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Cold plasma as an emerging catalytic route for oil modification

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ABSTRACT

The effect of cold plasma on the oxidation, hydrogenation, transesterification, and pyrolysis reactions is investigated. Also, the effect of cold plasma parameters on these reactions and the advantages and challenges of cold plasma are investigated. Cold plasma can produce low-trans partial hydrogenated oil at low temperature without catalyst. Besides, oil modified through transesterification and pyrolysis processes using cold plasma technique can be used for biofuel production. Oxidation during cold plasma treatment can be inhibited by omitting the oxygen from carrier gas and applying the lowest possible input power and treatment time. One of the main challenges of using dielectric barrier discharge device at large scale is providing high plasma intensity for large amounts of raw materials. In microwave discharge plasma device, high capital investment is the main challenge for scaling up. In conclusion, cold plasma technique can hydrogenate and transesterify oils at low temperature.

1. Introduction

In recent years, the plasma technique has been introduced as an emerging technology for microbial decontamination and altering the physicochemical properties of food components (Nwabor et al., 2022). Based on the Scopus search results, from 2014 to 2024, the number of cold plasma technique studies has enhanced by 43.49 % (Fig. 1). Plasma is an ionized gas that contains various reactive components such as free radicals, electrons, gas atoms, positive and negative ions, photons, and ground or excited state of molecules. The excitation frequency as well as pulsed, sinusoidal, and continuous wave patterns can affect the plasma characteristics (Dimitrakellis, Delikonstantis, Stefanidis, & Vlachos, 2022).

Various power sources such as direct current, microwave frequency, radiofrequency, and alternating current have been applied for plasma production. Different gases like argon, nitrogen, helium, and oxygen can be applied to produce plasma. Plasma can produce under vacuum or atmospheric pressure (Khani, Shokri, & Khajeh, 2017). The mechanism of plasma generation is providing large amounts of electrical energy to a gas at given pressure and temperature. This electrical energy excite and ionize the gas and generates electrons. When plasma is ignited, collisions between electrons results in various chemical reactions. These chemical reactions along with subsequent secondary chemical reactions between neutral particles and ions can generate various reactive species (Dimitrakellis et al., 2022). These reactive species include negative and positive ions, reactive nitrogen species such as NO_2, N_2O, and NO, charged particles, ultraviolet radiation, and reactive oxygen species such as O_3, OH, and O_2 (Fu et al., 2022).

Plasma technique can be classified into two distinct categories: temperature-based plasma technique and pressure-based plasma technique. Temperature-based plasma technique can be classified into low temperature plasma, high temperature plasma, and warm plasma, based on plasma production mechanism and the relative temperature between ions, electrons, and neutral species. In high temperature plasma technique, gas is heated at high temperatures (4000–20,000 K) to be converted to ionized form. In this technique, the plasma is highly ionized and the temperature of all chemical species (free radicals, electrons, gas atoms, positive and negative ions, and ground or excited state of molecules) is the same. Therefore, in high temperature plasma technique, there is a thermodynamic temperature equilibrium between all chemical species. In cold plasma technique, the gas temperature is near room temperature while the temperature of electrons is high (~10,000 K). In cold plasma technique, the plasma is weakly ionized and the temperature of all chemical species is not the same. Thus, in cold plasma technique, there is no thermodynamic temperature equilibrium between all chemical species. Cold plasma is generated by electric or electromagnetic fields. In warm plasma technique, the gas is heated at temperatures in the range of 2000–3000 K. In this technique, the ionization degree is high. Also, in warm plasma technique, the electrons energies are not the same (Li et al., 2020).

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In food processing applications, the cold plasma technique is commonly used. Pressure-based cold plasma technique can be divided into low-pressure cold plasma (10–100 Pa), atmospheric-pressure cold plasma (10^4–10^5 Pa), and high-pressure cold plasma (>10^5 Pa). In recent years, the atmospheric-pressure cold plasma technique has gained increased attention due to its low cost, easy operation, and the potential for continuous operation (Akhtar, Abrha, Teklehaimanot, & Gebrekirstos, 2022; Mudassir et al., 2023).

Reactive components present in cold plasma (free radicals, ions, and photons) can interact with the triacylglycerols of liquid oils. The interaction between cold plasma reactive components and triacylglycerols can result in different types of reactions such as oxidation, hydrogenation, transesterification, and pyrolysis (Palm et al., 2022).

Lipid oxidation is an undesirable reaction in liquid oils which can alter the nutritional quality and sensory characteristics of liquid oils. The peroxidation process is a spontaneous chain reaction that includes three stages of initiation, propagation, and termination. Cold plasma can generate free radicals. These free radicals can initiate the peroxidation process (Na, Mok, & Lee, 2020). Hydrogenation is an important manufacturing process in the food industry. During the hydrogenation process, liquid oil is converted to solid fat (Yepez & Keener, 2016). Transesterification is a conventional process for producing biodiesel. In this reaction, liquid oil triacylglycerols react with alcohols to generate fatty acid alkyl ester and glycerol (Palm et al., 2022).

Applying cold plasma technique for performing hydrogenation and transesterification reactions of liquid oils is more useful than conventional methods because cold plasma technique can hydrogenate at ambient temperature and atmospheric pressure. In addition, trans-fatty acids are not formed during the hydrogenation of liquid oil by cold plasma technique (Yepez & Keener, 2016).

Different parameters such as plasma source, processing time, input power, gas constituents, distance between the discharge and the sample, and gas flow rate can affect these interactions (Mir, Shah, & Mir, 2016).

The impact of cold plasma technique on lipid oxidation has been reported in some reviews (Gavahian, Chu, Khaneghah, Barba, & Misra, 2018; Jadhav & Annapure, 2021; Kopuk, Gunes, & Palabiyik, 2022; Rao et al., 2023; Saremnezhad, Soltani, Faraji, & Hayaloglu, 2021). However, the impact of the cold plasma technique on the hydrogenation, transesterification, and pyrolysis reactions of oils has been less investigated.

This review aims to describe and discuss the advantages and challenges of using the cold plasma technique for hydrogenating, transesterifying, and pyrolysis of liquid oils compared to the conventional methods for hydrogenation, transesterification, and pyrolysis reactions. Another aim was to investigate the effect of different cold plasma parameters such as gas type and concentration, gas flow rate, input power, electrode gap distance, and reaction temperature on these reactions. Besides, the limitations of using cold plasma technique for hydrogenation, transesterification, and pyrolysis reactions of liquid oil at industrial scale and the most important paths to the future are described.

2. Plasma technique

2.1. Basic principles

Plasma, the fourth state of matter is an ionized or partially ionized gas that contains free radicals, electrons, photons, ions, visible light, and excited or neutral state of atoms and molecules (Nikmaram & Keener, 2022). Plasma can be produced by applying energy in the forms of electrical, mechanical, thermal, nuclear, or electromagnetic waves (e.g., radio waves and microwaves) to a neutral gas or a combination of gases. Plasma production can be done under atmospheric pressure or under vacuum (Gholamazad, Hosseini, Hosseini, Ramezan, & Rahmanabadi, 2022). Gases which are commonly used for plasma production include oxygen, atmospheric air, nitrogen, helium, and argon. These gases can be used alone or in combination with each other (Mao et al., 2021). Cold plasma can be produced under atmospheric pressure or under vacuum conditions. At atmospheric pressure conditions, the reactive species are produced through collisions between electrons and heavier particles (Surowsky, Schlüter, & Knorr, 2015). By accelerating electrons in an electric field, several collisions occur between the atom, molecules, and electrons in the gas. The reactive species will be generated through primary and secondary processes. These processes include electronic impact processes (ionization, vibration, attachment, dissociation, and excitation), ion–molecule reactions, ion-ion neutralization, quenching, Penning ionization, three-body neutral recombination, photoemission, photo-ionization, and photo-absorption (Misra, Schlüter, & Cullen, 2016). Collisions among electrons, nitrogen, and oxygen molecules result in the generation of reactive nitrogen species ($^{\bullet}NO_3$, $^{\bullet}NO$, N_2O_5, NO_2^-, N_2O^-, ONOOH/ $ONOO^-$, and NO_3^-) and reactive oxygen species ($^1O_2$, atomic O, $O_2^{\bullet-}$, $^{\bullet}OH$, O3, $^{\bullet}OOH$, and H_2O_2). When oxygen is present, hydroxyl radical ($^{\bullet}OH$) is one of the major reactive species generated through primary and secondary collision processes in the plasma. The hydroxyl radical is usually observed in discharges containing humid air. This reactive species is an oxidizing compound which can abstract a hydrogen atom from organic compounds (RH) and produce other radicals (e.g. H_3O^+ and H_2O_2). In the case of reactive nitrogen species, $^{\bullet}NO$ plays an important role in generating other reactive nitrogen species

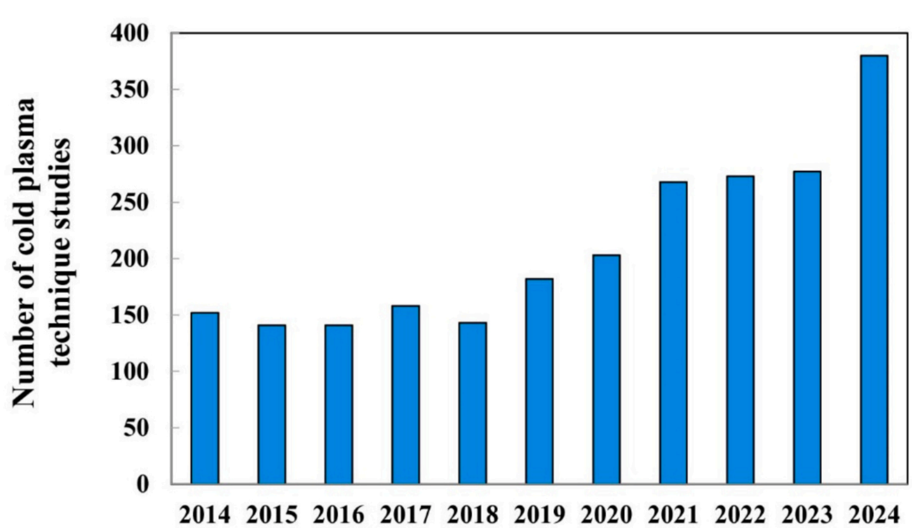

Fig. 1. Publications on cold plasma technique during past 10 years. The source of information was Scopus database.

