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THE INFLUENCE OF DISTILLATION METHODS ON THE FLAVOR PROFILE AND QUALITY INDICES OF WINE BRANDIES

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Abstract. The article examines the geographical area delimited for the production of the Divine and the specific characteristics and physical-geographical conditions of the Călărași vineyard through the prism of different methods of distillation and production of wine distillates. Technological regimes for distilling wine raw materials and organoleptic and physico-chemical indices such as: alcohol content, optical density, pH, aldehyde content, higher alcohol content and volatile acid content are scientifically argued. The quality indicators were determined for both wines and distillates obtained at the 'Charente' batch plant and the 'VAND-M-O' plant. The impact of the distillation method on the quality and production process of the wine distillate is evaluated.

Keywords: aroma compounds, distillation methods, grape varieties, physico-chemical indices wine distillates.

1. Introduction

Technological distillation is a process by which a low proof alcoholic beverage is heated in a controlled process in order to separate ethanol from water and other components, with the ultimate goal of concentrating the ethanol into a high proof spirit [1].

The raw material used in the production of the distillate is the technological wine produced through a special technological scheme and the observance of some technological requirements included in the national document SM: 186-2014 [2]. The general process by which brandy is obtained from grapes includes the main processes such as: grape \rightarrow fermentation directly \rightarrow submerged fermentation with pure culture of yeast \rightarrow liquid-state distillation \rightarrow aging in oak barrels.

Technological, after completion of the alcoholic fermentation of the must, the wine then is placed into a still where a heat source is applied. There are two types of stills: column stills (image a) and pot stills (image b) in figure 1 [3]. Each uniquely separates ethanol from other organic compounds of wine. The separation of ethanol occurs because ethanol boils at a lower temperature than water. When heat is applied to the wine, ethanol boils before the water in solution. The ethanol vaporizes, rising to the top of the still, where it collects and then is sent to a condenser where it is quickly cooled. The water and other organic compounds are later removed from the heated unit of the still [4].

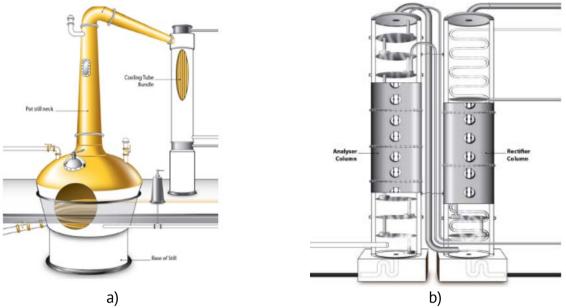


Figure 1. Appearance of winemaking distillation equipment.

Generally, aged spirits (like whisky, tequila and rum) and crafted spirits are made in pot stills since the pot stills also help add desired congeners to the spirit responsible for aromas and flavours in the spirit. Column stills are used to derive white or neutral spirits like vodka, gin and white rum.

Nationally, the geographical area delimited for the production of the Divine (Cognac alcoholic beverage called internationally) is situated in the continental area, which starts in the north between the Carpathian vineyards, extending to the Center - Codru region and continuing to the South – Bugeac steppes, including the vineyards from Călărași according to [5]. The quality, reputation and specific organoleptic characteristics of the Divine owe to the geographical environment, with its natural factors specific for the delimited geographical area, which extends over the geographical territory of the Republic of Moldova. One of the most essential factor that contributes to the ulterior production is climate of the delimited area, which ensures with the necessary conditions for ripening grapes with sunstroke of 2100 ÷ 2200 hours during the year, average annual temperature being of 10 ÷ 12°C and the sum of active temperatures being of 2900 ÷ 3100 °C. Due to different seasons, there can be noticed that the average value of the temperature in January is between -3.5 ÷ -4 °C and in July it is 20 ÷ 21 °C depending on continental climate. The climate of the Republic of Moldova is also influenced and sometimes even changed by the Carpathian Mountains (which protects from the northwestern winter winds) and the Black Sea (which buffers the summer heat in the South of the area). The presence of aquatic sources - the Dniester and Prut rivers and a large number of streams and brooks with ponds and the presence of spontaneous vegetation (essentially forests and tree strips), but also the balanced and developed relief, ensure a buffer of the extremes and respectively has a major impact on the quality and health of grapes. At last, but not least, a high diversity of soils such as gray, brown, black earth soils, typical, ordinary, tin, have a massive influence on the quality of the subsequent production and based on the geographical area, soils can be divided into two parts, first one- as soils with a great aeration and light, specific more for North and Center regions, and soils that are heavier and richer, especially for East and South regions [5].

