Effect of Propylene Glycol and Vegetable Glycerine Ratio in E-Liquid on Aerosol Formation: Overview of Relevant Properties

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BACKGROUND: Electronic nicotine delivery systems (ENDS) generate an aerosol by vaporising e-liquids that usually consist of propylene glycol (PG), vegetable glycerine (VG), and other ingredients (water, nicotine, and flavours). The chemical and physical properties of these components have a significant effect on aerosol formation and must be identified in order to improve product attractiveness and assess the degree of health risks. **AIM:** The aim of this article is to provide a description of the composition of the e-liquid base and its impact on the physical properties of the liquid used and the behaviour of the aerosol generated and particles separately. **METHODS:** For this purpose, 46 articles were selected using a series of keywords. English-language publications were chosen. **RESULTS:** The impact of the PG/VG ratio on the physical properties of the e-liquid (boiling point, viscosity, volatility, hygroscopicity), aerosol emission characteristics (refractive index, light scattering coefficient, particle size distribution, concentration, emission of harmful compounds), vape attractiveness (taste, "throat-hit", "cloud effect"), nicotine flux, coil temperature, and puff topography is presented. **CONCLUSIONS:** The PG/VG ratio is strongly correlated with the emission of carbonyls, which has adverse health effects and should be optimised. Furthermore, PG and VG also affect the other important characteristics of the aerosol generated by ENDS, which impact on both attractiveness and the consumption of harmful compounds. These findings could be considered for further research with the aim of improving electronic nicotine delivery systems as this can reduce levels of toxicants. This can be achieved by optimising the geometry of the components with respect to heating power and e-liquid.

Keywords | **E-Liquid – Propylene Glycol – Vegetable Glycerine – Electronic Nicotine Delivery Systems – Aerosol – Nicotine**

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1 INTRODUCTION

The popularity of electronic nicotine delivery systems (ENDS) is growing rapidly worldwide as they produce fewer harmful aerosols than conventional cigarettes do, since they do not burn tobacco and fewer chemicals are delivered to the users' bodies (Park & Choi, 2019).

The comparison between traditional tobacco cigarettes and ENDS has been explored in a plethora of studies (Glantz & Bareham, 2018; Glasser et al., 2017; Ward et al., 2020) which confirm that electronic nicotine delivery systems are popularly considered a less harmful alternative to traditional smoking, which is represented as a product with a substantial health risk. Increased attention to those systems has contributed to the rapid progress of the design of the devices. The first-generation electronic cigarettes are called "cigalikes"; they were designed like traditional cigarettes, while the second- and third-generation systems have changeable parts and their shape and design can vary widely (Keamy-Minor et al., 2019; Pepper & Brewer, 2014).

Numerous studies have been performed to evaluate the toxicological characterisation of vaping emission products, most of which remark that the chemical composition of ENDS is less harmful than traditional tobacco smoke (Belka et al., 2017; Dutra & Glantz, 2014; Son et al., 2020; Ward et al., 2020). However, vaped e-liquids can also generate substances that may lead to the formation of toxic components, especially carbonyls that are similar to those seen in conventional cigarettes (Son et al., 2020). Such observations argue in favour of continuing the study of the emission products of vape devices (El-Hellani et al., 2018, Jiang et al., 2020; Kosmider et al., 2014; Son et al., 2020). Additionally, a significant part of the findings has been focused on the consideration of differences in the impact on the human body of both ways of smoking (Park & Choi, 2019).

The emissions produced by different kinds of smoking systems depend on the design features of the devices' components, which is why the understanding of the dependence of some parameters of vape devices on others remains unclear and must be further examined. This scientific interest is clearly linked to the wide popularity of electronic nicotine delivery systems, which, in turn, raises concerns about the possibility of adverse health effects in users and environmental hazards (Marques et al., 2021). In addition, a preference for such devices is questionable because of the use of vape devices by nicotine-naive adolescents (Brett et al., 2021; Camenga et al., 2014; Dutra & Glantz, 2014).

In general, vape devices utilise a battery-powered coil to aerosolise an e-liquid that often contains nicotine and/or flavouring into an inhalable aerosol. This device is usually composed of electrical components that include a battery, chip, wire, button, and atomising components (a coil, wick, tank, and mouthpiece; Gao et al., 2021).