(Whitehead, 2016).

2.2. Cold plasma devices

Different cold plasma apparatus includes plasma jet (PJ), dielectric barrier discharge (DBD), corona discharge (CD), radiofrequency discharge (RFD), microwave discharge (MD), gliding arc discharge (GAD), and glow discharge (GD) (Kaavya et al., 2023). Among these apparatus, DBD device has been applied in most research. This can be due to the low cost, better availability, safety, environmental friendliness, and convenience of DBD device. Plasma produces between DBD electrodes (Rout & Srivastav, 2023; Ying et al., 2024). The advantages and disadvantages of different types of cold plasma devices are presented in Table 1.

2.2.1. DBD

One of the most common devices for plasma production is the DBD device. In DBD device, a direct voltage is applied across two electrodes to ionize the gas and generate plasma. It has a high voltage electrode at the top and a ground electrode at the bottom. The plasma region where the microfilaments generate a thin layer is between the two electrodes. One or both electrodes are covered with a dielectric barrier (Laroque, Seó, Valencia, Laurindo, & Carciofi, 2022). Glass, mica, ceramic materials, and silica glass can be used as dielectric barrier. The role of dielectric barrier is preventing an arc discharge and restricting the flow of current. In addition, the DBD device allows the gas to be ionized inside the inter-electrode space (Akhtar et al., 2022). Coaxial cylinder type and plate type are the most common configuration of electrodes in DBD plasma device (Wongjaikham, Wongsawaeng, Ngaosuwan, Kiatkittipong, & Assabumrungrat, 2023).

Although cold plasma is produced quickly by the DBD method, the cold plasma temperature and the reactive species concentration are low (Feizollahi, Misra, & Roopesh, 2021). To enhance the concentration of reactive species, a DBD plasma with needle-in-tube has been designed. In this device, a needle electrode is inserted into a hollow dielectric tube. This needle-in-tube device can force the generated free radicals to travel through the reactant layer before leaving the reaction chamber. This maximizes the physical contact with the reactant (Wongjaikham et al., 2023).

In DBD device, plasma can be generated at both low pressure and atmospheric pressure conditions. DBD device can operate at an alternating current voltage with 1–100 kV amplitude and a frequency of a few Hz to MHz for discharging (Kopuk et al., 2022). Also, in DBD devices, high-frequency operation is necessary for enhancing plasma quality and stability. A step-up transformer can be used for generating a high frequency electric field and obtaining a high frequency, high-voltage output for plasma generator. A transformer can reduce the switching losses and electromagnetic interference via applying voltage switching techniques (Yong-Nong & Chih-Ming, 2013).

Direct and indirect modes of exposure can be used to treat food samples by DBD device. For direct exposure, the plasma flow is directly applied to the sample. In direct exposure, the sample is placed between two electrodes (Fig. 2a). Applying direct exposure for large scale applications is difficult. In the case of indirect exposure, the sample is placed close to the plasma stream (Fig. 2b) (Misra, Keener, Bourke, & Cullen, 2015). The type of the gas used for generation of plasma, the electrical operation of the discharge, and the distance between the electrodes are important factors in DBD device (Surowsky et al., 2015). The benefit of DBD is that the various types of gas can be used for plasma production. In addition, the DBD generation is very rapid, and a relatively low gas flow rate is needed. Furthermore, it is possible to use different electrode geometries in this device and the gas consumption is low in DBD device. The DBD can provide a relatively simple and homogenous discharge ignition up to several centimeters and foods with solid and liquid states can be treated with this device (Nasiru et al., 2021). Also, this device can treat packaged food. Therefore, food can be

Table 1
Advantages and disadvantages of cold plasma devices.

Cold plasma device	Advantages	Disadvantages	Reference
DBD*	• Possibility to use various types of gas • Rapid DBD generation • Low gas consumption • Possibility to use different electrode geometries	• Requiring a high ignition voltage	Nasiru et al. (2021); Kopuk et al. (2022); Rout and Srivastav (2023)
PJ	• Generating a uniform and steady discharge • High concentration of reactive species • Small size of PJ device • High penetration depth	• Requiring high gas flow rate	Laroque et al. (2022); Lu et al. (2016); Neuenfeldt et al. (2023)
GAD	• Providing stable and flexible operation • Operating at low temperatures • Operating at atmospheric pressure • Possibility to use a wide variety of gas		Dasan et al. (2017); Kopuk et al. (2022)
CD	• Operating at atmospheric pressure • Low operating cost • Simple apparatus	• Small sample area • Non-uniform treatment • Operating at low temperatures	Coutinho et al. (2018)
GD	• Plasma production in a large volume • Operation at low temperatures		Sakudo and Misawa (2020)
MD	• Generate plasma without electrodes • Operating at atmospheric pressure or low pressure • Low gas consumption • High efficiency in generating reactive species • High electron density	• Necessity of applying a series of discharges for ensuring its applicability in large areas	Sakudo & Misawa, 2020; Wongjaikham et al. (2023); Kanca and Avşar (2023); Surowsky et al. (2015)
RFD	• Producing high-density plasma • Operating at atmospheric pressure • High production efficiency for industrial applications		Li et al. (2022)

* DBD: dielectric barrier discharge; PJ: plasma jet; GAD: gliding arc discharge, CD: corona discharge; MD: microwave discharge; RFD: radio frequency discharge

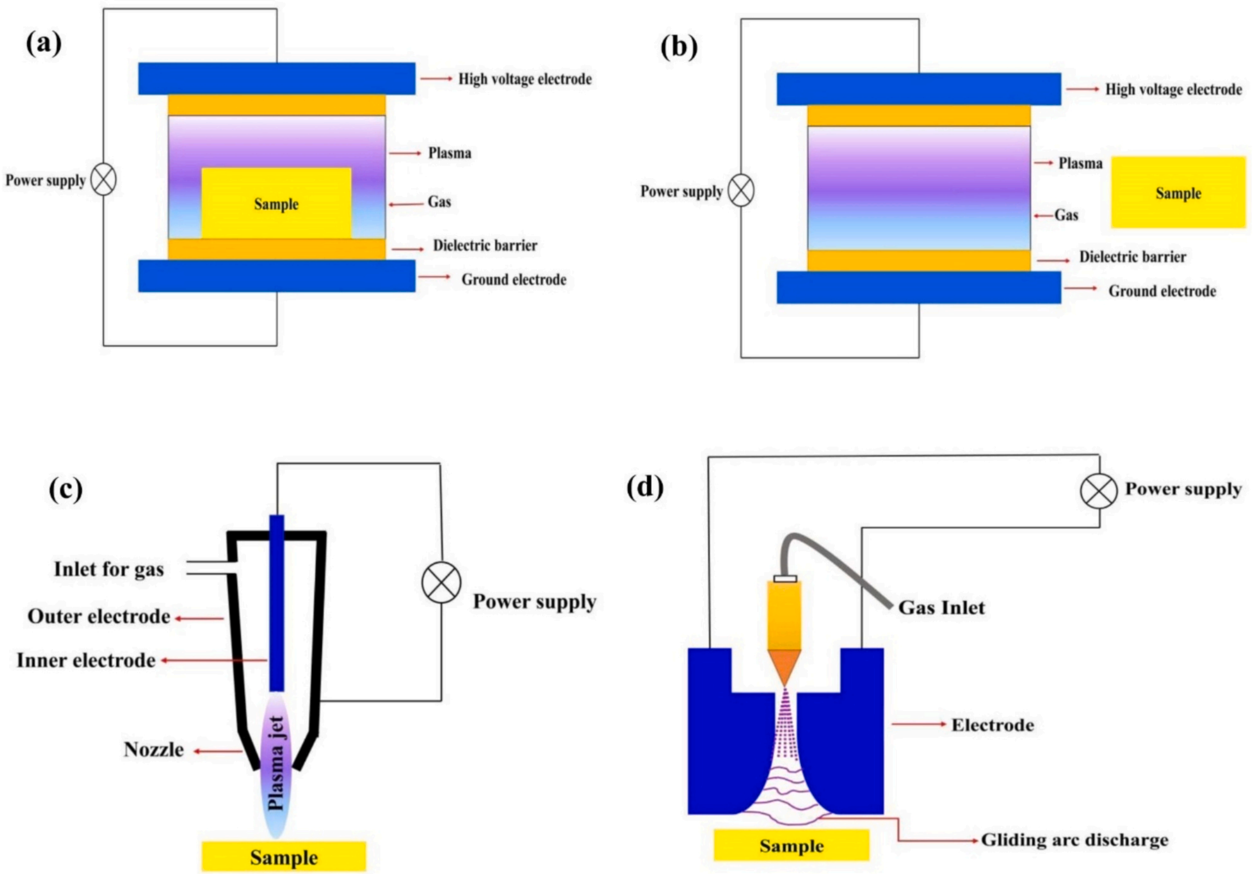

Fig. 2. (a) Direct dielectric barrier discharge device, (b) indirect dielectric barrier discharge device, (c) plasma jet device, and (d) gliding arc discharge device (Adapted from Jadhav and Annapure (2021) and Neuenfeldt et al. (2023)).

packaged before plasma treatment, and the contamination after plasma treatment is reduced (Rout & Srivastav, 2023). However, a high ignition voltage (> 10 kV) is needed for a DBD device. Accordingly, special isolations and precautions are essential for DBD devices (Kopuk et al., 2022). DBD device with parallel plate geometry successfully hydrogenated palm oil (Kongprawes et al., 2023) and soybean oil with high oxidative stability (Yepez & Keener, 2016). Also, needle-in-tube DBD device has been applied to hydrogenate refined palm olein oil. The cold plasma technique produced margarine with no trans-fatty acid with a texture similar to commercial margarine (Puprasit, Wongsawaeng, Ngaosuwan, Kiatkittipong, & Assabumrungrat, 2022). In addition, DBD device has been used for transesterification of vegetable oils (sunflower oil, palm oil, and coconut oil) and production of biodiesel with high efficiency (Nabilla, Anisa, Zara, & Bismo, 2020; Taki, Hosseinzadeh Samani, & Anasari Ardali, 2024).

