

Article

Study of Ultrasound-Assisted Technology for Accelerating the Aging Process in a Sugar Cane Honey Spirit

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Abstract: This study aims to obtain an alternative aging method using toasted white oak chips and ultrasound technology that yields a final product of similar quality to the one obtained by a traditional aging system in reduced time. Different conditions of ultrasound treatment and ethanol concentration during the maturation stage were studied. A sugar cane honey spirit was produced. The ultrasound treatments were applied to the distilled product to extract the color, aroma, and flavor compounds from the white oak chips used. Trials of spectrophotometry-evaluated color and e-sensing technology were applied to assess flavor and aroma. Very distinct color changes were obtained, indicating that ultrasound treatment facilitates the extraction of color compounds from the oak chips. The flavor profile obtained was similar to the one obtained for the unaged reference, indicating that the accelerated aging treatment may not influence flavor in a significant manner. The aroma profile achieved most descriptors found in the commercial rum aroma profile, indicating that the aging method studied influences the aroma profile. In general, the methods used allowed us to produce an aged spirit, offering a reduction in maturation time over the traditional system and a similar sensory profile for the final product.

Keywords: accelerated aging; ultrasound; oak chips; sensory analysis



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1. Introduction

Within the spirits industry, aging distilled products is crucial in improving the sensory profile of alcoholic beverages such as rum, brandy, whisky, and tequila. Aging is a common practice and has been studied for centuries [1]. Various factors influence the sensory profile of these products during the production process, such as the raw material, fermentation process, distillation operation, equipment used, and aging [2]. The spirit extracts flavor and aroma molecules from the wood barrel during the latter stage. The main compounds are phenolic acids and aldehydes such as vanillin, syringaldehyde, coniferyl aldehyde, gallic acid, and vanillic acid [3]. The concentration of these compounds is higher for samples matured for extended periods [4]. Various factors affect the final spirit's phenolic content, including the nature of the wood used in the aging process (region of origin, age, species) and charring in barrel production [2]. Oak species (*Q. alba* L., *Q. robur* L., *Q. petraea* (Matt.) L.) are commonly used. Nevertheless, species selection is often influenced by the region where the beverage is processed [4].

Traditional aging requires long periods and large storage areas, increasing the final product's price [5]. To develop new products or improve the characteristics of the existing ones, techniques to reduce the aging time while enhancing the organoleptic properties of the final product have recently been studied. A promising method, highlighted by its simplicity and effectiveness, is increasing the surface area of interaction between the wood and the spirit. This method involves the application of oak fragments, such as chips, cubes, staves, or powders. Several authors have reported similar organoleptic results compared to traditional aging while reducing the contact time [2,6]. Other techniques

studied to accelerate the aging process include micro-oxygenation, underwater aging, and the application of ultrasound. Micro-oxygenation consists of the controlled introduction of small oxygen dosages to the spirit. A positive effect for color and phenolic evolution was reported for this method [7]. A study was conducted for the underwater aging of agricultural rum from Madeira, where 14 months of maturation on the seafloor yielded positive results on the sensory profile of the final product [8]. Ultrasound energy has been studied to accelerate the extraction of compounds from the wood during the aging process [2,9]. Most studies have focused on the aging of wine spirits [10]. However, a study reported that the combination of ultrasound waves and oak chips could produce brandy with similar organoleptic characteristics compared to the product of traditional maturation [11]. Additionally, a comparable phenolic content and higher color intensity were reported for sugar cane spirits aged by application of ultrasound and oak chips compared to the traditional method [9].

Ultrasound technology has been widely used to enhance extraction processes, mainly through the cavitation phenomenon [12]. When high-energy ultrasound is applied to a liquid with elastic properties, the molecules expand and bubbles are formed. These cavities can explosively collapse, generating localized pressures to alter organic tissue, favoring the extraction of bioactive compounds [13]. In the food industry, power ultrasound (16–100 kHz) can generate emulsions, disrupt cells, and disperse aggregated materials [14]. The effectiveness of frequencies of 0.02–10 MHz has been studied for various purposes, including degassing and oxygen removal from products [15]. An ultrasound treatment with a frequency of 0.02 MHz showed potential as an excellent alternative method for aging a rice alcoholic beverage [16]. High-power and low-frequency ultrasound with wood fragments has been studied for spirit maturation, showing an improved extraction of phenolics [17]. Ultrasound enhanced the extraction of oak chips in sherry vinegar using micro-oxygenation. Successive cycles of the operation and stand-by time of ultrasound application have also been studied and proved successful [9,18]. Dosages of oak chips ranging from 3.5 to 7 g/L have been tested on various distillate beverages, showing positive effects on the final product, such as improved color and increased concentrations of phenolic compounds [17]. The duration and intervals of ultrasound application vary among previous studies. Ultrasound has been tested to enhance the extraction of phenolic compounds for optimizing the maceration process in winemaking [19]. This technology has also been studied for successful applications within the food industry and development [20].