First, the heater coil is activated during the puff and the liquid surrounding the coil heats up and vaporises. The vapour that has been formed comes in contact with the cold air and is condensed to form an aerosol of ultrafine particles that carries nicotine deep into the lungs. Aerosol particles are rapidly absorbed, then travel through the left heart, translocating to the brain in a few seconds. They can coagulate and evaporate, changing their size and behaviour suitably (Nides et al., 2014; Yingst et al., 2019).

Certainly, the design of these components may influence aerosol formation and delivery, which is one of the key parameters in aerosol formation. Understanding the impact these characteristics have on aerosol delivery at each stage may contribute to deepening our knowledge of the public health effects of vape devices.

Another equally important parameter is the composition of the e-liquid. The e-liquid usually contains a base that includes propylene glycol (PG) or vegetable glycerine (VG; or a mixture of both) with or without the addition of flavours, water, and nicotine (Uryupin et al., 2013). Variations in the ratios of the components of the e-liquid can affect aerosol formation in different ways, particularly in terms of particle size, visible effects, nicotine flux, etc. via the different chemical and thermodynamic properties of the substances used.

Consequently, the aerosols generated from vape devices have a significant impact on the human respiratory system. This is primarily related to the difference in the absorption of particles inside the human body with various sizes as a result of the influence of gravity, inertia, and Brownian motion. Hence, small particles are affected by Brownian motion with the ability to penetrate into lungs, whereas large particles can only settle in the upper respiratory tract via gravity (Manigrasso et al., 2015). Probably, the medium-sized particles move with air flow: inhale and exhale. Gaining a further understanding of the properties of e-liquids, features of device components, thermodynamic characteristics of the system, and other relevant parameters may assist in answering questions about the assimilation of aerosol particles pathing through airways and their health effect (Oldham et al., 2018).

This review seeks to improve our understanding of the general properties of PG and VG, features of their chemical structure, and its effect on physical and chemical properties.

2 METHODS

A literature review was performed to highlight the main properties of e-liquids containing PG/VG and their effect on various features. The research process was investigated using Google Scholar as this database ensures a wide range of articles. The research criteria were a combination of the keywords "e-liquid", "vegetable glycerine", "propylene glycol", "ENDS", "aerosol", and "nicotine". Moreover, only articles written in English were chosen. In general, 46 research articles were selected so as to highlight the main properties and combine them in order to provide a broad understanding of the characteristics of the substances used. Additionally, two safety sheets were included to provide information about the physical properties of PG and VG.

Table 1 | General physical properties of glycerol and propylene glycol

Properties	Units	Glycerol	Propylene Glycol
Density	$kg \times m^{-3}$	1.261	1.036
Molecular weight	$g \times mol^{-1}$	92.09	76.09
Viscosity	Pa × s	1.49	0.0499
Boiling point	∘ C	290	187
Refractive index		1.47399	1.4329
Heat of vaporization	$kJ \times kg^{-1}$	830	711
Specific heat capacity	$J \times kg^{-1} \times C^{-1}$	2350	2481
Surface Tension	$mN \times m^{-1}$	63.4	26
Vapor pressure (20 ∘C)	kPa	< 0.00033	0.0131

3 RESULTS

An e-liquid consists of a solvent system (PG and VG) in which other additives are dissolved in various mixture ratios. The main qualities of those substances can be summarised as shown in *Table 1.* The characteristics of each component of the electronic liquids play a significant role in the formation of the aerosol; therefore, determination of the physical and chemical properties is important in studying electronic nicotine delivery systems.

Propylene glycol is a colourless, odourless, non-turbid, viscous liquid with a slightly sweet taste, the physical properties of which are introduced in *Table 1.* It is easily miscible with a range of solvents (water, acetone, chloroform, etc.; Jacob et al., 2018; Technical Product Information, 2018). PG is a widely used compound in many industries, such as the food and tobacco industry, cosmetics, chemical intermediates, pharmaceuticals, paints and coatings, etc. In general, PG is classified as a safe product according to the Food and Drug Administration but it can irritate the respiratory tract and cause allergic reactions (Kulhánek & Baptistová, 2020).