2.2.2. PJ

PJ typically contains two concentric electrodes with a two-ring or coaxial geometry (Fig. 2c). The inner electrode is connected to a power that ionizes the gas, resulting in plasma discharge in a “jet-like” appearance from a nozel. The system is excited by radio waves (generally at 13.56 MHz frequency). When the radio waves excite the system, the outer electrode accelerates free electrons. The collision of free electrons with gas molecules results in generating various reactive species. A mixture of atmospheric gas (oxygen, nitrogen, etc.) and noble gas (argon, helium, etc.) is usually used as the working gas. The atmospheric gas can supply reactive species, while the noble gas provides easy ionization. A gas with high speed which is typically a noble gas, derives the produced plasma beyond the electrode zone, and incorporate the reactive species into the liquid oil. A uniform and steady discharge is generated by the PJ device (Neuenfeldt, Silva, Pessoa, & Rocha, 2023). In the PJ device, the density of charged species (10^{11}–10^{12} cm^3) and the concentration of reactive species (10–100 ppm) is high. The small size of PJ device and its high penetration depth are the benefits of PJ device. In addition, the PJ device can be effectively applied for confined or small areas. (Laroque et al., 2022; Lu, Cullen, & Ostrikov, 2016). However, applying PJ device for food industry is limited due to the high costs associated with maintaining the required gas flow (Neuenfeldt et al., 2023). PJ device has been applied for transesterification of sunflower oil with methanol to produce biodiesel. A high reaction conversion (83 %) was obtained using a PJ device (Ansari Samani, Hosseinzadeh Samani, Ghasemi-Varnamkhasti, Rostami, & Ebrahimi, 2023).

2.2.3. Gad

The GAD device consists of two identical electrodes. The electrodes are separated at one end via a narrow gap that progressively widen towards the opposite end (Fig. 2d). The working gas is provided by a tube inserted into the narrower section. Also, a gas flow meter regulates the gas flow. A high voltage is applied to the electrode resulting in the formation of an arc discharge that causes an electrical breakdown of the gas exists between two electrodes at the narrowest part of the electrodes (Onal-Ulusoy, 2021). The GAD device requires a current flow of approximately 10 A and a voltage difference of around 100 V to operate efficiently. The GAD can provide stable and flexible operation with a careful power supply system. This device can operate at low temperatures and atmospheric pressure. In addition, a wide variety of gases can be used for plasma generation in this device (Dasan et al., 2017; Kopuk et al., 2022). The rotating GAD device has been applied for converting waste rapeseed oil into high value compounds (Wu et al., 2015).

2.2.4. Cd

CD is an effect that was found in the regions of electrodes that have a sharp curvature or pointiness. In a CD device, a high voltage is applied between two electrodes. One of the electrodes can be a thin wire electrode or a point electrode. The thin wire electrode is called the emitter electrode, and the plane electrode is called the collector electrode (Fig. 3a) (Narimisa et al., 2024). The high voltage produces a strong electric field. This electric field is usually concentrates on surfaces with significant curvature like pinpoint structures, thin wires, or pointy ends. The interaction of this electric field with the neutral gas surrounding these regions results in gas ionization. In this device, the generated plasma has a filamentary appearance, which is concentrated at the tips of the surfaces (Neuenfeldt et al., 2023). CD device typically operates at atmospheric pressure. Also, the CD device operates in pulsed voltage or direct current modes. The low operating cost and simple apparatus are the benefits of this device. The small sample area and non-uniform treatment are the disadvantages of this device (Coutinho et al., 2018). The CD device has been applied for transesterification of waste frying oil and biodiesel production (Cubas, Machado, Pinto, Moecke, & Dutra, 2016).

2.2.5. GD

Glow discharge contains a moderate density of electrons and ionized gas with a high-energy density.

In this device, a direct voltage (> 100 V) is applied to a gas under a low vacuum (1–10^3 Pa) to generate plasma (Fig. 3b). In this device, the distance between electrodes is small. Enhancing the distance between the electrodes or enhancing the pressure can make the discharge unstable, resulting in a transition to corona discharge (Neuenfeldt et al., 2023). In a GD device, at least one electrode should be covered via an insulating layer. In this device, after turning the power on, the charged species will accumulate on the insulating layer surface, resulting in the formation of a potential difference between the insulating layer (Zhang et al., 2022). The main benefits of this technique are plasma production in a large volume and at low temperatures (Sakudo & Misawa, 2020).

2.2.6. MD

In this device, a magnetron produces MD without using electrodes (Fig. 3c). Absorbing the microwave by electrons increases their kinetic energy, inelastic collisions, and ionization reactions. Therefore, neutral components ionize, and electrons diffuse away from the gas phase. When the ionization rate is higher than the electron diffusion rate, plasma is generated. Based on the microwave energy consumed, the neutral gas can enhance the temperature from ambient temperature to 1000 K. Therefore, this system may need a cooling system. The MD device consist of a microwave power producer (magnetron), microwave power detectors (reflected power and incident power), circulator (an apparatus for protecting the magnetron from reflected power), microwave-to-plasma applicator, and discharge chamber. The most important component of MD device is microwave-to-plasma applicator because it allows microwave radiation to transfer into the plasma. The main benefit of this device is that it can generate plasma without electrodes. Therefore, the possibility of plasma or gas contamination by electrode corrosion is reduced (Kopuk et al., 2023; Wongjaikham et al., 2023). In addition, this system can operate at atmospheric pressure or low pressure. Furthermore, a low amount of gas is required to produce a large amount of active material (Sakudo & Misawa, 2020). High efficiency in generating reactive species, high electron density, and ease of

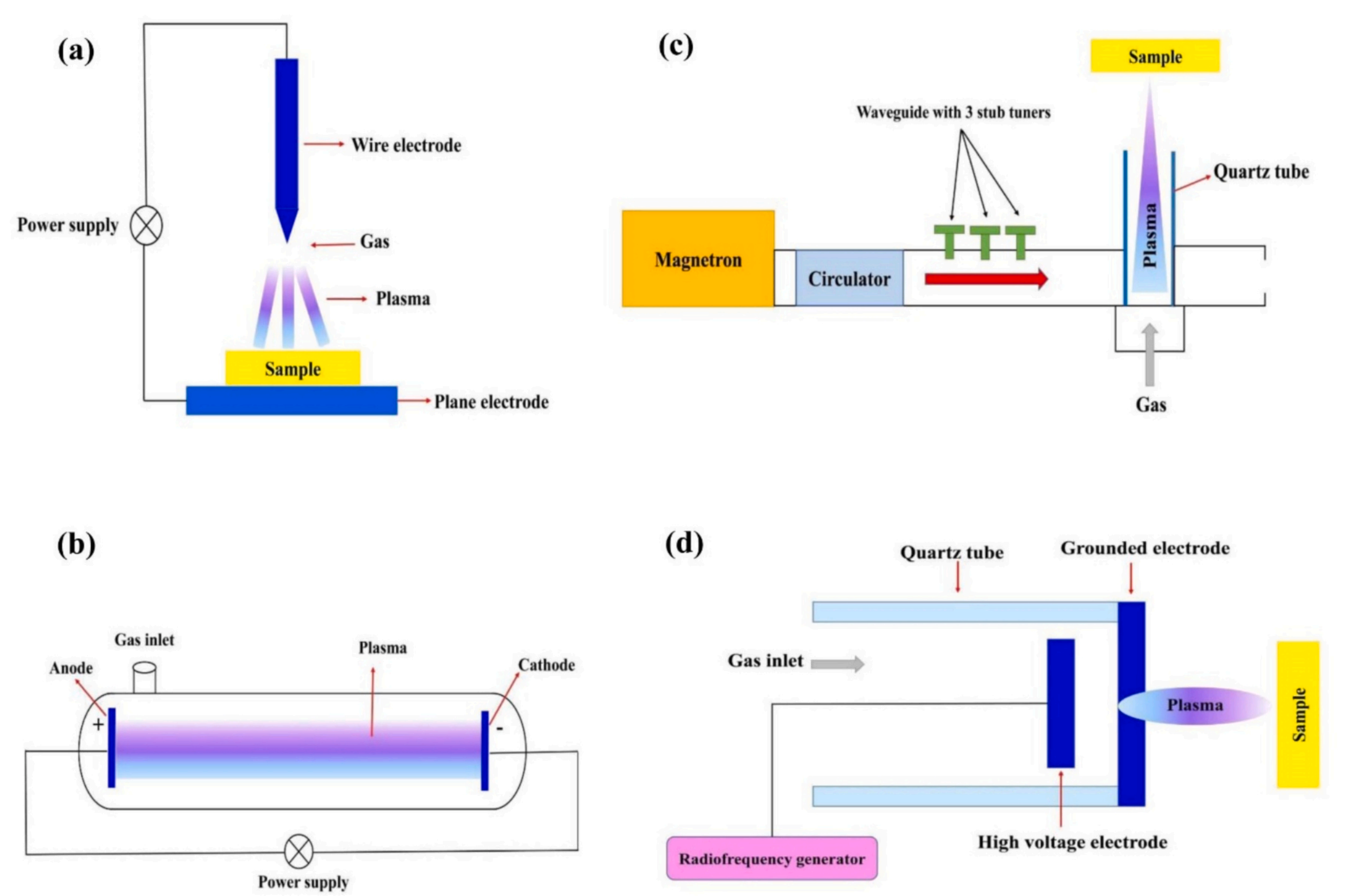

Fig. 3. (a) Corona discharge device, (b) glow discharge device, (c) microwave discharge device, and (d) radiofrequency discharge device (Adapted from Kumar, Pipliya, and Srivastav (2023); Surowsky et al. (2015); Dinescu, Ionita, Luciu, and Grisolia (2007)).

control of microwave-to-plasma conversion via applying an external applicator are the other advantages of MD device (Wongjaikham et al., 2023). The disadvantage of this system is the necessity of applying a series of discharges to ensure its applicability in large areas (Kanca & Avşar, 2023; Surowsky et al., 2015). The MD device has been used for the hydrogenation of palm oil. A margarine with suitable texture, low trans-fatty acids, and slip melting point was produced by MD device (Wongjaikham et al., 2022).