So, the quality of grapes, raw materials wines, distillates for the divine and the Divine is guaranteed not only by the specific and largely unchanged conditions of the delimited area, but also by the human factor, which combines the techniques and equipment used during the manufacture of products. Moreover, a skilled labor, born through tradition and common learning over the decades based on a concept of standard and level of quality, is also welcome. [6].

This research set out to evaluate the distribution of volatile compounds responsible for aroma and taste in *Aligote*, *Riton*, *Bianca*, *Alb de Suruceni*, *Riesling de Rhin* and *Sauvignon Blanc* grapes distillates obtained by two types of distillation equipment (Charente and VAND types). It was also tested the impact of distillation method and final alcohol concentrations in experimental samples on the presence of undesirable compounds, such as methanol, furfural and sulfur dioxide. Assessment was focused on finding the alcohol concentration in the main fractions that had good organoleptic characteristics and the lowest possible content of compounds harmful for human health.

2. Methodology

For the presented research there were used *Aligote*, *Riton*, *Bianaca*, *Alb de Suruceni*, *Riesling de Rhin*, and *Sauvignon Blanc* grape varieties grown in the Călărași vineyards from the Republic of Moldova to obtaining experimental wine distillates used destined for obtaining the Divin beverage. The raw material wines were obtained from the harvested grape varieties and they were subjected to all the examinations necessary for the determination of the physico-chemical and organoleptic indices included in the national document SM: 312-2014 [5 - 6].

Quality indicators and general ampelographic description of the grapes used include: **Aligoté** is a grape variety used to make the wine assortment of the same name and is also used in the manufacture of wines as a raw material for distillates. The grapes are small or medium, cylindrical or cylindrical-conical, winged or slightly widened at the base. Grains: medium, sometimes deformed, light green spherical, slightly yellowish with copper shades. It is used in the production of wines raw material for distillation, quality white wines, raw wines for sparkling wines and others.

Riton is a technical variety, which has a period of late ripening (mid-September, September), with high growth force, wood maturation is good. The grape is of medium or large size, it can be conical or cylindrical-conical, compact medium, often with wings. The bean is medium, round, the skin yellow-green on the sunny side - a faint tan. The flesh is juicy, the aroma is neutral, and it has a pleasant and harmonious taste. The wine is a golden-yellow color with a greenish tinge with specific characteristics of aroma, pleasant bouquet. It is used for the production of wines raw material for distillation to obtain the divine.

Bianca is to a technical variety that has an early ripening period (early September). The storage power of sugar is high, medium to high growth force. Grapes are small, cylindrical or cylindrical-conical forms. The berries are small, round or slightly oval, with a yellow-green hue, which can stay on the bushes for a long time, withering without altering, reducing the acidity. The berries weigh in medium 1.5 gr., which can be rounded or oval, yellow-green, juicy pulp, the skin is thin. Bianca is resistant to frost and mold, can be used for the production of wines as a raw material for distillation for the divine.