Glycerol is a clear, viscous, odourless, sweet-tasting hygroscopic liquid that can be extracted from natural oils and therefore it is called "vegetable glycerine" (Kulhánek & Baptistová, 2020). Glycerol is a chemical compound, whereas glycerine refers to commercial products that contain a percentage of water (a glycerol-water solution). The physical properties of glycerol are presented in *Table 1.* Glycerol is used in various applications, including the pharmaceutical, plastics, and tobacco and food industries, especially because of its useful solvent properties, which allow it to combine solutions with water, methanol, ethanol, and glycol (Wernke, 2014).

Glycerine and its aqueous solutions have the following essential characteristics: low saturated vapour pressure; high hygroscopicity and boiling point; a decrease in density and viscosity with an increase in temperature; less surface tension and specific heat of pure glycerine than water (Kulhánek & Baptistová, 2020; Segur, 1953).

It has been observed that glycerine is not a harmful compound. Therefore, the permissible dose of glycerol and propylene glycol in an aerosol is 10 mg/m^3 for an exposure time of eight hours, according to the Occupational Safety and Health Administration (Jacob et al., 2018; Segur, 1953).

PG and VG have different properties, the combination of which has an effect on the thermodynamic properties of the e-liquid. The PG/VG ratio as a solvent system may have a significant impact on the behaviour of the aerosol particles and their influence on taste, nicotine flux, and other key parameters that are determined by the viscosity, boiling point, volatility, and chemical structure of VG and PG. The molecular weight and viscosity of VG are greater than those of PG, meaning that the addition of VG to the base contributes to increasing the viscosity of the e-liquid (Talih et al., 2017). Therefore, the particles generated with a high VG content of the e-liquid may aggregate into large-sized particles because of their viscosity (Wu et al., 2021).

The situation related to boiling point is similar: the more VG there is in the e-liquid, the higher the boiling point is. Much attention should be paid to this because reaching boiling point by means of the coil leads to evaporation of the e-liquid and phenomena that occur during the vaporisation process and overheating cause the degradation of products, with the formation of harmful compounds (Talih et al., 2017; Wright et al., 2016).

The main difference between PG and VG is their hygroscopicity – the ability to absorb moisture from the ambient air or human respiratory system. For VG this property is expressed in the ability to accumulate or supply moisture until equilibrium with the ambient relative humidity is established. The hygroscopicity of the substances increases with the number of hydrophilic groups in their molecular structure. For that reason the hygroscopicity of VG is stronger than that of PG (the number of hydrophilic groups is three and two, respectively). The molecular structures of PG ($C_3H_8O_2$) and glycerine ($C_3H_8O_3$) are shown in *Figure 1 (a)* and *Figure 1 (b)* respectively. Hygroscopicity could lead to growing particle sizes and changing the conditions under which the experiment is performed if test samples are exposed to the air for some time (Segur, 1953; Wu et al., 2021).