2.2.7. RFD

In this device, plasma is generated by radio frequency at 13.56 MHz. In order to create a radiofrequency field, several methods can be used. These methods include applying radiofrequency voltage to electrodes with parallel-plate geometry (Fig. 3d) or using circulating radio-frequency current in antennas or coils by submerging in the plasma. Inductively coupled plasma, helicon wave sources, and capacitively coupled plasmas are the three main types of radiofrequency plasma. The advantage of an RFD device is producing high-density plasma at atmospheric pressure with little or no extra heat input. High production efficiency for industrial applications is the other benefit of RFD (Li et al., 2022). Kim, Lee, Choi, and Kim (2014) studied the effect of an RFD plasma device on the fatty acid composition of beef jerky. No significant change was observed in the fatty acid composition of beef jerky after exposing to RFD plasma of 200 W for 300 S (Kim et al., 2014). Upadhyay, Thirumdas, Deshmukh, Annapure, and Misra (2019) studied the effect of RFD plasma on the fatty acid composition of chia flour. The fatty acid composition of chia seed power was decreased by 9 % during RFD plasma treatment at 60 W for 900 s. However, trans-fatty acid was also found after RFD plasma treatment (Upadhyay et al., 2019).

3. Effect of cold plasma technique on liquid oil reactions

3.1. Oxidation

Lipid oxidation reactions can be classified into photo-oxidation, thermal oxidation, and auto-oxidation. In auto-oxidation reaction, food lipids spontaneously react with oxygen through the free radical chain reaction. Auto-oxidation reaction consists of three stages of initiation, propagation, and termination. During the initiation phase, the hydrogen atom (H) from the carbon next to the unsaturated fatty acid (RH) double bond is absorbed in the presence of an initiator (heat, light, and metal ions), and alkyl free radical ($R^{\bullet}$) is produced (Estévez & Cava, 2004; Fennema, Damodaran, & Parkin, 2008).

$$RH \rightarrow R^{\bullet} + H^{\bullet}$$

In the propagation phase, the $R^{\bullet}$ reacts with oxygen (O_2) and lipid peroxide radical ($ROO^{\bullet}$) is formed. The $ROO^{\bullet}$ abstract the hydrogens of other unsaturated fatty acid and generate lipid hydroperoxide (ROOH) and another $R^{\bullet}$ (Fennema et al., 2008).

$$R^{\bullet} + O_2 \rightarrow ROO^{\bullet}$$

$$ROO^{\bullet} + RH \rightarrow ROOH + R^{\bullet}$$

When the concentration of free radicals is high, the termination phase can occur. In this phase, free radicals are combined, and non-radical species are formed (Estévez & Cava, 2004; Fennema et al., 2008).

$$R^{\bullet} + R^{\bullet} \rightarrow R - R$$

$$ROO^{\bullet} + R^{\bullet} \rightarrow ROOR$$

The reactive species in plasma can initiate lipid oxidation in liquid oil, resulting in altering the fatty acid composition and reducing the nutritional value and sensory characteristics of liquid oils (Fig. 4). The reactive oxygen species present in cold plasma such as hydroxyl radicals and ozone can interact with unsaturated fatty acids and generate ozonide. Further oxidation results in the generation of aldehydes and ketones (Bayati, Lund, Tiwari, & Poojary, 2024). Díaz, Hernández, Ledea, Sazatornil, and Moleiro (2003) detected the formation of nonanal and 9-oxononanoic acid in the unsaturated fatty acids exposed to cold plasma. Summary of the researches on the impact of cold plasma technique on liquid oil oxidation is presented in Table 2.

3.1.1. Effect of cold plasma technique parameters on lipid oxidation rate of oils

Several researches have studied the effect of plasma technique parameters on the lipid oxidation of liquid oils. Palm oil was treated with atmospheric cold plasma at different powers (50 and 60 W) and time (50 and 60 W). After cold plasma treatment, the samples were stored for one week. After one week storage, the amounts of peroxide values (indicator of primary oxidation products (lipid hydroperoxide)) and anisidine

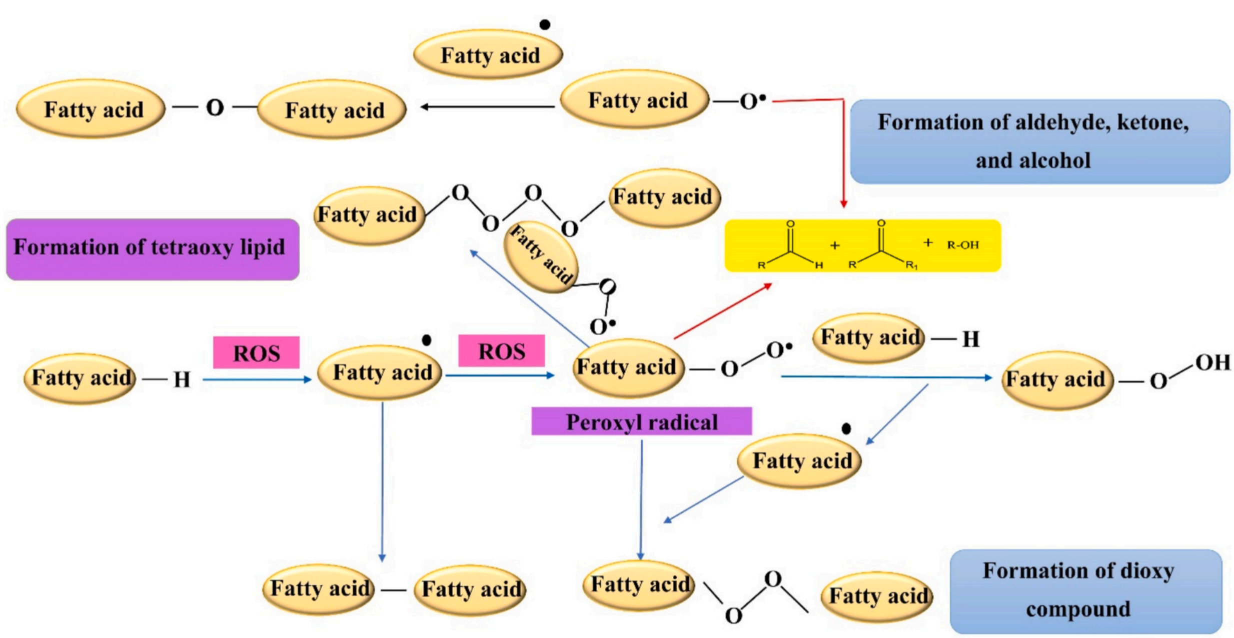

Fig. 4. Effect of cold plasma technique on lipid oxidation (Saremnezhad et al., 2021).

Table 2
Summary of the investigations on the impact of cold plasma technique on liquid oil oxidation.*

Oil type	Plasma device	Carrier gas	Treatment time (min)	Oxidation assay	Findings	Reference
Stripped corn oil	Corona discharge plasma jet, 220 V, 150 mA	Atmospheric air	10	CDA*	The amount of CDA in stripped corn oil significantly increased after 10 min of plasma treatment	Na et al. (2020)
Olive oil	DBD plasma jet, 50 kHz, 6 kV	99.9 % Ar + 0.1 % O_2	60	GC/MS	The amounts of secondary oxidation products increased during plasma treatment	Van Durme and Vandamme (2016).
Fish oil	DBD plasma jet, 50 kHz, 6 kV	99.4 % Ar + 0.6 % O_2	60	HS-SPME–GC/MS	Plasma treatment enhanced the formation of secondary oxidation products	Vandamme et al. (2015)
Palm oil	Bell-jar type plasma reactor, 13.56 MHz, 50 and 60 W	Atmospheric air	30 and 45	PV and AV	The amount of primary and secondary oxidation products was increased by increasing input power and treatment time	Niveditha et al. (2023)
Soybean oil fatty acid methyl ester	Parallel-plate DBD plasma, 25 kHz, 30 mA	75 % He +25 % H_2	330	Rancimat	Partial hydrogenation of fatty acid methyl ester by cold plasma technique enhanced its oxidative stability	Kongprawes, Wongsawaeng, Hosemann, et al. (2021)
Palm oil fatty acid methyl ester	Microwave discharge plasma, 400 W, 30 kV	H_2	180	Rancimat	Partial hydrogenation of fatty acid methyl ester by microwave plasma technique reduced its oxidation rate	Wongjaikham et al. (2021)
Palm biodiesel	DBD plasma, 20 kHz, 40 mA	80 % H_2 + 20 % He	300	Rancimat	Partial hydrogenation of biodiesel by DBD plasma improved its oxidative stability	Kongprawes, Wongsawaeng, Ngaosuwan, et al. (2021)
Soybean oil	DBD plasma, 120 V	H_2	5	PV	Soybean oil samples treated with plasma technique exhibited a higher PV than untreated samples	Yepez et al. (2021)
Fish oil	DBD plasma, 50 V	–	3	PV, AV, TOV, MDA, CD, and CT	Cold plasma treatment directly induced oxidation. Vitamin E showed a high antioxidant activity in fish oil treated with cold plasma	Wang et al. (2024)
Camellia oil	DBD plasma	Atmospheric air	1	PV	Cold plasma technique enhanced the PV by 17.6 %	Chen et al. (2024)

* CDA: conjugated dienoic acid; DBD: dielectric barrier discharge; GC/MS: gas chromatography/mass spectrometry; HS-SPME-GC/MS: headspace-solid phase microextraction combine with GC/MS; PV: peroxide value; AV: Anisidine value.

