Glycerol In Food, Cosmetics And Pharmaceutical Industries: Basics And New Applications

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Abstract: The world demand for a greener and sustainable environment has driven researchers all over the world to put effort in utilizing all the waste into valuable products. The circular economy is aiming to reduce the amount of waste being produced to almost zero waste by reutilizing and recycling as possible as we can. Glycerol being one of the main by-product from the biodiesel production is recently gaining interest as one of the emerging renewable feedstock for various applications. This mini review starts with a brief introduction on the history and physico-chemical properties of glycerol. The state-of-the-art for different types of glycerol production is also reviewed from the perspective of conventional chemical process as well as biological process. Finally this work highlights the potential of glycerol applications in food, cosmetics and pharmaceutical industries.

Index Terms: Glycerol, biodiesel, food, cosmetics, pharmaceutical, transesterification, used cooking oil.

1. INTRODUCTION

GLYCEROL was first accidentally discovered in 1779 by a Swedish scientist named K. W. Scheele. He did a chemical reaction between olive oil and lead monoxide and discovered a water-soluble substance with a sweet taste which later was identified as glycerol. This was the first recorded chemical isolation of glycerol and initially Scheele called glycerol as the 'sweet principle of fat' [1]. In 1836, Pelouze, a Frecnch scientist proposed $C_3H_8O_3$ as the empirical formula of glycerol and in 1886, the structural formula of $C_3H_5(OH)_3$ was accepted, based on the work of two scientists named Berthelot and Lucea [2].

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Glycerol or commercially known as glycerin is a colorless, odorless and viscous liquid. Its IUPAC nomenclature is propan-1,2,3-triol showing the presence of three hydroxyl groups, which are responsible for the hygroscopic character and solubility in water. Glycerol is soluble in different polar liquids but insoluble in higher alcohols, chlorinated solvents and non-polar compounds such as fatty oils and hydrocarbons. Due to the presence of inter and intramolecular hydrogen bonds, glycerol has a high boiling point of 290 °C at ambient pressure and high viscosity of 1.412 Pa s at room temperature. The lower-purity glycerol is generally called crude glycerol. Glycerol has diverse application in different fields such as food, cosmetics, pharmaceuticals and polymer industries. In food industry, glycerol is frequently added in food to increase the water-coating ability and act as solvent for various food additives. While in cosmetics industry, glycerol is used as demulcent and anti-inflammatory agent. In pharmaceutical formulation, high purity glycerol is added as lubricant and humectant. It is also useful in production of syrups, creams and balsams. The versatility of glycerol is mainly due to the physical and chemical properties. Due to the presence of polyhydroxyl structure of the glycerol, various valuable compounds can be produced by several catalytic routes such as lactic acid, acrylic acid, dihydroxyacetone, glycerol carbonate and 1,3-propanediol [3]. In the glycerol derivatives mentioned above, the most desired platform chemicals are lactic acid and its ester [4], [5], [6]. These derivatives are extensively applied in many filed of industries.

Recently, glycerol has attained increasing attention due to the expansion of the biodiesel industries since there is an increased in the production of crude glycerol (main byproduct) during the production of biodiesel [7], [8]. In the transesterification process, for each amount of biodiesel produced, it is estimated that 10 wt% amount crude glycerol is generated. A continuous increment in the production of biodiesel will accumulate an excessive amount of glycerol and by the year of 2020, it is predicted that the global production of glycerol will be around 42 billion liters [9]. Therefore, the development of various innovative uses of crude glycerol by industry and researchers is needed in order to open new markets in the near future. However, the crude glycerol generated from biodiesel industries contains a number of impurities which decrease its economic value and limit its application. Low purity crude glycerol contains impurities such as inorganic salts, matter organic non-glycerol (MONG) and water [10]. Large-scale biodiesel producers are able to transform the crude glycerol into a refined form with purities ranging from 95.5% to 99% by using expensive processing equipment, which prohibitive for small-scale producers. Nowadays, various cost-effective techniques are available for crude glycerol purification including membrane separation [11], adsorption and combination process of chemical extraction and adsorption [12]. Although these technologies have positive and promising future, additional research and development are still needed in order to make them more economically and operationally feasible. With the advancement in the glycerol purification technology, crude glycerol can be successfully refined to a higher purity grade and used as a platform for various industrial purposes.