Figure 1 | Molecular structure of propylene glycol *($C_3H_8O_2$)*

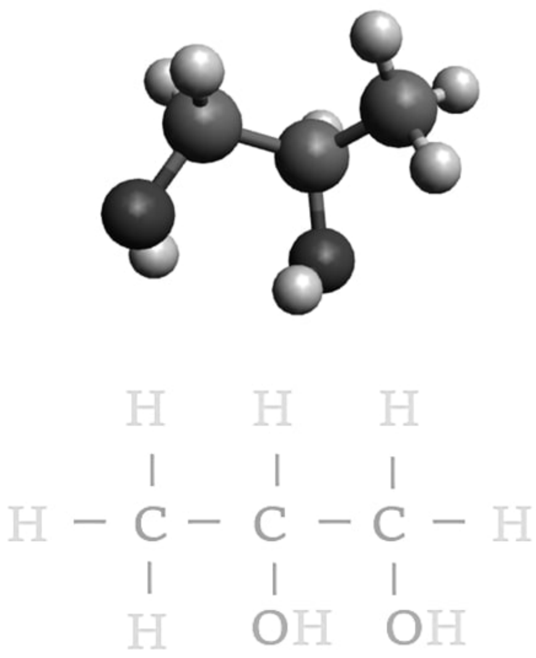
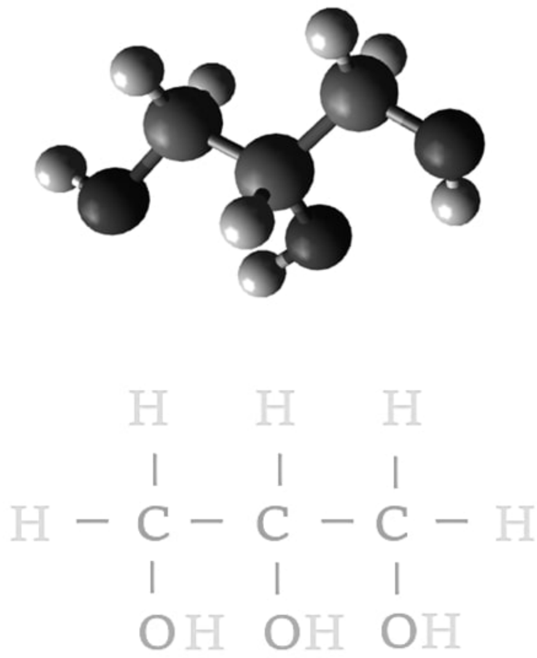


Figure 2 | Molecular structure of glycerine *($C_3H_8O_3$)*



The PG/VG ratio may affect the delivery of nicotine and actually the taste. First of all, a larger ratio of PG can cause a "throat-hit" effect associated with more nicotine delivery (Li et al., 2016). In contrast, a larger ratio of VG leads to "cloud effects". The "cloud effects" phenomenon influences product attractiveness, which, therefore, leads to consumers buying products with more artistic-looking exhaled aerosols more frequently (Brett et al., 2021). Meanwhile, the "throat-hit" is associated with satisfaction and the dependence on vaping because of the high nicotine flux in the e-liquid containing PG (Etter, 2016; Goldenson et al., 2016). This behaviour is the cause of the physical properties of the components, in particular, optical properties, particle size, and aerosol concentration.

One of the most significant characteristics that should be taken into account in aerosol particle analyses is the optical properties of the aerosol generated from the e-liquid. One of the important parameters is the refractive index, which includes real and imaginary parts reflecting the light-scattering and light absorption properties of the aerosol particles (Ingebrethsen et al., 2012; Liu et al., 2015).

Neither PG nor VG absorbs in the visible spectrum wavelength (400-780 nm), and thus they have a small imaginary part of the refractive index ($n \to 0$) (Baassiri et al., 2017). It must be noted that the real part of the refractive index is larger for VG than PG for the visible wavelength range, influencing more foggy "cloud effects" of the aerosol generated from e-liquids with a higher VG content (Wu et al., 2021). Baassiri et al. (2017) measured the refractive index of a PG/VG solution with various PG/VG ratios. They investigated the accretion of refractive index with an increasing VG content. Moreover, they explored the effect of the composition of the liquid on the light-scattering coefficient, and established that for 100:0 VG:PG this coefficient is less than in the case of a 0:100 VG/PG ratio.

In contrast, the mass concentration of the measured total particulate matter (TPM) in the filters increases with an increasing PG/VG ratio. It means that the more PG there is in the e-liquid, the more concentrated the aerosol is. In that case, it is expected that a more evident "cloud effect" will be seen, which contradicts the preliminary conclusions. The aerosol generated by an e-liquid containing more propylene glycol consists of particles smaller than that generated by an e-liquid containing more glycerol. Nevertheless, there are two explanations for that effect. Firstly, the lower vapour pressure of VG affects the slower evaporation process of particles with a high VG content, which leads to "long-lived" aerosol clouds. Secondly, an aerosol with a higher PG content has a lower light-scattering coefficient (Baassiri et al., 2017).