values (indicator of secondary oxidation products (aldehydes and ketones)) of palm oil samples were measured. The results showed that the amounts of peroxide values and anisidine values of palm oil samples treated with atmospheric cold plasma were higher than those of untreated palm oil samples. Also, the palm oil sample treated with cold plasma at 60 W for 45 min showed higher peroxide value and anisidine value than those palm oil samples treated at 60 W for 30 min or 50 W for 45 min. This can be due to the enhanced number of reactive species at a higher power of treatment when treated for a longer period of time (Niveditha, Jadhav, Ahlawat, Kalaivendan, & Annapure, 2023). The chemical composition of the gas used for plasma production can significantly affect the lipid oxidation rate. Van Durme, Nikiforov, Vandamme, Leys, and De Winne (2014) evaluated the effect of cold plasma technique on the formation of secondary oxidation products in vegetable oil. When the pure argon gas was used for plasma production, no oxidation product was observed. When O_2 or H_2O was present in the argon gas, the production of several secondary oxidation products was observed. The chemical composition of the secondary oxidation products detected in the cold plasma technique was similar to those observed in the natural peroxidation process (Van Durme et al., 2014). Also, it has been reported that when the working gas was dry air, $^{\bullet}NO_2$, O_3, $^1O_2$, and $^{\bullet}NO_3$ were the major reactive species. However, when humid air was used as the working gas, O_2NOOH and ONOOH were produced and lower amount of O_3 was detected. It was suggested that reactive nitrogen species ($^{\bullet}NO_3$, $^{\bullet}NO_2$, O_2NOOH, and ONOOH) resulted in the production of hydroperoxides, while O_3 mainly led to the production of carbonyl compounds through the trioxolane pathway (Liu et al., 2023). Sang et al. (2024) evaluated the impact of gas composition (gas A: 50 % N_2, 40 % CO_2, and 10 % O_2; gas B: air; gas C: 40 % CO2, 30 % N_2, 30 % O_2) on tilapia fillets auto-oxidation. The tilapia fillet samples treated with gas C group showed a higher oxidation rate than other samples. Therefore, enhancing the O_2 concentration in the working gas can increase auto-oxidation.

Liu, Wang, Wang, and Chen (2023) found that enhancing the treatment time and voltage of cold plasma can enhance the oxidation rate of tilapia fillets. This enhancement in the oxidation rate followed the zero-order reaction kinetic model. Also, they indicated that applied voltage affected the activation energy of oxidation to the higher extent than treatment time did. In addition, they observed that at voltage greater than 64.71 kV, the activation energy for primary lipid oxidation was higher than that of secondary lipid oxidation. Thus, in tilapia fillets the primary lipid oxidation was more susceptible to the cold plasma processing conditions. The auto-oxidation rate of fish samples treated with a DBD device at 70 kV was lower than those samples treated at 80 kV (Albertos et al., 2017). Liu, Van Paepeghem, et al. (2023) reported that the peroxide value and anisidine value of the stripped linseed oil methyl ester samples were enhanced by increasing the treatment time. Also, the peroxide value and anisidine value of the stripped linseed oil methyl ester samples were enhanced by increasing the energy input. A high correlation (R2 > 0.99) was observed between the peroxide value and energy input. They observed that treatment time is the most prominent factor influencing the auto-oxidation, followed by the power input and the plasma-sample distance (Liu, Van Paepeghem, et al., 2023).

In some research, the cold plasma technique did not significantly enhance auto-oxidation (Lee et al., 2018). The cold plasma parameters such as gas type, input power, and treatment time can affect the auto-oxidation rate. Also, the fatty acid composition of liquid oil, and storage conditions before and after cold plasma treatment can significantly impact lipid oxidation rate (Gavahian et al., 2018). The lipid oxidation rate during cold plasma treatment of liquid oils can be minimized by carefully selecting the plasma parameters such as plasma power, duration of plasma treatment, and chemical composition of the gas used for plasma production (Jadhav & Annapure, 2021).

3.1.2. Strategies to reduce lipid oxidation rate during cold plasma treatment

Incorporating antioxidants into liquid oil can significantly enhance its oxidative stability during cold plasma treatment. Na et al. (2020) observed that α-tocopherol and sesamol which are hydrogen donator antioxidants significantly decreased peroxidation rate in stripped corn oil. The amount of conjugated dienoic acid in the samples containing α-tocopherol, sesamol, and the control sample were 0.16 %, 0.17 %, and 0.24 %, respectively. This indicates the production of free radicals in stripped corn oil treated with cold plasma technique. Adding β-carotene which is a singlet oxygen quencher significantly reduced the conjugated dienoic acid value compared to the control sample. The amount of conjugated dienoic acid in the sample containing β-carotene was 0.22 %. This indicates that singlet oxygen may be the main oxidizing agent produced by cold plasma technique in stripped corn oil. Also, they observed that α- and γ-tocopherol showed different stability in the plasma treatment. After plasma treatment, α-tocopherol reduced faster in medium-chain triacylglycerol and stripped corn oil than γ-tocopherol (Na et al., 2020). Wang et al. (2024) investigated the effect of curcumin, tea polyphenols, vitamin E, and β-carotene in reducing fish oil oxidation during cold plasma treatment. Incorporating antioxidants during cold plasma treatment reduced the peroxide and malondialdehyde values. Vitamin E showed higher efficiency than other antioxidants in inhibiting fish oil oxidation (Wang et al., 2024).

The storage condition of liquid oil before plasma treatment can also affect lipid oxidation rate during plasma treatment. Exposure of liquid oil to light and oxygen can initiate the peroxidation process and make the liquid oil more susceptible to the reactive species present in plasma. Therefore, liquid oil should not be exposed to oxygen and light during the storage period before plasma treatment (Gavahian et al., 2018).

As stated in section 3.1.1, cold plasma processing parameters can significantly affect the lipid oxidation rate. Setting the plasma parameters at the lowest possible input power and shortest possible duration and omitting oxygen from the gas used for plasma production can reduce the lipid oxidation rate during plasma treatment (Gavahian et al., 2018). According to Sang et al. (2024), the lipid oxidation rate of tilapia fillets was reduced by decreasing oxygen concentration of the gas used for plasma production form 30 % to 10 %. After 8 day storage, the peroxide value of the tilapia fillet treated with 10 % O_2 was 0.49 meq/kg, while the peroxide value of the tilapia filet treated with 30 % O_2 was 0.57 meq/kg (Sang et al., 2024). It has been reported that the peroxide value of tilapia fillets treated at 80 kV was 22 nmole/kg, while the peroxide value of tilapia fillet treated at 40 kV was 2.2 nmole/kg during storage at 25 °C. Also, when the samples were treated at the same voltage, enhancing the storage temperature from 4 °C to 25 °C resulted in a significant increase in peroxide value ($P < 0.05$). Besides, the peroxide value was decreased from 15 nmole/kg to 6 nmole/kg by reducing the treatment time from 300 s to 60 s (Liu, Wang, et al., 2023).

Taken together, lipid oxidation during cold plasma treatment can be minimized through storing liquid oil at suitable conditions before plasma treatment, incorporating efficient antioxidants into liquid oil before cold plasma treatment, and choosing appropiate input power, treatment time, and carrier gas for cold plasma treatment (Gavahian et al., 2018).

3.1.3. Cold plasma as a novel non-thermal technique for monitoring oxidative stability of liquid oil

Cold plasma technique can accelerate the oxidation rate at a shorter time compared to UV radiation and thermal oxidation. It has been reported that the amount of conjugated dienoic acid in stripped corn oil significantly increased after 10 min of plasma treatment. The same degree of conjugated dienoic acid was obtained after 48 h storage at 60 °C and 2.5 h storage at 100 °C (Na et al., 2020). Also, Vandamme et al. (2015) stated that DBD-PJ with the gas composition of argon and 0.6 % oxygen (O_2) can be used as a faster and more reliable oxidation test compared to thermal oxidation test for accelerating fish oil oxidation. In addition, they observed that α-tocopherol at 1000 ppm concentration showed antioxidative effect when incorporated into fish oil before plasma treatment, while it showed pro-oxidative effect in thermal oxidation test. Accordingly, cold plasma is a more reliable technique than the thermal oxidation test for determining the efficiency of antioxidants in fish oil (Vandamme et al., 2015).

3.2. Hydrogenation

3.2.1. Hydrogenation of liquid oil by conventional technique

In the hydrogenation reaction, unsaturated double bonds of liquid oils are converted to saturated bonds, and a partially hydrogenated oil is produced. In the conventional hydrogenation process, the liquid oil is hydrogenated in the presence of hydrogen gas and a chemical catalyst such as nickel, platinum, or palladium. This process is usually carried out under high pressure (0.07–0.4 MPa) and temperature (120–250 °C). The high temperature and the catalyst needed for conventional hydrogenation can result in the formation of trans-fatty acids via the conversion of cis isomer of unsaturated fatty acid into the trans isomer (Wongjaikham et al., 2023).

3.2.2. Hydrogenation of liquid oil by cold plasma technique

In the cold plasma technique, hydrogen gas alone or a mixture of hydrogen gas and other inert/noble gas is used for plasma production in the hydrogenation process. Different forms of hydrogen can be generated during plasma treatment. These include atomic (2H), ionic ($2H^+$, $2H^{2+}$, and $2H^{3+}$), and vibrational activated (H_2*) forms. The reduction capacities of these forms are in the following order: $H^+ > H_2^+ > H$ (Sabat, Rajput, Paramguru, Bhoi, & Mishra, 2014). The atomic hydrogen converts the unsaturated bond to the saturated bond. In cold plasma technique, high-energy electrons can generate radicals and active species which can act as catalyst, rapidly interact with lipids, and promote hydrogenation pathways. Accordingly, no physical catalyst would be needed for the hydrogenation reaction. Cold plasma technique can facilitate hydrogenation reaction to occur at ambient temperature and atmospheric pressure without catalyst (Wongjaikham et al., 2023). Yepez, Baykara, Xu, and Keener (2021) reported that high voltage atmospheric cold plasma can generate reactive species which can hydrogenate and polymerize triacylglycerols of soybean oil. Besides, isolated double bonds containing bis-allylic hydrogens can be converted to conjugated double bonds under high voltage atmospheric cold plasma treatment (Yepez et al., 2021). In the cold plasma technique, electrons with high energy can form radicals and active species. These radicals and active species can act as catalyst and promote hydrogenation reaction (Wongjaikham et al., 2023).