This study aims to obtain an alternative aging method using toasted white oak chips (*Quercus alba*) and ultrasound technology that yields a final product of similar quality to the one produced by the traditional aging system. The developed method should offer advantages regarding reducing maturation time over the conventional system, offering a similar sensory profile for the final product. For this purpose, laboratory experiments were conducted under different ultrasound durations and ethanol concentrations through the maturation stage. The results were compared analytically and sensorily to reference rums that were aged using the traditional system. Various commercially available rums were studied, analyzing parameters comparable to the sugar cane spirit obtained in this study.

2. Materials and Methods

2.1. Sugar Cane Spirit

The sugar cane spirit was produced using the industrial process overview shown in Figure 1. Briefly, 3.5 L of 1.355 specific gravity (SG) sugar cane honey was diluted with 8.5 L water to 1.096 SG. The pH of the mash was adjusted to 4.5 with tartaric acid. Then, 6 g of Distilamax yeast (*Saccharomyces cerevisiae*, Lallemand Biofuels & Distilled Spirits, Montreal, QC, Canada) was activated with 6 g of yeast nutrient (Springferm NAB-3 from Fermentis) and added to the mash. The fermentation process was conducted at 30 °C for 15 days, with a reinoculation of yeast and nutrients on day 7. Acidity, temperature, and specific gravity (SG) were monitored during fermentation. The fermented mash was then distilled to obtain a concentrated ethanol spirit. Heads and tails were discarded.

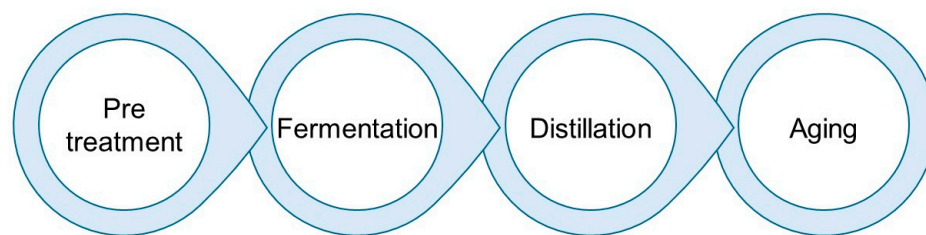


Figure 1. Sugar cane spirit production outline scheme.

2.2. Accelerated Aging Method

A laboratory-scale method for maturation accelerated by static ultrasound-assisted extraction of the sugar cane honey spirit was used. The trials were carried out in 60 mL amber glass vessels with the sugar cane spirit and oak wood chips (*Quercus alba*, Jack Daniels, Lynchburg, TN, USA). The experimental design included the duration of the ultrasound treatments, whether the method included successive cycles of operation and stand-by time, and the alcohol by volume (ABV) during the aging process. The treatments for all samples were repeated every 24 h for 5 consecutive days. The design, including the levels of operation for all samples, is presented in Table 1. All the measurements were performed in triplicate. The operation conditions were selected according to the successful reports of previous studies. The dosage of oak chips was constant for all samples at 6 g/L of distillate. The ultrasound application was performed with an ultrasound bath of 40 kHz (Branson 2510, Brookfield, CT, USA).

Table 1. Experimental design to evaluate optimal conditions for the accelerated maturation process.

Samples	Factors		
	Alcohol by Volume (ABV)	Ultrasound Duration (min)	Successive Cycles (Yes/No)
1	60	10	Yes
2	60	20	Yes
3	60	10	No
4	60	20	No
5	70	10	Yes
6	70	20	Yes
7	70	10	No
8	70	20	No

2.2.1. Alcohol Content

The alcohol content in the samples was monitored after each treatment to consider potential ethanol losses due to the application of this technology. Additionally, ultrasound treatment durations of up to 60 min were studied to analyze the possible loss of ethanol due to volatilization.