2 PRODUCTION OF GLYCEROL: CHEMICAL AND BIOLOGICAL ACTION

2.1 Biodiesel Production

Conventional Transesterification Process

Glycerol is generally produced as a major by-product from the chemical reaction between vegetable oil or animal fat with the addition of short chain alcohol (normally methanol or ethanol) in the presence of catalyst (normally sodium hydroxide). This process is called the transesterification process and is the principal process for biodiesel production. Figure 1 shows the general process of transesterification for biodiesel production. All biodiesel production regardless of what source of raw material it utilizes must meet with the ASTM (American Society for Testing and Materials) D6751 and European standard EN 14214 before it can be commercialized or use for other application. Glycerol produced from this process is considered as unrefined glycerol [13]. Generally, each of 100 kg of the biodiesel production will generate 10 kg of glycerol [14]. The biggest challenge for the biodiesel industries is the high cost of raw materials which is around 70-95 % from the total production cost [15]. Despite of using virgin vegetable oils or crude palm oil, scientist is exploring other sources for the raw material due to the issue raised for the concern of long-term competition between the raw material and the food supply [16]. Hence, an alternative for the high cost of raw materials has been introduced by exploring several types of inedible oil such as Jatropha curcas (Jatropha), rubber, tobacco, cotton seed and many more [16]. . The potentiality of used cooking oil (UCD) [17]. Other impurities affecting the glycerol purity from the transesterification process consist of soaps, excess methanol, fatty acid methyl esters, glycerides and water. The concentration of all the impurities is highly dependent on the type of oil feedstock used, efficiency of the overall production process and also the separation technique being used at the end of the manufacturing process [18]. In industrial production, chemical catalyst is normally been used in the process as it acts very fast and robust. However, the glycerol produced from this stream is in the unrefined version which has been a bottleneck towards further application. This unrefined glycerol has become a potential pollutant for the environment. Hence, to enable glycerol to be suitable for other applications, it must go through several purifications or refining steps to remove all the impurities affecting the glycerol yield.

Enzymatic Transesterification

Besides the general biodiesel production using chemical catalyst such as the harsh sodium hydroxide, scientists have also found enzymes to be a good substituents to eventually replace the chemical catalyst for a better and sustainable process. Selecting the right enzyme for the transesterification process is a very critical process in order to maximize the production yield without sacrificing the operation cost. The suitable type of enzyme for the enzymatic transesterification of any oils and fats is called lipase. The lipase mode of action in the enzymatic transesterification process is to catalyzes the hydrolysis of the oils and fats. The application of lipase for the transesterification process poses several benefits compared to the conventional chemical such as lower by-product and waste production as well as easier product separation [16]. However, as enzymes are known to be highly expensive in price compared to the chemicals, it must immobilized onto a support in order to enable the enzymes to be recycle and reuse. This is to avoid single use of the enzymes which can increase the overall production cost. In order to achieve high yield of glycerol, as well as the free acid methyl esters (FAME) which is normally been produced for the biodiesel production. Several parameters that can be optimized for achieving high targeted product are methanol-to-oil ratio, enzyme concentration, reaction time and reaction temperature. The enzyme recycling can also be optimized further to increase the efficiency of the overall process.

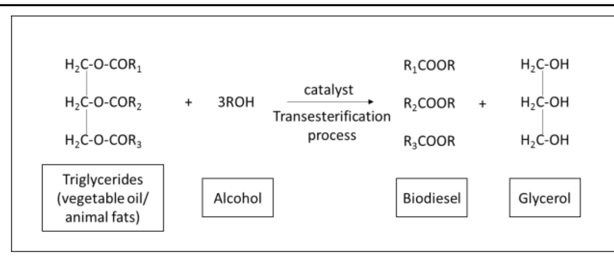

Fig. 1. *Transesterification process of triglycerides into biodiesel and glycerol*

2.2 Glycerol Residue

Glycerol may also be extracted from the glycerol residue. Glycerol residue is a residue collected after a glycerol refining process from a palm kernel oil methyl ester plant from an oleochemicals industry. From a study carried out by Ooi et al., the average content in the glycerol residue contains 20.2 % and 6.6 % of glycerol and fatty acids, respectively [19]. The glycerol content can range from as low as 8 % up to 36.3 %. Almost similar with the conventional transesterification, glycerol residue is produced from the transesterification process of the specific raw material of crude palm kernel oil with the addition of methanol as well as in the existence of catalyst. The product obtain is in heterogeneous mixture containing glycerine and methyl esters where the glycerine is a mixture consisting of methyl esters, methanol, and soap.

Chemical treatment has been proven to increase the glycerol content in the crude glycerine [20]. Several methods can be used to purify this glycerol and the level of purity can be determined by using chromatography such as the High Performance Liquid Chromatography (HPLC).