The use of high temperature and catalyst are associated with the generation of trans-fatty acids. Therefore, hydrogenation with cold plasma technique can reduce the formation of trans-fatty acids. The main advantage of using a catalyst for hydrogenation reaction is high productivity and high reaction selectivity. The low reaction selectivity is the main drawback of hydrogenation by cold plasma technique. The low reaction selectivity can result in the formation of unwanted side products (Wongjaikham et al., 2023). Hydrogenation of soybean oil by cold plasma technique was carried out at 90 kV. The treatment time was 120 min, and the gas flow rate was 10 L/min. Atomic hydrogen species identified by optical emission spectroscopy were the main reactive species responsible for the hydrogenation reaction by high-voltage atmospheric cold plasma (Yepez & Keener, 2016). Also, microwave plasma technique was applied to produce low trans-fat margarine via partial hydrogenation of palm olein. Hydrogen flow rate of 4 L/min, temperature of 32 °C, microwave power of 600 W, reaction time of 4 h, and negative voltage of 60 kV was suggested as the best processing parameter for production of margarine with low trans-fatty acid, slip melting point, and good texture. The percentage of trans-fatty acid was 4.23 %, which was lower than that of conventional hydrogenation (Wongjaikham et al., 2022). The results of above studies indicate that DBD plasma technique and microwave plasma technique can use as green technique for hydrogenation of liquid oil at low temperature, without catalyst, and without formation of trans-fatty acids. However, besides the benefits of hydrogenation by cold plasma technique, the major disadvantage of hydrogenation by cold plasma technique is the high amount of gas required for plasma production (Kongprawes et al., 2021). A Summary of the researches on the application of cold plasma technique for liquid oil hydrogenation is presented in Table 3. Gas type and concentration, gas flow rate, input power, electrode distance, and reaction temperature can affect hydrogenation by cold plasma technique (Fig. 1S) (Wongjaikham et al., 2023).

3.2.3. Factors affecting hydrogenation by cold plasma technique

3.2.3.1. Gas type and concentration. Hydrogen is typically used as the feed gas for hydrogenation of liquid oil. A mixture of hydrogen with other gases can also be used for hydrogenation. The impact of H_2:He ratio (25–100 % H_2) on the hydrogenation rate of refined palm olein for production of margarine in a DBD cold plasma with needle-in-tube configuration was investigated. The hydrogenation rate was increased by increasing the H_2 concentration from 25 % to 100 %. Also, the hydrogen concentration affected the temperature of the chamber. For 100 % H_2 and 100 % He, the temperature of the reaction chamber was measured to be 50.25 ± 0.35 °C and 48.75 ± 0.35 °C. The higher temperature in the presence of 100 % H_2 can be due to the exothermic nature of hydrogenation, which resulted in the release of the highest amount of heat when the hydrogenation rate was the highest. Although the high temperature can cause the formation of trans-fatty acids, the production of trans-fatty acids was not observed when 100 % H_2 was used (Puprasit et al., 2022). Yepez and Keener (2016) used a mixture of H_2 and N_2 (5 % H_2:95 % N_2) for hydrogenation of soybean oil by high-voltage atmospheric cold plasma technique. They observed considerable conversion of linolenic acid and linolenic acid to oleic acid and stearic acid during hydrogenation for 12 h. Also, Kongprawes, Wongsawaeng, Hosemann, et al. (2021), (Kongprawes, Wongsawaeng, Ngaosuwan, Kiatkittipong, & Assabumrungrat, 2021) investigated the effect of the H_2:He ratio (5 % H2:95 % He to 25 % H2:75 % He) on palm oil biodiesel hydrogenation. They reported that H_2 concentration at 80 % showed higher hydrogenation rate of palm oil biodiesel than those of 52.5 % and 25 % in DBD cold plasma device. When H_2 was used at 80 % concentration, the concentration of palmitic acid and stearic acid was increased by 1.4 % and 20.4 %, respectively. In comparison, the linoleic acid and linolenic acid concentrations were decreased by 13.4 % and 38 %, respectively. In contrast, the mixture of 15 % H_2:85 % He resulted in the faster hydrogenation of palm oil than those mixtures of 20 % H_2:80 % He and 25 % H_2:75 % He in a DBD plasma with parallel-plate configuration (Puprasit, Wongsawaeng, Ngaosuwan, Kiatkittipong, & Assabumrungrat, 2020). This was probably because when the H_2 concentration was high, the large amount of produced H_2 free radicals experienced smaller mean free paths and recombination became significant, which resulted in lower amount of atomic hydrogen being present in the reaction chamber to hydrogenate vegetable oil (Grozdanov, 2014). More studies are needed to find efficient and cheap hydrogen donors for hydrogenation by cold plasma technique.

3.2.3.2. Gas flow rate. The gas flow rate, which indicates the plasma density and stability, can impact the hydrogenation rate. The gas flow rate can affect the convection and balance of excited species and charged particles in the plasma region (Höft, Becker, & Kettlitz, 2016). Kongprawes et al. (2023) examined the effect of gas flow rate at 0.2, 0.5, and 0.8 L/min on the hydrogenation of palm oil using DBD cold plasma. Glycerol was used as a hydrogen donor. They observed that the best gas flow rate was 0.5 L/min, followed by 0.8 and 0.2 L/min, respectively. The lower efficiency of 0.8 L/min gas flow rate than that of 0.5 L/min might be due to the low residence time of reactive species in the reaction chamber and the elimination of the reactive species from the reaction chamber before interacting with glycerol. The lower efficiency at 0.2 L/min flow rate can be due to the presence of lower amounts of reactive species in the reaction chamber, resulting in lower hydrogenation reaction rate (Kongprawes et al., 2023). According to Kamjam et al. (2022) different flow rate of H_2 gas led to different hydrogenation rate and plasma density. Increasing H_2 flow rate led to production of higher amounts of active hydrogen radicals. This can result in providing higher homogeneity of plasma distribution and higher plasma density and enhancing the hydrogenation rate. However, a medium gas flow rate can provide the best effect on the hydrogenation reaction. It has been suggested that when the flow rate is too low, fewer free radicals are generated, and the generated plasma is unstable (Kamjam et al., 2022). In addition, applying a high gas flow rate can enhance production

Table 3
Summary of the investigations on the application of cold plasma technique for liquid oil hydrogenation.

Oil type	Plasma device	Carrier gas	Treatment time (min)	Findings	Reference
Palm oil	Parallel-plate DBD plasma, 20 kHz, 15 kV*	He	120	Hydrogen radicals extracted from glycerol by plasma technique hydrogenated unsaturated fatty acids	Kongprawes et al. (2023)
Soybean oil	Parallel-plate DBD plasma, 90 V	95 % N_2 + 5 % H_2	120	Plasma technique reduced the iodine value similar to the traditional hydrogenation process	Yepez and Keener (2016)
Refined palm olein	Needle-in-tube DBD plasma, 40 W	H_2	900	Plasma technique produced margarine with no trans-fatty acid with a texture similar to commercial margarin	Puprasit et al. (2022)
Palm oil	Parallel-plate type DBD plasma, 25 kHz	85 % He +15 % H_2	240	Margarine with low trans-fatty acids was produced by plasma technique	Puprasit et al. (2020)
Soybean oil fatty acid methyl ester	Parallel-plate DBD plasma, 25 kHz, 30 mA	75 % He +25 % H_2	330	DBD plasma hydrogenation significantly reduced iodine value	Kongprawes, Wongsawaeng, Hosemann, et al. (2021)
Palm oil fatty acid methyl ester	Microwave discharge plasma, 600 W, 60 kV	H_2	180	Microwave plasma effectively hydrogenated palm oil fatty acid methyl ester at low temperatures without using catalyst	Wongjaikham et al. (2021)
Palm biodiesel	DBD plasma, 20 kHz, 40 mA	80 % H_2 + 20 % He	300	DBD plasma exhibited a better performance than conventional hydrogenation technique	Kongprawes, Wongsawaeng, Ngaosuwan, et al. (2021)
Soybean oil	DBD plasma, 120 V	H_2	5	Cold plasma treatment decreased the amount of polyunsaturated fatty acid and increased the amount of saturated fatty acids	Yepez et al. (2021)
Palm oil	Microwave discharge plasma, 600 W, 60 kV	H_2	240	Microwave plasma produced margarine with good texture, slip melting point, and low trans-fatty acids	Wongjaikham et al. (2022)
Palm oil	DBD plasma, 35 W	He	720	Applying glycerol as hydrogen donor resulted in the production of trans-fat-free margarine with high hydrogenation rate	Priyanti et al. (2024)
Soybean oil	DBD plasma, 230 V	H_2	780	The total content of unsaturated fatty acids was decreased. The amounts of trans-fatty acids were very low.	Sirati, Gharachorloo, Ghomi Marzdashti, and Azizinezhad (2025)

* DBD: dielectric barrier discharge.

expenses unreasonably. Furthermore, high flow rates can result in rapid species recombination. Besides, a high flow rate can lead to rapid leaving of the reactive species from the system through the outlet port. This can result in the reaction of fewer hydrogen free radicals with unsaturated double bonds (Wongjaikham et al., 2023).

The plasma generation source can affect the optimum gas flow rate for the hydrogenation reaction. For instance, the optimum gas flow rate for biodiesel hydrogenation in a DBD torch device was 0.35 L/min (Mustafa et al., 2018), while that for a DBD parallel plate device was 4 L/min (Wongjaikham et al., 2022). In the case of microwave plasma technique which can produce higher density plasma than that of DBD plasma, the optimum gas flow rate was 8.5 L/min and the excess hydrogen was recycled (Wongjaikham et al., 2021). From the above results, it can be concluded that the optimum gas flow rate depends on the type of plasma device used for plasma production.