2.2.2. Titratable Acidity

The titratable acidity was represented as acetic acid due to its domination in sugar cane spirits [21]. The titratable acidity was measured using 0.1 N NaOH by the Association of Official Agricultural Chemists (AOAC) method with slight modifications [16]. The titratable acidities and pH values of an unaged control sample and of all accelerated aging treatments were measured. This gave an indication of acidity and the level of sour taste of the alcoholic beverage; it was also an index of possible contamination [21].

2.2.3. Color Determination

After the accelerated aging treatment, the color of the aged spirit samples was quantified via visible spectrophotometry. The absorbance of the matured samples was measured using a spectrophotometer (Thermo Scientific GENESYS 10S Series, Waltham, MA, USA)

at wavelengths of 450, 520, 570, and 630 nm. The CIELab method was used to obtain the colorimetric coordinates. This method is widely recognized as one of the most accurate methods for color definition in the food industry. Moreover, it is commonly used to analyze aged spirits' chromatic characteristics [8,9]. The Research Color Group developed the MSCV_7 software at the University of La Rioja to determine the color of wines and brandies using four absorbance values. This software was used to determine the color of the CIELab coordinates. These parameters are L^* (lightness), a^* (CIE red(+)/green(−) color attribute), and b^* (CIE yellow(+)/blue(−) color attribute). Furthermore, the total color difference (ΔE) was used as the reference method to quantify the color changes in the aged samples. Equation (1) was used to calculate ΔE .

$$\Delta E = \sqrt{(L^* - L_0)^2 + (a^* - a_0)^2 + (b^* - b_0)^2}, \quad (1)$$

where L_0 , a_0 , and b_0 were the reference values of unaged sugar cane spirit and L^* , a^* , and b^* are the color parameters of the samples after the extraction process.

2.2.4. Electronic Tongue Evaluation

Samples were diluted to 5% ABV and assessed using an electronic tongue system (TS-5000Z, INSENT, Atsugi, OL, Japan). The system incorporated an array of five basic chemical sensors, including AAE (umami), CT0 (salty), CA0 (sour), C00 (bitter), and AE1 (astringent). An array of three aftertastes was also included in the system (aftertaste-A for astringency, aftertaste-B for bitterness, and richness for umami). Firstly, the primary taste was measured as the potential difference between the sample and the reference set to zero. Successively, the sensors were rinsed in the reference solution and the aftertaste was calculated as the potential difference compared to the reference solution. The reference solution consisted of 30 mM potassium chloride solution and 0.3 mM tartaric acid solution. For each measurement, 35 mL of diluted spirit was placed into a dedicated beaker and the sample was continuously monitored at 20 °C. The reference sample used was food-grade ethanol diluted to 5% ABV. Each sample was measured four times; the average data were applied for further analysis. The obtained data were standardized against the reference solution. The electronic tongue test simulated the state of the human mouth with only saliva present.

2.2.5. Electronic Nose Evaluation

The aroma profile of the samples was performed using an electronic nose system (PEN3, Airsense Analytics GmbH, Schwerin, MV, Germany). The system comprises a sampling unit and a gas detection system comprising 10 Metal Oxide Semiconductor (MOS) sensors. Each sensor is sensitive to a characteristic volatile compound, referred to in [22]. Table 2 shows the sensors and their corresponding target substances. For preparation, 10 μ L of the diluted samples (5% ABV) was dispensed into a 20 mL glass vial and capped with a Polytetrafluoroethylene (PTFE) septum. Each vial was incubated at 20 °C for 24 h to reach the headspace equilibrium. Subsequently, the electronic nose system injected filtered air for 120 s to calibrate the sensors. Afterward, the headspace of each vial was injected into the system for 50 s; the sensor signals were recorded at each second. The average response of periods between the 40 and 42 s values, which exhibited a stable response curve, was considered for sample detection and used for further analysis.

Table 2. Electronic nose PEN3 sensors and their target substances. Adopted from [22].

Sensors	Target Substances
W1C	Aromatic compounds
W5S	Nitrogen oxides
W3C	Ammonia and aromatic compounds
W6S	Hydrogen
W5C	Hydrocarbons, aromatic compounds
W1S	Methane in the environment, with a broad range
W1W	Sulfur compounds, pyrazine, many terpenes (i.e., limonene)
W2S	Ethanol, some aromatic compounds, broad range
W2W	Aromatic components, sulfur compounds
W3S	Methane and some high-concentration compounds

3. Results

3.1. Sugar Cane Spirit

Acidity, temperature, and specific gravity (SG) were continuously monitored during fermentation. Figure 2 shows the mash's profiles of acidity, temperature, and specific gravity (SG) during the nine days of fermentation.

