEPABEADS EC-EP/M application in TLL (Thermomyces lanuginosus lipase) immobilization on the total production costs was conducted. In addition to this, the effects of activity retention and utilization of the oxirane groups on SEPABEADS ECEP/M on

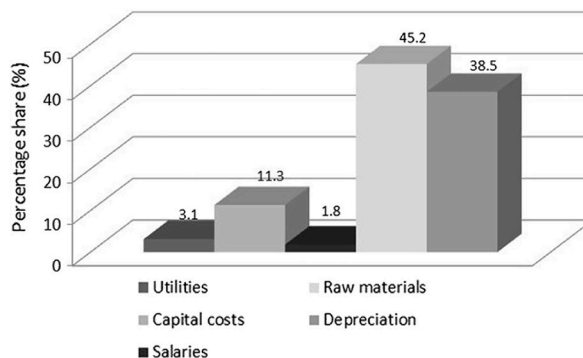


Fig. 34. Share of individual input parameters in the total production costs [115].

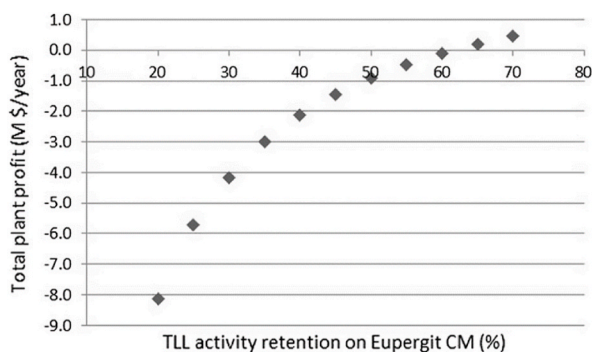


Fig. 35. Influence of the increase in activity retention on the total plant profit with Eupergit CM [115].

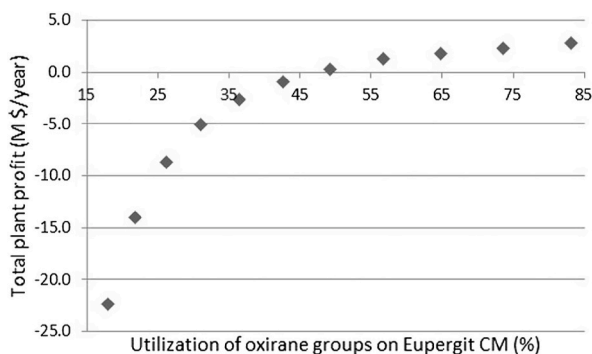


Fig. 36. Influence of the increase in utilization of the oxirane groups on the total plant profit with Eupergit CM [115].

total plant profit were examined. The results showed the application of TLL immobilized on SEPABEADS EC-EP/M on an industrial scale has a potentially profitable outcome due to a significant reduction in operating costs. Nevertheless, by recognizing and solving key process bottlenecks, the biodiesel synthesis in micro-flow packed-bed reactors can have a promising future at industrial scale production [115].

A few cases of enzymatic biodiesel production in packed-bed microreactors were compared against a real-time, industrial plant in Croatia operating at 10,000 t/year, which uses rapeseed oil to produce biodiesel in batch mode. For the industrial rapeseed biodiesel plant, the raw material was the biggest contributor to the production costs (71 %), followed by the capital costs (11 %). Total plant profit for the first year of production for case 2&5 were much better than the batch operating mode. In Case 2, the number of reuses of