Alb de Suruceni is a universal variety (both table and technical), white in color with a late ripening period. The variety can accumulate sugar very well; it represents high resistance to transportation. The growth of the logs is medium to high; the maturation of the wood is good. Characteristics of grapes include: grapes can be of medium and large size, as well as cylindrical-conical. Grains are large, round, greenish-yellow, tanned in the sun. The pulp has a harmonious taste and is juicy. The wines obtained have the color of straw, light straw with neutral, specific aromas, slightly acidified. They are intended - for the production of wines raw material for distillation, for the production of brandy, but can also be consumed fresh.

Riesling de Rhine (white) has a medium to long vegetation period. High growth vigor, with a good maturation of the wood. The tip of the shoot is fluffy white, may be greenish, with a slight pink tinge. The grapes are small and medium in size; they can be winged, reminiscent of the juice and peach, more often cylindrical, thick with a short tail. Their average weight is included of 100 and 150 gr. Characteristics of the grains: they are small, round, with a coloration that beats yellowish-golden green on the sunny side with small brown dots, with a thin and very durable pellet. The pulp is juicy, with a pleasant, specific and harmonious taste. The wines produced from these varieties are full, harmonious, firm, with a balanced taste, with a bouquet that evolves well through aging in glass. They are intended for the production of wines raw material for distillation, high quality dry white wines and also sparkling wines.

Sauvignon Blanc (white) has a medium ripening period; medium vigor to the maturation of the wood is good with the growing tip of the shoot with a scaly rosette that has a greenyellow color with reddish hues on the edge. Characteristics of the grains: they are small in size, can be rounded, with a yellowish-golden green coloration on the assorted side with small brown dots and a thin, durable pellet. The pulp is juicy with a very pleasant, specific and harmonious taste. The wine obtained shows remarkable quality, fine, with a characteristic aroma of the variety, especially the aroma of currant; some can distinguish the aroma of juice and peach, which evolves into a valuable bouquet through aging. It is a wine with a lot of life, pleasant, slightly acidic and aromatic. These variety show good qualities acquired in the area and conditions of the company "*Călărași Divin*". It is used in the production of wines raw material for distillation for divine, brandy, quality wines, sparkling and sparkling.

The quality of the must and the raw material wines intended for distillation largely depends on the quality of the grapes that are used during the process of production. The factors that influence the quality of the grapes are: the geographic area, the soil, the climatic conditions, the quality of the raw material, which can vary from year to year, from one vineyard to another. During ripening, grapes can accumulate not only sugars, but also other substances, which participate in the production and formation of divinities.

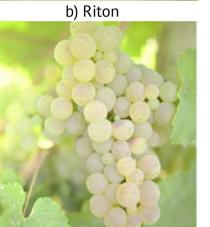
Therefore, the climatic conditions are characterized by average temperatures in the warmest month of about 20÷22 °C and the amount of precipitation is about 800 mm/years. On the other side, the best soils for cultivating grape varieties for the divine are limestone with a free subsoil layer. Soils of this type regulate moisture well, absorbing water during rainfall and conserving it in dry periods. Furthermore, in order to determine the physico-chemical indices and the organoleptic properties of the obtained wines, analysis methods were used according to the standards in force, as well as those recommended by the OIVV reglementation.

Distillation was performed on discontinuous installation (Charente type) and on continuous-run VAND-M-01 installations and the characteristics are included in table 1 according to [7].



a) Aligote







c) Bianca



d) Alb de Suruceni	e) Riesling de Rhin	f) Sauvignon Blanc	
Figure 2. Characteristics of experimental grape varieties.			

Table 1

Distillation methods studied				
Method	Description of the method			
Double distillation method with stills of Charente type	Represent a pot still, also called alembic still. It is typically made from copper. Copper was used traditionally because it is a good conductor of heat and it extracts sulphur from the distillate. The bottom part is called kettle where the ferment is heated. The ferment is evaporated and the vapors travel through the swan-shaped neck and it reaches the worm, a spiral-shaped copper tube attached to the condenser where the vapour is condensed to a liquid. Distillation takes place in two stages. In the first stage, the simple distillation is done, when the total alcohol content is extracted from the wine, and in the second stage, the fractional distillation, when it has their separation of "heads", "hearts" and "tails" fractions. At the company " <i>Călărași Divin</i> " the Charente type equipment of a capacity 400 liters, double distillation is usually performed. Compared to other methods that provide (thermal agents or direct fire), after which we obtain unsatisfactory results, distillation performed in copper boilers, is the most efficient.			