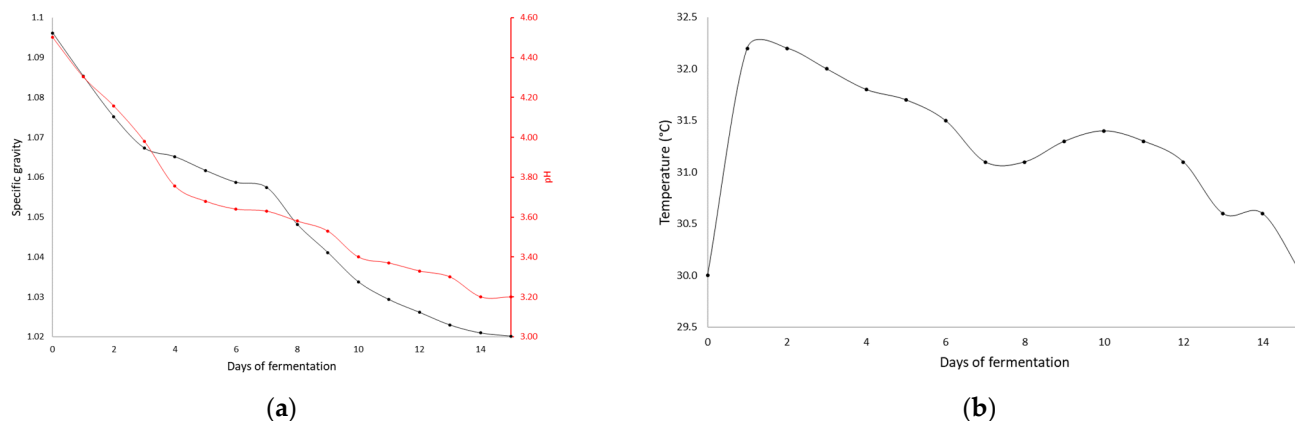


Figure 2. Specific gravity and acidity profiles (a) and temperature profile (b) of a sugar cane honey mash fermentation.

To obtain the initial specific gravity (SG) of the sugarcane honey at 23 °Bx, the formulas presented in Table A1 were used. During fermentation, the specific gravity decreased from 1.096 to 1.020, representing an ethanol concentration of about 10.99% ABV. Acidity was adjusted to an initial pH of 4.5 with tartaric acid to favor the performance of the yeast, as this is the optimal range for fermentation [21]. The mash became more acidic over time, reaching a pH of 3.2 on the final day of fermentation. Figure 2 shows that the specific gravity decreases over time. The main components of the mash used are water and soluble solids. Thus, the latter, primarily fermentable sugars, is responsible for a specific gravity value higher than one. In concordance with this, the sugar content in the mash decreases over time, indicating glucose utilization (it is likely converted into alcohol and CO₂) [21]. However, the rate of sugar consumption decreases as fermentation progresses. The rate of sugar consumption is higher in the initial days after each yeast inoculation, decreasing over time. Moreover, the specific gravity and pH decrease rapidly at the beginning of the process, and later, their values have minor changes per day. Likewise, the temperature profile shows an increase of 2.2 °C, reaching 32.2 °C on the first day of fermentation. From that point forward, the temperature decreases at a lower rate, reaching 30 °C in the remaining 14 days of fermentation. The increment in temperature can be understood as the fermentation process consisting of an exothermic reaction. The three phases of fermentation can explain the acidity, specific gravity, and temperature profiles. The initial phase is yeast