3.2.3.3. Input power. The input power is one of the most important parameters affecting the hydrogenation rate. In DBD plasma, the plasma discharge power is enhanced by enhancing the input power. Kongprawes, Wongsawaeng, Ngaosuwan, et al., 2021reported that by increasing the input power from 50 to 100 W, the discharge voltage was enhanced from 1.76 to 2.1 kV. They stated that the highest hydrogenation rate was obtained at an input power of 100 W. The high input power provided higher plasma density, resulting in higher amounts of H_2 to react with unsaturated fatty acids. In addition, higher discharge voltage generated higher amounts of high energy electrons which can provide higher collision frequency with H_2 molecules and increases the hydrogenation reaction rate (Kongprawes, Wongsawaeng, Ngaosuwan, et al., 2021). The effect of input power at 35, 50, and 75 W on the hydrogenation of palm olein oil for margarine production was investigated. The conversion of unsaturated fatty acids to saturated fatty acids was 36.2 % at the input power of 75 W, while the conversions at 35 and 50 W were 15.7 % and 26.8 %, respectively. The lower conversion rate at lower input power may be related to the relatively lower energy levels of excited helium, which can result in weaker chemical bond cleavage (Priyanti et al., 2024). Puprasit et al. (2022) evaluated the impact of various input power of 20, 40, 60, and 80 W on the hydrogenation rate of refined palm olein in a DBD cold plasma device with needle-in-tube configuration. The input power of 40 W showed the lowest iodine value. The discharge voltage and the plasma density are enhanced by increasing the input power. This was attributed to the higher dissociation of H_2 molecules into free radicals (Žigon, Petrič, & Dahle, 2018). However, it was observed that the hydrogenation rate was decreased when the input power was higher than 40 W. This was explained by the enhancement in temperature at higher input power. By increasing the input power from 40 to 80 W, the temperature was enhanced from 48.5 °C to 74.75 °C. High temperatures may cause higher desorption of H_2 molecules and reduce the hydrogenation rate (Puprasit et al., 2022). The effect of microwave power (400, 500, and 600 W) on palm oil partial hydrogenation was investigated. Palm oil samples treated at 600 W microwave power showed lower iodine values than other samples (Wongjaikham et al., 2022). Also, Wongjaikham et al. (2021) observed that the amounts of saturated fatty acids, especially stearic acids, were enhanced by increasing microwave power from 300 W to 500 W, while the amounts of polyunsaturated fatty acids (linoleic acid and linolenic acid) were decreased by increasing microwave power from 300 W to 500 W. Also, the amount of oleic acid was slightly decreased by enhancing microwave power from 300 W to 500 W. Higher amount of microwave power provided higher plasma density and a higher level of hydrogen-free radicals to saturate the C=C bonds (Wongjaikham et al., 2021). From the above results, it can be concluded that increasing input power to a power that does not cause a sharp increase in hydrogenation temperature is helpful.

3.2.3.4. Electrode gap distance. In a DBD device, the electrode gap distance is one of the most important factors which can impact the hydrogenation performance. A large electrode gap distance needs higher energy to generate plasma (Kongprawes, Wongsawaeng, Hosemann, et al., 2021). The optimum electrode gap distance is different for DBD devices with different configurations. The optimum electrode gap distance for DBD device needle-plate and parallel-plate configurations for biodiesel hydrogenation were 1 and 3.5 cm, respectively (Kongprawes, Wongsawaeng, Ngaosuwan, et al., 2021). Kongprawes et al. (2023) reported that the gap distance of 1 cm showed a higher reduction in linoleic acid than those of 0.5 and 1.5 cm in a DBD plasma device with a parallel plate configuration. Also, Puprasit et al. (2022) reported that the gap size of 0.5 cm showed a lower iodine value than those of 0.25 and 1 cm during the production of margarine in DBD plasma with needle-in-tube geometry. A too-small gap size between electrodes can provide a high-intensity electric field but with a lower amount of plasma generation. In addition, a small gap size can form a lower intensity microfilament discharge to react with C=C bond which results in lower hydrogenation rate (Uhm, Jung, & Kim, 2003). In conclusion, a medium electrode distance is suggested for liquid oil hydrogenation. Also, the optimum electrode distance is different for each plasma device.

3.2.3.5. Reaction temperature. In the conventional hydrogenation technique, increasing the temperature can enhance the hydrogenation rate. However, in cold plasma hydrogenation technique, enhancing the temperature can have a negative impact since trans-fatty acids can form at high temperature. Hydrogenation of palm oil at 50 °C in a DBD plasma device with parallel-plate geometry was significantly faster than hydrogenation at 30, 80, and 100 °C (Puprasit et al., 2020). Also, the hydrogenation rate of biodiesel decreased by enhancing the temperature from 30 to 80 °C in a microwave plasma device (Wongjaikham et al., 2021). The higher hydrogenation rate at lower temperature was related to the exothermic nature of hydrogenation process (Sági, Holló, Varga, & Hancsók, 2017). In addition, the solubility of hydrogen gas in liquid oil is decreased at high temperature, resulting in lower hydrogenation rate (Tomoto & Kusano, 1967). Wongjaikham et al. (2021) reported that the palm oil hydrogenation rate was increased slightly by increasing the temperature from 32 to 80 °C in a microwave plasma device. However, since trans-fatty acids were formed during hydrogenation at high temperature, 32 °C was chosen as the optimum temperature for production of low trans-fatty acid margarine by microwave plasma technique. It can be concluded from the above studies that it is better to hydrogenate liquid oil at medium temperatures (30–50 °C) to minimize the formation of trans-fatty acids and enhance the hydrogenation efficiency.

3.3. Transesterification

3.3.1. Transesterification by conventional technique

Transesterification is a reaction in which the glycerol moiety of the triacylglycerol is substituted by another alcohol. Transesterification of liquid oil with alcohol is a well-known chemical reaction that can be used at large scale for producing biodiesel (Wu, Bashir, Hsieh, Krosuri, & McDonald, 2019). Methanol is the most common alcohol used for transesterification reactions. In the conventional method, the transesterification reaction can occur in the presence of heat and chemical catalysts. Acid (sulfuric acid and phosphomolybdic acid) or alkali (potassium hydroxide and sodium methoxide) catalysts are usually used for transesterification reaction (Dehghan, Golmakani, & Hosseini, 2019; Palm et al., 2022). The reaction temperature is in the range of 30–280 °C (usually 50–70 °C) (Wu, Deng, Zhu, Bashir, & Izuno, 2019). However, the conventional method is time-consuming and energy-intensive. Therefore, developing new technique to enhance transesterification rate is essential (Palm et al., 2022). To improve the transesterification rate, microwave (Nayak, Bhasin, & Nayak, 2019) and ultrasound techniques (Tan, Lim, Ong, & Pang, 2019) are applied. Although these techniques can enhance the esterification rate, they require high temperatures (Dehghan et al., 2019; Golmakani, Dehghan, & Rahimi-zad, 2022).

3.3.2. Transesterification by cold plasma technique

Transesterification by cold plasma technique usually carry out at low temperature (25–40 °C). This can be related to the production of sufficient energy for breaking chemical bonds by electric discharge generated by the cold plasma technique. The main benefit of this technique is that esters can be produced without chemical catalysts. Also, the production of unwanted side-products is limited (Kamjam et al., 2022; Palm et al., 2022; Wongjaikham et al., 2023).

The transesterification mechanism by cold plasma technique includes attacking free electrons and transferring energy in the photons to the pairs of electrons that are covalently bonded. This results in exciting and breaking the covalent bonds (Jiang et al., 2014). When methanol bonds are broken, OH, H, and CH_3 radicals are generated. The OH group bonds are weaker and break at the beginning of cold plasma treatment. The breakdown of OH bonds results in the generation of methoxide ions. The produced methoxide ions can initiate a transesterification reaction (Lee & Kim, 2013). By continuing the ionization process, excited or broken C—C bonds can adsorb free atoms. The OH group of fatty acids are replaced with hydrocarbon chain and oxygen atom of methanol and the glycerol backbone is separated from triacylglycerol molecule. The reaction pathways in transesterification by cold plasma technique is similar to that of conventional transesterification technique, except that conventional technique needs catalyst, high temperature, and stirring during transesterification reaction. Besides, the reaction time in conventional technique is higher than in cold plasma technique. In cold plasma technique, electrons with high energy can act as electrocatalyst and reduce the activation energy of transesterification reaction. However, when catalyst is not used in high voltage cold plasma techniques, controlling the transesterification reaction and producing biodiesel with high selectivity and purity is difficult. This can be due to the unpredictability of the electron performance (Istadi, Yudhistira, Anggoro, & Buchori, 2014). Thus, it is suggested to use a catalyst to control the transesterification reaction by cold plasma technique (Buchori, Istadi, & Purwanto, 2017).

Furthermore, the consumption of electrical energy in the cold plasma technique is low (Nabilla et al., 2020). Using a DBD plasma reactor, the highest biodiesel yield (88.97 %) was obtained at 40 °C, reaction time of 120 min, oil to methanol molar ratio of 1:1, and plasma voltage of 10.2 kV. Bashir, Wu, and Krosuri (2021) used a continuous liquid-phase plasma discharge device for biodiesel production. The cold plasma treatment was performed at ambient temperature for 2 min. Then, the reaction mixture was kept for six h at room temperature. The reaction conversion was measured to be 78 %. They stated that this new liquid-phase plasma discharge device can decrease the energy input and time compared to conventional technique (Bashir et al., 2021). Summary of the investigations on applying cold plasma technique for transesterifying oil for biodiesel production are presented in Table 4.

3.3.3. Factors affecting transesterification by cold plasma technique

Temperature has a critical role in the esterification reaction rate. Nabilla et al. (2020) evaluated the effect of temperature on biodiesel production in a DBD plasma device. The highest biodiesel yield (88.97 %) was observed at 40 °C. Increasing the temperature from 40 to 60 °C decreased the biodiesel yield. This decrease in biodiesel yield by increasing the temperature can be due the occurrence of chain chemical reactions which resulted in the production of undesirable products such as paraffins (Istadi et al., 2014).

The oil to alcohol molar ratio is another important parameter affecting biodiesel yield. Wu, Bashir, et al. (2019) found that the biodiesel conversion rate was enhanced by increasing the methanol to oil molar ratio from 3:1 to 6:1 (99.2 %). Also, Bashir et al. (2021) reported that the biodiesel conversion rate was increased by increasing the methanol to oil molar ratio from 4:1 to 10:1. A further increase in molar ratio from 10:1 to 12:1 did not have any significant effect on increasing biodiesel conversion rate. The increase in the biodiesel yield by enhancing the alcohol to oil molar ratio can be related to the enhancement of contact area between alcohol and oil (Lin, Hsu, & Lin, 2014). Also, Elsheikh et al. (2022) observed that the yield of transesterification of castor bean seeds via flying jet plasma enhanced from 80.5 % to 91.7 % by increasing the methanol to seed molar ratio from 9:1 to 12:1. However, further increase in molar ratio from 12:1 to 15:1 showed a negative impact on the transesterification yield. This was related to an enhancement in the biodiesel portion prone to solubilizing in the glycerol (co-product) (Elsheikh et al., 2022).