Continuation Table 1

The continuous distillation system uses the principle that the distillation plant must be supplied with raw material continuously, but also takes into account the uninterrupted separation of different fractions coming from more or less slightly volatile. Column stills are made from stainless steel and comprises a tall column structure attached above the top of the boiling kettle designed to attain purer Continuous vapours. It is divided into chambers using perforated plates. The still is distillation constantly heated at the bottom, so when the ferment is poured from method at the the cooler top part and when it hits the steam, it vaporizes and sends VAND-M-01 the alcohol back to the top. When the alcohol carrying vapour passes through the perforated plates, it condenses the heavier particles and only alcohol carrying vapours continues to the top, travels through the pipes attached to the condensers where the vapours are condensed to the desired liquid. Installations with continuous operation, such as Vand - M-01 ensure the obtaining of the middle distillate directly from the wine, avoiding the stages of formation of intermediate products.

Sensory assessment of the wine distillates was performed using the ONV model of positive ranking and all the samples were diluted before sensory analysis to 40 % vol. All experimental samples were prepared and analyzed in triplicate. Results obtained were expressed as average ± SD.

3. Results and discussion

In order to determine the time of harvest, it is necessary to monitoring the progress of grape ripening by quantitatively modifying the sugars and acids in the average sample. Tables 2 and 3 show the main indications of must and raw material wines for distillation.

Table 2

Main physico-chemical indices of the analyzed grape varieties				
Ampelographic variety	Mass concentration of sugars expressed in reducing sugar, g/dm ³	Mass concentration of titratable acidity, expressed in tartaric acid, g/dm ³	Permissible sugar standard, g/dm³	Permissible norm of mass concentration of titratable acidity,expressed in tartaric acid g/dm ³
Aligote	188 ± 2	7.2 ± 0.55		
Sauvignon	196 ± 4	7.4 ± 0.55		
Bianca	197 ± 6	7.3 ± 0.78	170 ÷ 190	4 ÷ 8
Riesling	191 ± 3	6.9 ± 0.55		
Riton	189 ± 5	7.0 ± 0.68		

The characteristics of the raw material wines intended for distillation are determined by a specific aroma, acidity, alcohol, tannins and other factors that must be taken into account in the manufacture of the divine. Dhysical chamical indicas of raw material wines intended for distillation

Table 3

Physico-chemical indices of raw material wines intended for distillation					
Type of wine raw material	Alcohol content, * % vol.	Mass concentration of sugars expressed in reducing sugar,** g/dm ³	рН***	Mass concentration s1/4 of titrable acides, expressed in tartaric acid **** g/dm ³	Mass concentration of volatile acids, expressed in acetic acid,***** g/dm ³
Aligote	11.36 ± 0.42	Max4	3.3 ± 0.3	7.11 ± 0.82	0.40 ± 0.17
Sauvignon	11.70 ± 0.25	Max 4	3.2 ± 0.3	7.20 ± 0.55	0.50 ± 0.13
Bianca	10.80 ± 0.25	Max4	3.3 ± 0.2	7.03 ± 0.80	0.53 ± 0.15
Riesling	10.90 ± 0.25	Max 4	3.2 ± 0.6	6.83 ± 0.68	0.48 ± 0.08
Riton	11.20 ± 0.30	Max4	3.3 ± 0.3	6.90 ± 0.55	0.47 ± 0.12

* $p \le 0,05$ According to OIV rules, OIV – MA – AS312-01A;

** $p \le 0,1$ According to OIV rules, OIV – MA – AS311-01; *** $p \le 0,05$ According to OIV rules, OIV – MA – AS313-15;

**** $p \le 0.05$ According to OIV rules, OIV - MA - AS313-01;

***** $p \le 0,05$ According to OIV rules, OIV - MA -MD-AS313-02ACIVOL.

The raw material wines intended for distillation were identified as light wines, with low extract, moderately alcoholic and acidic, with a subtle neutral aroma and floral fruits with a minimal presence of sulfur dioxide.