multiplication, which requires oxygen. During the primary phase, sugars are converted into alcohol and CO₂, forming bubbles in the mash. In the final phase, the CO₂ release, the formation of bubbles, and the increase in temperature stop [21]. During distillation, the spirituous product is divided into “heads”, “hearts”, and “tails”. The volatile fractions are separated through cuts in the distillate to obtain desirable compounds and remove undesired ones. The “heads” fraction includes aldehydes, esters, and methanol. Methanol is undesirable in the distillate due to its toxicity; controlling the distillation can remove this compound effectively [23]. The “tails” fraction includes acetic acid and furfural, which must be removed to reduce acidity. The presence of volatile compounds in each fraction depends on their boiling points and affinity for water, alcohol, or both. Consequently, there are four groups of volatile compounds: compounds fully or partially soluble in alcohol, compounds soluble in water, those soluble in both alcohol and water, and, finally, compounds that are not soluble in alcohol but can be transported by water vapor (hydro-distillation) [24]. The “heads” and the beginning of “hearts” include volatile compounds that are soluble in alcohol and have a low boiling point (i.e., acetaldehyde and ethyl acetate), compounds entirely or partially soluble in alcohol and water that have a boiling temperature of less than 200 °C (i.e., methanol and superior alcohols), and compounds entirely or partially soluble in alcohol with high boiling temperature (i.e., fatty acids and their esters, isoamyl acetate, ethyl octanoate, ethyl decanoate, and ethyl dodecanoate) [24]. The middle of the “hearts” and the “tails” contain volatile compounds that are entirely or partially soluble in water with boiling points higher than the boiling temperature of water (i.e., acetic acid, 2 phenyl- ethanol, ethyl decanoate, and diethyl succinate) and compounds soluble in water with high boiling points (i.e., furfural). Hydrophilic compounds, such as furfural and higher alcohols, are present in all distillate fractions [24]. The “heads”, “hearts”, and “tails” were collected and quantified during the distillation process carried out for this study. Appendix A presents the calculations made to obtain the quantification of the fermentation and distillation processes.

The efficiency of the ethanol yielded during the fermentation process was 88.85%, as the absolute ethanol obtained during this process was 10.99% and the theoretical ethanol yield was 12.37%. This value indicates that 88.85% of the ethanol that could be produced in this fermentation, according to the fermentable sugar content, was made during this process. Additionally, the fermentation process efficiency was 63.98%. For the distillation process, the efficiency was 94.03%. This result indicates that 94.03% of the ethanol in the fermented wash was recovered during the distillation process. A higher distillation efficiency represents a more efficient separation of the ethanol from the fermented wash and a higher yield. Table 3 presents the distillation results and ethanol content of the cuts made during this process.

Table 3. Distillation results.

	Total Volume (L)	Ethanol Content (% ABV)	Ethanol Volume (L)	Fraction of Total Volume (%)	Total Ethanol Fraction (% ABV)
Wash	12.00	10.99	1.32	100.00	100.00
Head	0.18	85.00	0.15	1.50	11.60
Heart	1.56	65.00	1.01	13.00	76.89
Tail	0.35	21.00	0.07	2.92	5.57
Vinasses	9.91	1.00	0.10	82.58	7.51

The head contains high levels of unwanted secondary components, including most of the methanol produced, and should be discarded. This cut represented 1.5% of the total volume of the fermented wash or 11.6% of the total ethanol recovered. On the other hand, the heart is the desired distillate, containing most of the ethanol produced and some concentrated aromas produced during fermentation. This cut represented the volume equivalent to 13% of the total volume of the fermented wash or 76.89% of the total volume

of ethanol obtained. The tail represents 2.92% of the total volume of the fermented wash or 5.57% of the total ethanol fraction.

The overall yield of the process was 60.16%. This value indicates that 60.16% of the initial sugar content in the sugarcane juice was converted into ethanol and recovered during the distillation process. A higher overall yield represents a more efficient use of the sugar content in the sugarcane juice and a higher ethanol yield.

3.2. Accelerated Aging Method

3.2.1. Alcohol Content

The alcohol content of the spirit samples was monitored during the aging process. Ethanol concentration might be affected by the ultrasound technology through the cavitation phenomenon. When high-energy ultrasound is applied to a liquid, molecules expand and bubbles are formed. These cavities can explosively collapse, generating localized abrupt changes in temperature that could volatilize some of the ethanol in the spirituous samples. Moreover, the alcohol concentration in the samples studied was constant throughout the accelerated aging process. The alcohol by volume (ABV) of the aged spirits was the same as in Table 1 for the untreated samples. There were no recognizable changes in the ethanol concentration of samples treated for up to 60 min with the ultrasonic bath. The conditions of the experiment might explain this behavior. For instance, the ultrasonic bath's operation frequency might not be high enough to cause ethanol volatilization with the scheme used. Additionally, the airtight vessels used in the batch process might prevent the liberation of volatile compounds, including ethanol. Previous studies have reported positive results for the ultrasound aging technique for continuous schemes. These results were based on color change and total polyphenol index values on continuous ultrasonic treatments for sugar cane and wine spirits [9,13]. Moreover, the potential loss of ethanol might be carefully checked when scaling the process, especially in continuous methods, as the airtightness might be challenging to guarantee.