3 BASIC AND NEW APPLICATIONS OF GLYCEROL

3.1 Glycerol in Food Industries

At present, glycerol is mostly utilized as an intermediate chemical for the production of a wide range of products. Glycerol can be converted into other valuable compounds such as sweetener in foods and soft drinks which is important in food industries by conventional or fermentation process [21]. . For example, a production of a higher-value product, D-lactic acid (D-lactate) and L-lactic acid (L-lactate) which exhibits a number of applications in the food, pharmaceutical, and polymer industries, was reported from engineered strains of Escherichia coli (E. coli) under optimized conditions [22]. However, crude glycerol always associated with a problem related to impurities which make microbial fermentation challenging. Thus, a systematic engineering of E. coli was developed providing the improved strain which has the ability of homofermentative production of optically pure D-Lactate [23]. The engineered strains of Klebsiella pneumoniae also hold great promise for the efficient conversion of low-value glycerol streams, a major by-products from current biofuel industry, to a higher-valued product of optically pure D-lactate [24]. Osmophilic yeast, Candida magnolia was also capable to convert cheap raw material of glycerol into mannitol under gaseous condition [25]. Mannitol, a type of sugar alcohol, is widely used as sugar substitutes in food industry. The production of mannitol by fermentation technology is preferable due to the problems encountered with its production via chemical process [26]. The Polyalcohol, L- or D-arabitol, can be produced by yeasts via bioconversion or biotransformation of glycerol [27]. In the food industry, arabitol can be used as a natural sweetener, a texturing agent, a color stabilizer and a low-calorie sweeteners (only 0.2 kcal g^{-1}) which also act as low-glycemic, low-insulinemic, anti-cariogenic, and prebiotic agent that suitable for diabetic patients [28]. Glycerol can be converted into glycerol monolaurate that is important in food industry as a surfactant, preservative and emulsifier. Glycerol monolaurate is a product of esterification process of glycerol with lauric acid with the help or support of an acid or base catalyst [29]. In addition, glycerol monostearate is considered as a harmless and efficient emulsifier that is extensively utilized in food industry particularly in the manufacture of flour products. The addition of glycerol monostearate during the flour processing can improve the quality of the flour which can make it look and taste better [30]. Glycerol monooleate, a product of glycerolysis process of camellia oil with the aid of lipase as catalyst, was used in the production of ice cream to the desirable characteristics especially sensory and physical features, e.g., mouthfeel, overrun, firmness and melting resistance [31]. Another excellent food emulsifier agent is reported from polyglycerol ester group. Polyglycerol is a combination of glycerol oligomers produced from the reaction of metal-based catalyst for instance potassium hydroxide with glycerol at high temperature (range from 250 to 300 °C). Recently, the long-carbon fatty acid polyglycerol esters exhibited the most excellent properties as a food emulsifier compared to medium- and short-carbon fatty acid polyglycerol esters [32]. Humectants in foods are considered as additives that capable to increase water holding capacity also manage water activity. The addition of humectants to food products can increase the product stability, lessen microbial activity and protect the food texture. Glycerol is categorized as one of the most effective humectant polyols. The moisturization effect of glycerol in foods is owing to the capability of its hydroxyl groups to attach and retain water. Glycerol is an ideal ingredient in food due to several properties including nontoxic, digestible, safe to environment and provides a good flavour and pleasant odour [33]. Glycerol becomes an alternative agent in reducing water activity which elicits the similar general effects as frequently used humectants, NaCl. At the same time, the addition of glycerol in food minimizes the growth of the microbiota, including foodborne pathogens [34]. Previously, humectants have been widely employed in meat products to improve the physical properties including emulsifying ability, water binding capacity, textural and appearance quality. Glycerol has been recognized as one of the useful humectants that can improve the meat product quality by increasing the antioxidant activity and metal chelating capability hence to some extent can inhibit oxidative damage during meat processing and storage [35]. The incorporation of glycerol in jerky product made from spent hen meat exhibited better quality by showing the lowest water activity, decrease protein aggregation and the smallest shear value as compared to other humectants [36]. In addition, glycerol is generally recognized as sustenance safe (GRAS) which suggested the suitability of this sugar alcohol to be put as a humectant specifically in high-moisture diets for cats thus provide energy without compromising health [37].