The aroma of the wines obtained is a primary one acquired from grapes, but it is also a secondary one, which was formed during the fermentation of the must. Essential oils are the basis of primary aromatic substances, the main ones being terpenoids, high-boiling aliphatic alcohols and their esters. This group of compounds is responsible for the so-called floral-fruit aromas.

The basis of secondary aromatic substances is alcohol and their esters, which form the basis of the aroma of wine. During fermentation, it is necessary to mention that aromatic alcohol with high boiling and their esters are also formed, thus enhancing the floral nuances of the aroma. After obtaining the raw material wines intended for the production of divines, they were circulated for distillation.

The quality of distillates for the divine is directly dependent on the technological schemes applied to wine distillation.

For this purpose, the methods of distilling raw material wines intended for the production of divines were experimented, research was carried out, which refers to both technological and economic aspects.

Although the raw material was qualitative and the regimes and parameters of the wines were observed, the quality of the distillates obtained was different due to the method of distilling the raw material wines. The grapes, which were harvested, were transported to the company by means of transport by trailers [8].

The objective of the study was to evaluate the influence of raw material distillation methods on obtaining wine distillates from grape varieties such as: *Aligote, Sauvignon, Riton, Bianca*, others at the *Charente* and *VAND-M-01* installations.

Tables 4 and 5 indicate the organoleptic indices and physico-chemical indices of wine distillates obtained by different distillation methods.

Table 4

Org	anoleptic indices of distill	ates obtained by 2	methods of distilla	ition
Distillatio	n Clarity	Color	Flavor	Ta

Distillation methods	Clarity	Color	Flavor	Taste
Characteristic of wine distillates according to the Specifications for Divine	Clear liquid, without foreign inclusions	colorless to light straw	Clean of wine, with light shades of flowers	Clean, slightly burning
Distillation method with batch operation at the type <i>Charente</i> installation	Clear liquid, without sediment and suspended particles	Colorless, without foreign nuances	Clean of wine, with light shades of flowers. The intensity of the aroma is stronger, more floral.	Clean, not stinging, soft, sweet taste
Distillation method with continuous operation at the <i>VAND-M-01</i> installation	Clear liquid, without sediment and suspended particles	Colorless, with slight yellowish hues	Clear of wine, with aroma of aromatic aldehydes, pure wine, burner, slightly sour taste	Clear wine, burner, slightly sour taste

It is important to emphasize that the boiling point of compounds is not the sole criterion for their separation during distillation. The separation of compounds is strongly influenced by their solubility in alcohol and water mixture as well as by the distillation equipment employed. For instance, if the alembic still was used, the higher alcohol (2-methyl-1-butanol, 3-methyl -1-butanol, 2-methyl-1-propanol) content was higher in the hearts than in the tails, despite their having boiling points superior at 100°C. In turn, the solubility of the methanol in water, owing to its capacity to form hydrogen bridges, ensures that it is present in all fractions (often in the greatest concentration in tails), regardless of its lower boiling point [9].

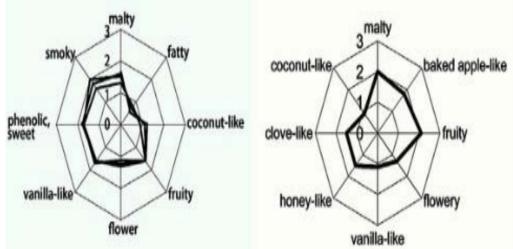


Figure 3. Spider diagrams of organoleptic characteristics in 2 experimental distillate samples.