3.2.2. Titratable Acidity

All aged samples studied were diluted to 40% ABV to study titratable acidity and pH. The reported values for titratable acidity and pH are presented in Table 4. The dual evaluation of pH and acidity yield a more robust perception of how the different aging treatments studied influence the final sensory profile of the sugar cane honey spirit, highlighting the relevance of these parameters in quality control as they indicate the right fermentation process and no contamination [21].

Table 4. Titratable acidities and pH values of unaged control sample (REF) and accelerated aging treatment samples.

Sample	Titratable Acidity (g/100 mL)	pH
REF	0.012	4.309
1	0.011	4.375
2	0.008	4.228
3	0.009	4.360
4	0.009	4.387
5	0.010	4.599
6	0.010	4.627
7	0.011	4.541
8	0.009	4.582

The highest pH value was reported for sample 6, while the lowest pH value was exhibited by sample 2. Overall, the aged samples had higher pH values when compared to the unaged reference. Titratable acidity, measured in grams of acetic acid per 100 mL, impacts the sensory profile of the final product. The reference sample had the highest

values of acidity, representing a potential difference in sensory perception when compared with other samples. This suggests that the aging process influences the drop in acidity due to chemical reactions that decrease the concentration of organic acids, including acetic acid [21]. Sample 2 exhibited the lowest value of acidity (0.008 g/100 mL), which suggests that samples with the longest aging treatment yield products with lower acidity and thus an attenuated flavor complexity. These findings suggest an alteration in acidity due to the aging process and, therefore, also an alteration in the sensory profile.

3.2.3. Color Determination

Volatile congeners are produced during fermentation, and their concentrations are only measurable in the final spirits, as monitoring procedures are generally not applicable. Esters and aldehydes are essential in spirits' sensory profiles. Their production depends on the abundance of their corresponding alcohols and acyl-coA radicals involved in yeast metabolism, such as n-propyl, isobutyl, and isoamyl alcohol. Distillation purifies and concentrates the compounds formed during fermentation to enhance the sensory profile, significantly impacting the final product's organoleptic characteristics [21].

The mean values of the absorbance readings for all aged samples and the untreated reference sample and their standard deviations are presented in Table A2 in Appendix B, which provides the chromatic characteristics data. The CIELab parameters are presented in Table 5. They include the trichromatic components (X, Y, Z), clarity (L*), red/green attribute (a*), yellow/blue attribute (b*), chroma (C*), tone (H*), and the calculated total color difference (ΔE) of each sample compared to the unaged reference. The total color difference between pairs of aged samples and unaged references is presented in Table A3.

Table 5. Chromatic characteristics of the aged samples of sugar cane spirit and total color difference against the unaged sample (REF).

Sample	X	Y	Z	L*	C*	H*	a*	b*	ΔE
REF	93.23	98.42	103.62	99.4	1.29	94	−0.09	1.29	-
1	85.53	90.88	81.82	96.4	11.1	95.96	−1.15	11.04	10.26
2	88.96	94.41	92.93	97.8	5.68	99.79	−0.97	5.6	4.68
3	83.86	89.01	80.51	95.6	10.72	95.09	−0.95	10.68	10.17
4	85.34	90.49	79.42	96.2	12.59	93.61	−0.79	12.56	11.74
5	87.56	93.03	85.05	97.2	10.24	96.3	−1.12	10.18	9.22
6	85.92	91.25	81.45	96.5	11.62	95.2	−1.05	11.57	10.72
7	89.07	94.72	89.63	97.9	8.19	99.1	−1.3	8.09	7.07
8	88.37	93.88	84.86	97.6	10.97	95.89	−1.13	10.91	9.84

In general, the clarity (L*) values range from 0 to 100, where 0 represents black and a value of 100 relates to a colorless beverage. Overall, the samples studied reported low intensity in color, with an interval of 95.6–97.9 for the clarity attribute. This parameter's highest value (99.4) was measured for the unaged sugar cane spirit reference sample, followed by sample 7 (97.9). The lowest value of clarity was reported for sample 3 (95.6), followed by samples 4 (96.2), 1 (96.4), and 6 (96.5). A lower value of clarity means a higher intensity of color. In addition, the data obtained for the parameters a* and b* for all samples indicate green (when a* < 0) and yellow (when b* > 0) hues, respectively. Unaged sugar cane spirit is colorless due to the absence of phenolic compounds; the acquired color changes are due to aging [4]. This phenomenon occurs primarily due to oxidative reactions, condensation, esterification, and the extraction of aromatic aldehydes, tannins, and lignin-related substances that give aged rum its characteristic color [25]. Figure 3 presents the visual color representation of the samples studied.