The type of acyl donor can also affect the biodiesel production by cold plasma technique. Palm, Pienta, Duarte, de Carvalho Pinto, and Catapan (2024) compared the transesterification reaction of monoesters with that of vegetable oil in a DBD plasma device with liquid-phase discharge. They observed that the transesterification of vegetable oil showed lower conversion rates than that of monoester, which was related to the lower miscibility of reactants (Palm et al., 2024).

The chemical composition of the gas used for plasma production can affect biodiesel production. In a DBD device, the biodiesel yield for argon and a mixture of argon and CO_2 (50:50) were 53 % and 29.3 %, respectively. When a mixture of argon and CO_2 was used as the carrier gas, the amounts of unwanted products such as methyl and alkoxy compounds was higher than when argon was used as carrier gas (Nabilla

Table 4
Summary of the investigations on the application of cold plasma technique for transesterifying oil for biodieseol production.

Oil type	Alcohol	Plasma device	Carrier gas	Treatment time (min)	Findings	Reference
Fresh palm oil, used palm oil + fresh palm oil (50:50), used palm oil + castor oil (50:50)	Methanol	DBD, 220 V, 30 mA*	Ar, Ar + CO_2 + H_2O	120	The cold plasma DBD reactor converted about 47–89 % mixture of triglycerides to various product such as fatty acid methyl ester, green diesel paraffin, and fatty alcohols	Nabilla et al. (2020)
Soybean oil	Methanol	Liquid-phase plasma discharge, 1.2 kV	–	15.38	Plasma technique produced biodiesel with high purity	Wu, Bashir, et al. (2019)
Soybean oil	Methanol	Continuous liquid-phase plasma discharge, 1.08–2.37 kV	–	–	The biodiesel was continuously produced at room temperature and at a higher rate than conventional method	Wu, Deng, et al. (2019)
Palm and coconut oils	Methanol	DBD plasma, 10.2 kV	Ar, Ar + CO_2 + H_2O	120	• Biodiesel was produced by plasma technique without catalyst at low temperature	Nabilla et al. (2020)
Sunflower oil	Methanol	PJ, 20 kV	Ar	0.5–1.5	• The transesterification conversion was 83 %.	Ansari Samani et al. (2023)
Sunflower oil	Methanol	DBD, 20 kV	0.42–1.25	1	• A biodiesel with high conversion (94.75 %) was produced during 1 min plasma exposure	Taki et al. (2024)

* DBD: dielectric barrier discharge; PJ: plasma jet.

et al., 2020). From the above results it can be concluded that the transesterification reaction temperature, oil to alcohol molar ratio, acyl donor type, and chemical composition of the gas used for plasma production can significantly affect transesterification rate by cold plasma technique.

3.4. Pyrolysis

Pyrolysis of vegetable oil triacylglycerols is an alternative method for the production of biofuels. Pyrolysis is the thermal degradation of organic compounds through breaking chemical bonds. In this process, oil is heated at high temperatures (300–1000 °C). Pyrolysis is one of the best methods for recovering energy because of its non-restrictive conditions, limited environmental impact, high conversion efficiency, and feedstock flexibility. In pyrolysis process, biodiesel is produced from cheap material such as byproduct of cook oil factory. Pyrolysis has several advantages compared to transesterification for biofuel production. These advantages are lower processing expenses and better compatibility with fuel standards. Besides, the chemical composition of bio-fuel produced by the pyrolysis method is similar to that of liquid fuel (e.g., diesel fuel). However, production of biofuel through pyrolysis of vegetable oils by conventional techniques needs high temperature and high energy (Lu, Li, & Zhu, 2009). In recent years, cold plasma technique is applied for vegetable oil pyrolysis and production of biofuel at room temperature. Meeprasertsagool, Watthanaphanit, Ueno, Saito, and Reubroycharoen (2017) used cold plasma technique for pyrolysis of palm oil at room temperature. Hexadecane was the major liquid product detected by gas chromatography/mass spectrometry. They stated that plasma poses enough energy to convert palm oil into biofuel at room temperature. Fan et al. (2023) used cold plasma technique to convert heavy oil into C_2H_2, H_2, and carbon nanomaterials. In the cold plasma technique, the highest heavy oil conversion was 50.4 %. The mass yields for C_2H_2 and H_2 were 19.7 % and 3.3 %, respectively. Cold plasma technique enhanced the heavy oil conversion by 12 % with above 95 % reduction in discharge power, compared to the thermal plasma technique (Fan et al., 2023).

4. Advantages and challenges of using cold plasma technique for modification of the chemical structure of liquid oils

Applying cold plasma technique for oil modification provides more benefits than conventional methods since cold plasma technique can operate at atmospheric pressure and low temperature. Also, the hydrogenation of liquid oil by cold plasma technique does not result in the formation of trans-fatty acids. *Trans*-Fatty acids can enhance low-density lipoprotein and enhance the risks of hypertension, cardiovascular diseases, Alzheimer's disease, certain cancers, diabetes, and stroke. Therefore, reducing the formation of trans-fatty acids can decrease the risk of various diseases (Meeprasertsagool et al., 2017). Low energy consumption, short treatment times, and requiring low electricity are other advantages of cold plasma technique (Coutinho et al., 2018). A comparative data has indicated that the expense of energy requires for applying cold plasma technique is equivalent to the energy requires for a light bulb (Misra & Roopesh, 2019).

One of the most important challenges of using cold plasma technique for liquid oil is the occurrence of lipid oxidation reactions during cold plasma treatment. In order to prevent lipid oxidation, optimizing plasma parameters such as input power, treatment time, and type and composition of the gas used for plasma production is necessary (Gavahian et al., 2018).

Another challenging issue of using cold plasma technique is related to the commercialization and scale-up at industrial scale (Sruthi et al., 2022). In order to scale up some cold plasma techniques such as DBD with parallel plate geometry, their configuration need to be developed to obtain higher plasma intensity for large amount of feed stock. Microwave plasma technique can be readily scaled up for industrial application. However, the equipment and devices required for plasma production and protection equipment for safety issues are expensive. Thus, microwave plasma technique needs high capital investment for scaling up in industrial scale. Accordingly, to produce hydrogenated oil and biodiesel by cold plasma technique at industrial scale, the economic issues should be considered and compared to that of conventional technique (Wongjaikham et al., 2023). Another disadvantage of using the cold plasma technique is the cost of gas used for plasma production. In order to minimize this cost, the gas used for plasma production can be recycled by a recirculating pump in a closed system (Kongprawes, Wongsawaeng, Hosemann, et al., 2021).

5. Conclusion and future perspectives

In this review, the effects of reactive species present in cold plasma were investigated on the oxidation, hydrogenation, transesterification, and pyrolysis reactions of liquid oils. Although the reactive species present in cold plasma can initiate the peroxidation process, but setting the plasma parameters at the lowest possible input power and the shortest possible duration as well as omitting oxygen from the gas used for plasma production, can inhibit peroxidation during cold plasma treatment. Also, cold plasma technique can be used as a new technique for monitoring oxidative stability of liquid oils at lower temperature and shorter time. In addition, cold plasma technique can be used to hydrogenate unsaturated fatty acids at room temperature and low pressure without forming trans fatty acids. The main application of hydrogenation by cold plasma technique in food industry is production of hydrogenated oils and margarines with low trans fatty acids. Furthermore, cold plasma technique can be used as an emerging technique for transesterification or pyrolysis processes and production of biofuel at low temperature. Although several researches have been carried out on evaluating the effect of cold plasma technique on oxidation, hydrogenation, and transesterification reactions of liquid oil, but further research is required for clarifying other aspects of cold plasma technique for oil modification. For instance, the effect of cold plasma technique on the stability of antioxidants and other bioactive compounds present in liquid oil such as squalene is not apparent. In addition, various types of cold plasma devices might have different impacts on the oxidation, hydrogenation, transesterification, and pyrolysis reactions of liquid oil. Besides, factors such as gas type, gas concentration, gas flow rate, electrode gap distance, input power, and temperature can affect oxidation, hydrogenation, transesterification, and pyrolysis reactions of liquid oils. Therefore, further research is needed to compare the effect of different cold plasma devices on these reactions and choose the best cold plasma device for each application. Also, to minimize adverse effects on these reactions, further researches are needed to clarify the plasma chemistry and reaction mechanism as well as optimizing its parameters.

Several researches have been carried out on the biodiesel production by cold plasma technique through transesterification of liquid oil with alcohols. However, other transesterification reactions of liquid oils such as transesterification reactions of liquid oil with fatty acid esters for altering the physicochemical properties of liquid oils has not yet been investigated. In order to produce confectionary fats with desirable properties, the melting behavior of the oil can be modified through transesterification reactions. More investigations are needed for applying the cold plasma technique to produce confectionary fats at ambient temperature.

Most of the studies carried out on cold plasma technique are at lab scale. More studies are required for scaling up and commercialization of oils modified by cold plasma technique. These studies should not be limited to the initial equipment needed for scaling up and commercialization, but also considering the energy and economic requirements of scaling up to commercial scale.

One of the main limitations of applying cold plasma technique for oil modification is the utilization of high amounts of gas during cold plasma treatment. In order to solve this problem, it is recommended to equip the cold plasma device with gas recycling pump. Besides, combining cold technique with other techniques such as microwave, ohmic, and ultrasound is suggested for oil modification. More studies are required to determine the long-term impact of reactive species on human health and to develop the standards for modified oils produced by cold plasma technique.

CRediT authorship contribution statement

Malihe Keramat: Writing – original draft, Visualization, Validation, Investigation, Conceptualization. **Mohammad-Taghi Golmakani:** Writing – review & editing, Visualization, Validation.

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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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Appendix A. Supplementary data

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Data availability

Data will be made available on request.

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