The reason for most of the effects is the volatility of the substances under study. The volatility of substances has been associated with vapour pressure: the low vapour pressure of VG causes less volatility. Such observations help draw the conclusion that a liquid with a greater PG ratio evaporates more rapidly than a liquid with a greater VG content. Of course, it is also affected by the enthalpy of vaporisation and specific heat but since these parameters are almost equal for PG and VG, the volatility could be only determined by vapour pressure. The main reason for this behaviour is the difference in molecular structure. The point is that VG has one more OH group than PG, which leads to stronger hydrogen bond intermolecular forces in the e-liquid solution (Li et al., 2021). E-liquid could contain water, which characteristics must also be taken into account. Water is more volatile than PG but has greater specific heat capacity than PG at a given temperature. Hence, more energy must be input to evaporate water with the same rate as PG does. Consequently, PG evaporates faster than water anyway (Baassiri et al., 2017).

The composition of the e-liquid also has an influence on the aerosol dynamics. Li et al. (2020) illustrated the correlation between the particle loss rate and PG/VG ratio. In general, the particle loss rate increases with a rise in the PG/VG ratio from 0:100 to 100:0 without nicotine content, while adding nicotine promotes the reverse behaviour. The reason for this behaviour may be the saturated vapour pressure of nicotine, which could impact on the overall e-liquid saturation vapour pressure (Li et al., 2020).

The PG/VG ratio affects the nicotine flux. The nicotine flux is the rate at which the aerosol is emitted from a vape device per unit of time (Eissenberg & Shihadeh, 2015). That parameter is used as a potential regulatory value, being an essential factor that can influence the perception of aerosol emission by users. Separately, Talih et al. (2017) previously showed that an e-liquid with a higher PG content transports more nicotine than an e-liquid with a higher VG content because of its threshold for evaporation and therefore greater volatility. Thus, PG may be called a vehicle of nicotine (Lechasseur et al., 2019). That phenomenon is related to the "throat-hit" effect that was discussed above (Baassiri et al., 2017). Spindel et al. (2018) confirmed that a pure PG liquid or 55PG:VG delivered more nicotine than an e-liquid containing VG. In that finding users took shorter puffs using devices with PG content than VG content but they obtained a higher plasma nicotine concentration (Spindle et al., 2018). Talih et al. (2017) described a transport model that clearly explains the observation of greater TPM emissions and nicotine in an e-liquid with a high PG content and therefore confirmed the assumption of greater nicotine transfer by PG (Talih et al., 2017).

Another important characteristic that is influenced by the PG/VG ratio is the particle size distribution. The particle size distribution may be measured by various techniques (Belka et al., 2017; Fuoco et al., 2014; Ingebrethsen et al., 2012; Oldham et al., 2018). Baassiri et al. (2017) showed that a PG-dominant e-liquid emits smaller particles than a VG-dominant e-liquid but, at the same time, the number of particles is greater in the first case. Identical results were obtained by Zhang et al. (2013). Pourchez et al. (2018) also found that a high-PG e-liquid generates smaller particles than a high-VG e-liquid at low wattage, while reaching 22 W leads to their convergence. Moreover, Zervas et al. (2018) noticed that the particle size distribution of propylene glycol gives a linear size distribution. In contrast, the distributions of VG and a mixture of PG and VG have some peaks that are connected to the high boiling point of glycerine compared to PG.

VG and PG undergo decomposition to molecular carbonyl compounds at high temperatures (Jiang et al., 2020; Y. Li et al., 2021). El-Hellani et al. (2018) showed that there is no correlation between the PG/VG ratio and the formation of carbonyls. But Kosmider et al. (2014) found that high-PG-level liquids generate more carbonyls than VG-based e-liquids. The levels of formaldehyde, acetaldehyde, and acetone are dramatically increased with a higher PG content for a certain range of battery output voltage. This is particularly clear for a high battery output voltage, which indicates that the level of carbonyls increases with increased battery output voltage, as it leads to a higher coil temperature (Kosmider et al., 2014). Y. Li et al. (2021) also found that some carbonyl degradation products dramatically decreased with an increasing VG fraction in the e-liquid but the fraction of acrolein rose in that case. Conklin et al. (2018) confirmed that PG generates a higher level of acetaldehyde and crotonaldehyde, whereas VG leads to higher levels of formaldehyde and the formation of the unsaturated aldehyde acrolein. Moreover, they indicated that lower levels of carbonyls were emitted by a mixture with a 25PG:75VG ratio, which indicates that some proportions of PG and VG can reduce the level of carbonyls. Such observations may be related to heat transfer, which is strongly correlated with the chemical and physical properties of the components of e-liquids discussed above (Talih et al., 2017). The emission of carbonyls is one of the key parameters as it has adverse health effects. For instance, formaldehyde and acetaldehyde are classified as potential carcinogenic substances. Acrolein has a strong irritating effect on the mucous membranes of the eyes and respiratory tract. Overall, these compounds are potentially hazardous and their consumption may be regulated by the PG/VG ratio or optimising the device's characteristics.