REF	1	2	3	4	5	6	7	8
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Figure 3. Average visual color representation of unaged (REF) and aged samples (1–8).

The total color difference (ΔE) is presented in Table 5, considering the classification used by Abreu [9], where $\Delta E < 1.5$ represents a slight difference, $1.5 < \Delta E < 3$ indicates a distinct difference, and $\Delta E > 3$ is classified as a very distinct difference. All aged samples present distinct differences in total color compared to the unaged sample. Therefore, the aging treatment induces a higher intensity of color in the samples. The highest color change was presented in sample 4. Overall, the color change increased as the extraction time increased. Most treatments with a longer ultrasound duration (20 min) showed larger differences in total color compared with the samples that had a shorter ultrasound duration (10 min), regardless of the ethanol concentration or successive treatment cycles. Previous studies found a linear relationship between ΔE and the total polyphenol index, with a high positive correlation between these parameters [9]. Furthermore, a previous article on a rice alcoholic beverage presented results where the values of ΔE for samples aged by sonication technology achieved in a short time (<1 day) were comparable with a conventional process (control treatment) that lasted for about 1.5 months to achieve a similar color [26]. Among the aged samples, as shown in Table A3, eight pairs (1–3, 1–5, 1–6, 1–8, 6–3, 6–4, 8–5, and 8–6) presented minor differences in total color. Distinct differences were obtained in 10 pairs (1–4, 2–7, 3–4, 3–5, 3–8, 4–5, 4–8, 5–6, 5–7 and 7–8). The remaining pairs of samples presented very distinct differences in total color.

3.2.4. Electronic Tongue Evaluation

The electronic tongue method was used to evaluate the flavor characteristics of ultrasound-aged samples (1–8) with various alcohol concentration levels and ultrasound durations. Additionally, neutral ethanol 5% ABV (NE), an unaged sample (Ref), and commercially aged rum brand samples were tested using the E-tongue technique. Figure 4 shows the radar chart of the E-tongue responses for neutral ethanol 5% ABV (NE), the unaged sample (Ref), and the average aged samples (1–8). The E-tongue average response and standard deviation values of the eight sensors for the studied samples are presented in Table A4 in Appendix C, which exhibits flavor characteristics data. The average response and standard deviation for food grade neutral ethanol 5% ABV (NE), the untreated reference (Ref), and the average commercial sample are also exhibited. According to the manufacturer's recommendations, the data for the saltiness taste indicator should be discarded for further analysis in alcoholic samples. The ultrasound-aged samples exhibited sourness values below the taste threshold. The reported bitter aftertaste (aftertaste-B) values are below the taste threshold, except for samples 2, 5, and 6. The umami aftertaste (richness) also presented values below the taste threshold for most aged samples, except for samples 7 and 8. The values for the remaining taste indicators surpassed the taste threshold, serving as effective taste indicators. Aged samples demonstrated strong bitterness, astringency, and umami, ranging from stronger to less strong. Ultrasound-aged samples had a weak astringency aftertaste (aftertaste-A). The bitter aftertaste (aftertaste-B) and astringency aftertaste (aftertaste-A) presented the most significant data variability among the samples. Samples 3, 4, 5, and 7 presented the most taste indicator values within one deviation of the average aged-sample response. Samples 5 and 6 exhibited significant differences in most taste indicators compared to the unaged reference sample. Overall, the untreated reference showed similar taste indicator values compared to the average aged responses. The unaged reference presented values for sourness, astringency aftertaste (aftertaste-A), bitter aftertaste (aftertaste-B), and umami taste indicators within one deviation of the average aged-sample response. When compared to the sugar cane honey spirit (aged or not) and the commercial samples, the neutral ethanol 5% ABV (NE) presented higher values for the bitterness, astringency, astringency aftertaste (aftertaste-A), and umami indicators

while presenting a lower response value for sourness. This may indicate that the rectification process may intensify bitterness, astringency, astringency aftertaste (aftertaste-A), and umami flavors while attenuating the sour flavor. However, sourness, bitter aftertaste (aftertaste-B), and umami aftertaste (richness) exhibited values below the taste threshold for the NE sample.