Volatile compounds of wine distillates may originate from raw materials, as well as being formed as by-products during alcoholic fermentation and distillation. Together with compounds responsible for aroma, fruit distillates contain undesirable compounds, such as methanol, furfural and SO₂, which can have adverse effects on human health [10].

The testers detected in first samples a sharp, acrid, solvent, pungent aroma and taste when compared with the second samples as a consequence lower concentration of volatiles. On the other hand, all the heart fractions were assessed as sharp and burning, owing to the predominance of ethyl alcohol, which excessively reduced the concentrations of aroma components. Esters contribute to the pleasant fruity aroma of fruit distillates with the main ester produced by yeast during fermentation is ethyl acetate. At low concentrations (up to 200 mg/dm³) ethyl acetate has a floral and fruity aroma. At higher concentrations it has a negative effect on the sensory quality of spirits, and its presence is related to acetic bacterial spoilage [9]. Numbers from 0 to 3 in the spider diagrams of 2 wine distillates mean odour intensity from not perceivable to strongly perceivable; and from 0 to 5 in these spider diagram liquor mean odour intensity from not perceivable to strongly perceivable.

Table 5

Physico - chemical indices of wine raw material	Charente Method	VAND-M-01 Method	Permissible limits after SM 312: 2014 Wine distillate [6].
Ethyl alcohol, % vol, at 20 °C,	64.50 ± 0,55	76.10 ± 0.19	52.0 ÷ 86.0
Mass concentration of higher alcohols, g / dm ³ anhydrous alcohol	3.100 ± 0.097	2.78 ± 0.06	1.0 ÷ 6,0
Mass concentration of aldehydes, expressed as acetaldehyde, g/dm ³ alc. anhydrous	0.110 ± 0.079	0.050 ± 0.027	0.03 ÷ 0.5
Mass concentration of average esters, expressed as ethyl acetate, g/dm ³	0.85 ± 0.35	1.360 ± 0.097	0.2 ÷ 2.5
Mass concentration of volatile acids, expressed in acetic acid, g/dm ³ , anhydrous alcohol	0.111 ±0.019	0.720 ± 0.079	0.02 ÷ 0.8
Mass concentration of furfural, mg/dm ³ anhydrous alcohol	0.500 ±0.079	0.20 ± 0.06	Max 3.0
Mass concentration of methyl alcohol, g/dm ³	0.31 ±0.06	0.230 ± 0.079	Max 2.0
Mass concentration of total sulfur dioxide, mg/dm ³	4.031 ± 0.612	4.021 ± 0.612	Max 45

Physico - chemical indices of distillates obtained by Charente and VAND-M-01 methods

According to the literature [8 and 10], head compounds typically include aliphatic aldehydes and esters (ethyl acetate, isoamyl acetate, methyl acetate), because of their low boiling points and high solubility in ethanol. Of the aldehydes in the head fractions, acetaldehyde occurred in the largest concentrations, regardless of the distillation variant

used (*Charante* and *VAND-M-01*). Acetaldehyde can give beverages a fruity character when present in low concentrations, but in higher concentrations it causes a pungent smell, producing poor-quality distillates.

Interestingly, the concentrations of acetaldehyde in the hearts fractions obtained from two-stage distillation Charante were approximately double those obtained following the VAND installation (p < 0.05). This fraction also contained large quantities of other aldehydes, such as isobutyraldehyde, isovaleraldehyde and hexanal, as well as acetaldehyde diethyl acetal. This can be explained by the fact that, when a second distillation is performed, part of the water in the wash has already been removed by the first distillation, which increases the concentrations of ethanol and volatile secondary compounds in the second distillate. Single-stage distillation resulted in the separation of head fractions with acetaldehyde concentrations ~50% lower than those in head fractions separated following two-stage distillation [10].