Thus, the thermal degradation of products is the effect of the coil temperature, which is determined by the composition of the e-liquid. The coil temperature may also be influenced by the PG/VG ratio and the addition of flavours, nicotine, and water, which could change the heat capacity, boiling point, viscosity, airflow, and other relevant characteristics. For instance, Korzun et al. (2018) noticed that the solvent consumption of an e-cigarette increases significantly with increased airflow, which could be a reason for the faster cooling effect. It may also be assumed that the delivery of the e-liquid to the heating zone occurs faster with a high PG content. In general, the characteristics of coils and their dependence on devices' parameters have been examined in a number of studies (Saleh et al., 2020; Y. Li et al., 2021; Zhao et al., 2016) but further research is still needed, especially regarding the influence of the composition of the liquid on the coil temperature.

It is also noteworthy that the PG/VG ratio was found to have an impact on the puff topography via taking larger and longer puffs using a VG-based e-liquid in order to bring more nicotine (Y. Li et al., 2021) However, the puff volume and puff duration were not controlled, thus bringing the results obtained into question. Therefore, that research study should be repeated in order to gain accurate results.

A number of the properties that were discussed above may have a huge impact on the taste experienced by the user. Undoubtedly, the PG/VG ratio determines the particle size and therefore its absorption. To be exact, the particles generated by a VG-based e-liquid are mainly large, sweet, and with the property of adhesion. On the other hand, e-liquids with a high PG ratio emit particles with a clearly "true" taste because of the small particle size, which penetrates into the alveoli.

4 CONCLUSIONS

Overall, the composition of the e-liquid is the essential parameter in vape emission characteristics. This study highlighted the features that have been determined by the VG and PG ratios and their impact on the properties of the aerosol that is formed.

The attractiveness of vape devices ("cloud effect" and "throat-hit") is influenced by the PG/VG ratio. The "throat-hit" is linked to the possibility of PG evaporating rapidly compared to VG, while the chemical structure and capacity to generate particles with a greater refractive index and light-scattering coefficient of VG leads to clear visual effects that are preferred by users.

The properties of PG and VG described in this study help to establish a link between the thermodynamic properties of the e-liquid and transport phenomena during vaping that regulate the quantity of thermal degradation products in general. On the basis of this fact, the most important field of the impact of the PG/VG ratio is the emission of carbonyls, as their action is directly related to the user's health. Formaldehyde, acetaldehyde, acrolein, and other compounds have adverse health effects such as causing damage to the lining of the lungs and irritation of the mucous membrane and can also cause cancer, etc. The regulation of the PG/VG ratio may reduce the emission of these compounds and thereby minimise the risk to users' health. The selection of the safest PG/VG ratio could be investigated with deep knowledge of the chemical and physical properties of PG, VG, and heat transfer parameters.

Further study of the dependencies of the base components of e-liquids and other device characteristics is important in research into electronic nicotine delivery systems. The attractiveness and health safety of ENDS are essential aspects of the vaping industry and both of them are regulated by the PG/VG ratio. In addition, other features play a significant role in aerosol formation and its composition, for instance, heating element characteristics. The optimisation of heating element characteristics and the composition of the e-liquid is a way of improving vaping devices.

Broadly speaking, the properties described above are determined not only by the PG/VG ratio, but also by the addition of flavours and nicotine. Consequently, studying aspects of their impact is also important in studying the emissions from vape devices.

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