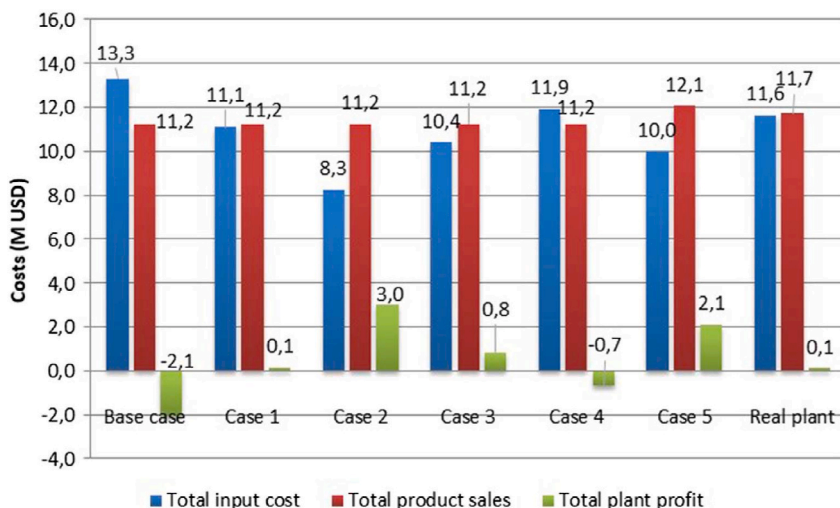


Fig. 37. Total plant profit for the first year of production for each case [124].

the immobilized TLL were 50 days. Case 2 showed a total plant profit of 3 M USD (Fig. 37). The immobilization cost and share in total input costs was 9.9 %, while the raw materials contributed to 67.2 % of total input costs. The cash flow analysis showed a positive economic balance after the second year of production, with an NPV of 0.05 M USD. The cost analysis showed that Case 2 can be a viable enzymatic biodiesel production process (Fig. 38). The influence of increased utilization of oxirane groups on SEPABEADS EC-EP/M on total plant profit was investigated in Case 5. A total plant profit of 2 M USD was achieved at 83.1 % utilization of oxirane groups. The raw materials contributed to 59.5 % of the total input costs, followed by the immobilization at 20.4 %. The cash flow analysis for Case 5 showed a positive NPV after three years of production [124].

An economic analysis over a 15-year design life proved that the production of biodiesel in a micromixer-microreactor system is a more feasible alternative to the production of biodiesel in a batch reactor. Both reactors were compared on an equal basis of a 900 kg/year production rate of biodiesel. According to the study, while energy consumption for the micromixer-microreactor system and batch reactor were 55 and 130J/g of biodiesel, the capital cost was \$2000 and \$615 respectively. On the other hand, the operating cost of the micromixer-microreactor system and batch reactor was \$170/year and \$ 416/year respectively. Although microreactors have a larger capital cost than batch reactors, the operating cost of microreactors is significantly lower than the operating cost of batch reactors, making it a more feasible alternative in the long run for microreactor biodiesel production [125].

11. Environmental impact and benefit of micromixers

When producing biodiesel, the use of micromixers and microreactors presents a significant environmental advantage over batch processing techniques. In batch production methods, vegetable or animal fat is reacted in a batch reactor with a catalyst [126]. Usually, the reaction is conducted under high pressure and temperature conditions. The biodiesel is separated from the glycerin and other by-products after the reaction is finished [127]. In continuous microreactor systems, vegetable or animal fat is reacted in a microreactor with a catalyst. Usually, the reaction is conducted at lower pressures and temperatures than in batch production techniques. Continuous production of biodiesel involves separating it from glycerin and other by products. Depending on the process employed, the production of biodiesel may have a considerable negative impact on the environment. Due to their higher energy requirements, batch production techniques usually have a greater environmental impact than continuous microreactor systems. This is because reactants are usually heated to high temperatures and pressures during batch production methods. Conversely, continuous microreactor systems can function at reduced pressures and temperatures [128].

In comparison to continuous microreactor systems, batch production methods also use more water. This is because batch production techniques usually require cleaning the biodiesel to get rid of contaminants. Conversely, biodiesel with less water content can be produced by continuous microreactor systems [31,68]. Continuous microreactor systems produce less waste than batch production methods. This is because techniques for batch production generally yield higher levels of glycerin and other by-products. Conversely, biodiesel with less glycerin can be produced using continuous microreactor systems [126]. Micromixers encourage complete and quick mixing of reactants, which results in a more complete feedstock to biodiesel conversion. This reduces waste generation and improves reaction efficiency by minimizing waste streams that contain partially converted products or unreacted feedstock [37,129]. Processing times are greatly shortened by reaction rates that are accelerated by efficient mixing in micromixers. In contrast to batch processes, this results in reduced energy usage for heating and agitation [34]. Microreactors' high surface area-to-volume ratio promotes effective heat transfer, which may allow for lower reaction temperatures or shorter heating times [130]. In comparison to batch reactors, microreactors also require smaller volumes of reaction media. As a result, less solvent waste is produced throughout the procedure [131]. Continuous processing is made possible by microfluidic systems, which also provide real-time reaction parameter monitoring and control. By doing this, the possibility of uncontrolled exothermic reactions and other batch process-related safety risks is reduced.