3.2 Glycerol in Cosmetic Industries

The hygroscopicity and hydrophilicity nature of glycerol (by having three alcoholic hydroxyl groups) and its solubility in water makes this versatile compound a great humectant in cosmetics. Glycerol provides hydration, an important properties required for most topical application of cosmetic products. Mice model deficient in the epidermal water/glycerol transporter aquaporin-3 showed recovery in the skin hydration, elasticity and barrier function after topical application of glycerol. In other study, a skin hydration test proved a significant skin hydration and skin barrier function after 24 hours after applying hyaluronic and Centella asiatica stem cells extract moisturizing fluid in glycerol [38]. In addition, glycerol has wound healing properties, induces keratinocyte proliferation and reduces melanin intensity [39]. It also acts as skin barrier. Glycerol is reported to enhance physical texture of the skin. Glycerol is also used as humectant in toothpaste to keep the toothpaste wet and also protect gingiva and dental tissue [40]. A demand for natural butter and oil usage in cosmetics such as shea butter, jojoba oil, coconut oil etc., has been increasing lately. Formulations of these oils in water-based system for use in shampoos, conditioners and shower gels require an emulsifier. Glycerol is a great emulsifier to spread oily active substance into its liquid excipient. Body cosmetic that is applied to wetted skin for moisturizing effect is delivered as oil-in-water emulsion. Oily ingredients in this product are dispersed in the aqueous solution using water soluble glycerol. In addition to blending the oily ingredients in water, glycerol also give the effect of moisturizing to the skin and smooth feeling [41]. Glycerol is used as co-solvent in topical gel products containing oily active ingredients. Glycerol is also incorporated in water phase ingredients of sunscreen which is delivered as water-in-oil emulsion [42]. Some lipophilic bioactive components in cosmetics are encapsulated using oil-in-water nanoemulsion as a delivery system. Nanoemulsion provides better dispersion and penetration of active ingredients to be applied to body parts. The small size of nanoemulsion are sterilized using 0.2 um filters and hence could develop a preservative-free products [43]. Using this approach, glycerol has been used as co-solvent in the formation of the nanoemulsion. Glycerol modifies the interfacial properties between aqueous solution and surfactant to help forming small droplets during spontaneous emulsification. The use of glycerol has demonstrated an improved properties of vitamin E nanoemulsions in terms of reduction in droplets diameters, low surfactant level used and enhanced stability for long-term storage [44]. A food grade glycerol fatty acid ester (a glycerol derivative) is also used as emulsifier in non-fluoride toothpaste enriched with oil soluble vitamin [45]. Glycerol derivatives have also been used for various purposes in cosmetics. For example, glycol with a hydrocarbon-chain and/or of a hydroxylated ester, is ingredient in skin or lips cosmetic that reinforces their natural flesh tint to give natural volumized effect. One of the hydroxylated esters is the result of glycerol esterification with carboxylic acid [46]. In product containing skin lightening agents, glycerol ether of aliphatic alcohol is claimed to reduce the negative effects of the agents to skin, such as irritation. The presence of bisabolol and glycerol ethers of aliphatic alcohol in combination was shown to reduce the skin inflammation caused by skin active ingredients of niacinamide and N-undecylenoyl-L-phenylalanine [39].

3.3 Glycerol in Pharmaceutical Industries

Glycerol is the by-product of biodiesel industry. It has been used in various industrial applications such as in the pharmaceutical, cosmetics and food industries due to the nontoxicity and good biocompatibility [47]. The demand of glycerol in pharmaceutical and cosmetics industries increased from 26% to 34% during 1995–2006 [48]. Crude glycerol formed from trans-esterification reaction for biodiesel production can be used after several purification processes. By using the complex process and distillation, 99% purity of glycerol can be achieved [14], [49]. Glycerol assists to maintain the texture and adds humectancy, controls the water activity and prolongs the life span in a host of applications [50]. In pharmaceuticals, it provides lubrication and smoothness to many cough syrups, ointments, expectorants, anaesthetics, lozenges, gargles and elixirs. Moreover, the glycerol also used in ear infection medicines and plasticizers for medicine capsules as well as a carrier for antibiotics and antiseptics [51]. The glycerol is applied in medical grade nitroglycerin production as well, which is used to widen the blood vessels. The glycerol and its aqueous solutions have showed various promising applications as lubricants and heterogeneous catalytic reactants [52,53]. In the molecular scale, the lubrication of glycerol and chemical conversion are related with the confinement of glycerol and water molecules within the restricted nano spaces.

4 CONCLUSION

This mini review has demonstrated the high potential of glycerol to be used in various applications especially in the food, cosmetics and pharmaceutical industries. High production of glycerol can be obtained from the result of biodiesel production as biodiesel industry is a huge developing industry worldwide. Applying a good and correct separation techniques to purify the unrefined glycerol would be a cornerstone for its wider application in the future.

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