Another compound as furfural showed a different distribution from that of the aliphatic aldehydes. The highest concentrations were measured in the hearts fractions, especially in those obtained after two-stage distillation *Charante* (p < 0.05) with 0.3 g/dm³ superior of VAND equipment. Those results for the distribution of furfural following distillation are in agreement with those reported in the literature [8, 10]. Varying concentrations of furfural in spirit beverages can result from different distillation conditions and equipment. Furfural is generated by the dehydration of pentoses, and occurs during distillation involving Maillard reactions. Its constant increase was probably due to furfural synthesis occurring in the heated pot still [11].

The esters of acetic acid and alcohols such as propyl acetate, butyl acetate and hexyl acetate occur in fresh plums. Acetate esters of higher alcohols are significant aroma compounds in alcoholic beverages [12 - 14]. These compounds can give a pleasant fruity fragrance to the general aroma of fruit and wine distillates. In this study esters were separated mainly in the head fractions. As a consequence, the concentrations of these compounds were much lower in the heart fractions (see Table 5). Of the esters in the heart fractions, ethyl acetate was present in the highest concentrations, regardless of the VAND distillation method used (p > 0.05).

A compound harmful to human health as methanol is subject to restrictions owing to its high toxicity. Given the low boiling point of methanol (64.7 °C) and its high volatility, it might be assumed that the highest concentrations of this compound would be present in the head fractions. Methanol should be distilled mainly at the beginning of the process, and its concentration may be expected to decrease steadily along the distillation path and not exceed au maximum the value 2 g/dm³. The heart fractions obtained by booth equipments contained low contents (p < 0.05) of methanol ≤ 0.3 g/dm³ and total sulfur dioxide ≤ 4 mg/dm³ fact which attests the inoffensiveness and quality of the obtained experimental distillates during the study.

4. Conclusion

The eco-climatic conditions of the Călărași vineyards are favorable for the cultivation of wine varieties such as *Aligote, Riton, Bianca, Sauvignon* and *Riesling de Rhin* and *Sauvignon Blanc* that allow to produce the raw material wines that are essential in order to produce divines with the corresponding and necessary sugars and titratable acidity concentrate.

The physical-chemical indices of the raw material wines destined for distillation fit within the acceptable limits established by the technical conditions. Therefore, in order to improve the quality of the obtained wine distillates, it is essential to make an appropriate choice of the distillation methods and working parameters that need to be used during the wine distillate production. As a consequence, the quality of the wine distillate using the *Charente* method was identified as being of a higher level than those produced by using VAND-M-01. This result is owing to the fact that by utilizing the VAND-M-01 method, the volatile acidity increases approximately by 6 times and produces wine distillate with a slightly quality downgrade. So, the number of aldehydes obtained by distillation with ordinary installations was 0.11 g/dm³, and in a continuous flow 0.05 g/dm³. The reasons for such experimental data are due to the fact that the distillation process in the first case took longer than was expected and some of the alcohol oxidized due to aldehydes. Moreover, it was established that the higher alcohol content in the distillates that were initially obtained by both methods Charente (3.10 g/dm³) and VAND-M-01 (2.78 g/dm³) correspond to the acceptable limits between 1 and 6 g/dm³).

The results of this study indicate that both method of distillation can be used to improve the chemical and organoleptic qualities of wine distillates. The composition of volatile components in the heart fractions of wine distillates varied greatly depending on the distillation methodology used, on the separation of the head fractions from the heart and on the final concentration of alcohol in the heart fractions. Increases in the alcohol by volume strength of the heart fractions (from 65 to 75% vol.) resulted in a gradual reduction in the concentrations of volatile compounds such as aldehydes, esters and higher alcohols, as well as in lower levels of methanol, furfural, anhydrous alcohol and acetaldehyde.

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