Compared to batch processes, continuous processing with smaller reaction volumes may result in lower fugitive emissions of

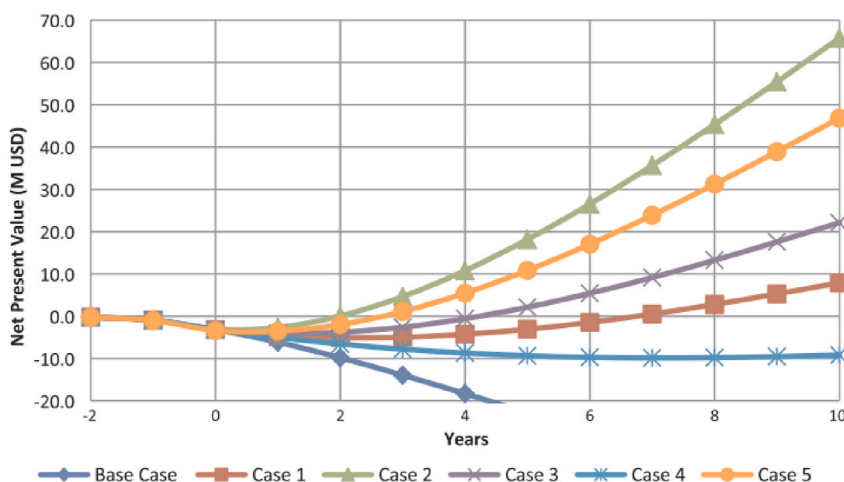


Fig. 38. Cash flow analysis for each case for a 10-year production period [124].

volatile components like methanol [31,132]. Thus, the use of microreactors and micromixers offers a viable method for producing biodiesel that is not harmful to the environment. They are an important tool for a more sustainable biofuel industry because of their capacity to encourage effective mixing, minimize waste generation, and lower energy consumption [133]. Therefore, batch production methods are less environmentally friendly than continuous microreactors with micromixer systems when producing biodiesel. This is because continuous microreactor systems produce less waste, water, and energy.

12. Future perspectives and challenges on micromixers

Finding building materials for microfluidic devices that can effectively handle corrosive and reactive reactants and products remains a significant challenge. Polymers are mainly used to make the majority of microreactors due to their ease of fabrication. However, microreactors made of polymers are susceptible to chemical attacks, which means they may react with reactants, by-products, and/or products, reducing their shelf life. Another challenge for plastic polymer microdevices is avoiding the risk of leakage within the joints between each microreactor bundle during biodiesel production in microreactors and micromixers [102]. As a result, using gaskets and fasteners to prevent reactant and product leakage in microreactors and micro mixers is not a viable solution and necessitates additional research.

The clogging of micromixers and microreactor channels caused by the viscosity of oil and biodiesel poses a severe obstacle to the generation of biodiesel in microdevices. When oil with a high free fatty acid level is used for biodiesel production in a microreactor, saponification is another cause of microchannel clogging. Cleaning the fouling of microchannels with descalers and chemicals is extremely challenging due to their microscopic size. As a result, research must be done to reduce the risk of fouling, and cleaning solutions must be devised.

Research has not yet adequately analyzed the use of solid base and acid heterogeneous catalysts in microreactors due to their practical limitations. The immiscible fluids and catalyst mass transfer restrictions affect the reaction rate. So, it is essential to carry out additional research in this area. Even if a packed bed microreactor composed of solid acid or base heterogeneous catalyst as a random packing has been employed and generates a fair yield, it is still challenging to use packed beds constantly on a larger scale. This is because their catalytic activity is reduced continuously as a result of loss of strength of basicity or acidity and blockage of active pores or sites by oil, glycerol, and biodiesel. Another problem that needs to be resolved is the production of a large pressure drop when packed into the micromixer-microreactor pressure building to a significant level [49,102].

Because the size of the microreactor is on a micro-scale it is quite challenging to get at different residence times the reaction and flow behavior data inside the microreactor. This makes the validation of experiments less practical. Therefore, the use of microreactors on an industrial level faces practical limitations. Hence, more research on the methods to upgrade lab-scale microreactor biodiesel production to industrial-scale production should be done. Also, it is uncertain how successfully the same process will reproduce itself in large-scale manufacturing given how difficult it is to control the flow behavior of each bundle of microreactor online. Hence, an intelligent automated monitoring system is required to accurately regulate their operation [58]. In conclusion, solving these issues has the potential to transform the industrial production of biodiesel while simultaneously advancing the technological capabilities of micromixers and microreactors. The efficient and sustainable production of biodiesel can be achieved by innovative solutions to material selection, leakage prevention, fouling reduction, catalyst optimization, reactor scaling, and data validation. Furthermore, the incorporation of intelligent automated monitoring systems can guarantee accurate control over the functioning of microreactors, hence augmenting their feasibility in extensive industrial environments.

The goal of ongoing research is to improve the performance of micromixers by investigating new designs, materials, and fabrication methods. New geometries are explored in micromixer design research. It refines micromixer designs for more scalable integration and improved mixing efficiency. In addition, it investigates materials that are highly resistant to chemicals and corrosion to be both affordable and biocompatible. In the future, micromixers may be combined with nanotechnology, microfluidics, and microelectronics for use in biodiesel production. There are still difficulties, though, mainly with increasing production, guaranteeing dependability, and lowering fabrication costs. To fully realize the potential of micromixers and enable their wider adoption in academic research and real-world applications, these issues must be resolved.

13. Conclusion

In summary, the challenges posed by the higher reaction time and product costs in biodiesel synthesis, coupled with the immiscibility of oil and alcohol, necessitate an intensification approach through micromixers. This method enhances conventional mixing principles, crucial for achieving target outcomes in microreactors during transesterification reactions. The comprehensive review undertaken in this study covers various micromixer types, their mixing efficiency, the role of co-solvents in intensification, and the hydrodynamics contributing to conversion enhancement. Analyzing advancements in micromixer technology across different systems reveals the profound impact of micromixer and microreactor shape and microsize level on biodiesel synthesis yield. Particularly, research outcomes are particularly effective at the smaller (lab-scale) biodiesel synthesis level. The widespread utilization of homogeneous base catalysts is attributed to their higher catalytic activity and positive influence on macromixing efficiency.

Conversely, reports indicate challenges with heterogeneous catalysts in microchannel-packed bed micro mixers, specifically hindering alcohol and oil macromixing.

Looking ahead, for effective industrial applications, several crucial aspects demand further research. These include the development of microheating and cooling systems for microreactors, ensuring the maintenance of efficient flow regimes like slug and droplet flows. Additionally, investigating high-free fatty acid oil transesterification employing an acid catalyst is vital. The enhancement of

methods for joining laminae to modules and the production of efficient, affordable gaskets for microreactors are also areas requiring focused exploration. While micromixer-microreactor systems showcase efficiency in lab-scale biodiesel production, addressing these future research points is pivotal for their successful industrial application. This includes advancing heating and cooling technologies, optimizing flow regimes, exploring acid-catalyzed conversion of high-free fatty acid oils, improving modular design methods, and developing cost-effective gaskets. The pursuit of these research avenues will undoubtedly contribute to the ongoing evolution and effectiveness of biodiesel production through micromixer and microreactor systems.

Data availability statement

No data was used for the research described in the article.

CRediT authorship contribution statement

Yonas Desta Bizualem: Writing – review & editing, Writing – original draft, Visualization, Resources. **Amare Gashu Nurie:** Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Talbachew Tadesse Nadew:** Writing – review & editing, Visualization, Supervision, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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