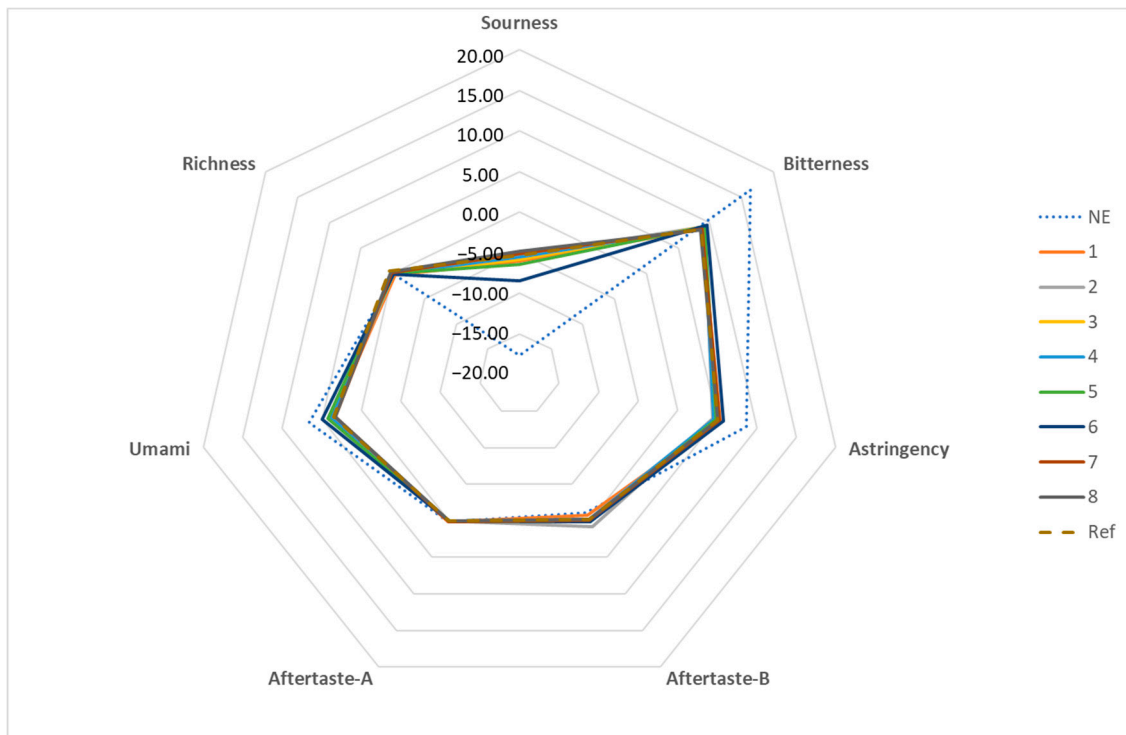


Figure 4. Radar chart of E-tongue responses of neutral ethanol 5% ABV (NE), unaged sample (Ref), and average aged samples (1–8).

Generally, the aged samples tend to present lower response values for the sourness, astringency, astringency aftertaste (aftertaste-A), and umami aftertaste (richness) indicators than the untreated sample. The ultrasound-treated samples exhibited higher response values for the bitterness, bitter aftertaste (aftertaste-B), and umami indicators than the unaged reference. Consequently, the aging process enhances the bitterness, bitter aftertaste, and umami flavors while impairing the sourness, astringency, astringency aftertaste, and umami aftertaste of sugar cane honey spirits. Additionally, the sugar cane honey spirits' overall flavor profile presents notable bitterness, astringency, and umami indicators while exhibiting a slight tendency to the astringency aftertaste indicator and is below the taste threshold for sourness, umami aftertaste, and bitter aftertaste. This flavor profile is comparable to the untreated reference sample flavor profile. Therefore, the differences between the aged samples and the untreated sample are not remarkable.

Furthermore, commercially aged rum brand samples were also tested using the E-tongue technique. Samples 1, 5, 7, and 8 showed response values closest to the average commercial sample results in most taste indicators. Some aged samples (3, 5, and 7) were selected for further analysis because of their proximity to the commercial rum brands studied, their closeness to the average aged-sample values (1–8), and their significant difference from the untreated sample. Figure 5 shows the radar chart of the E-tongue measurement for the unaged sample (Ref), average aged samples (3, 5, and 7), and average commercial samples.

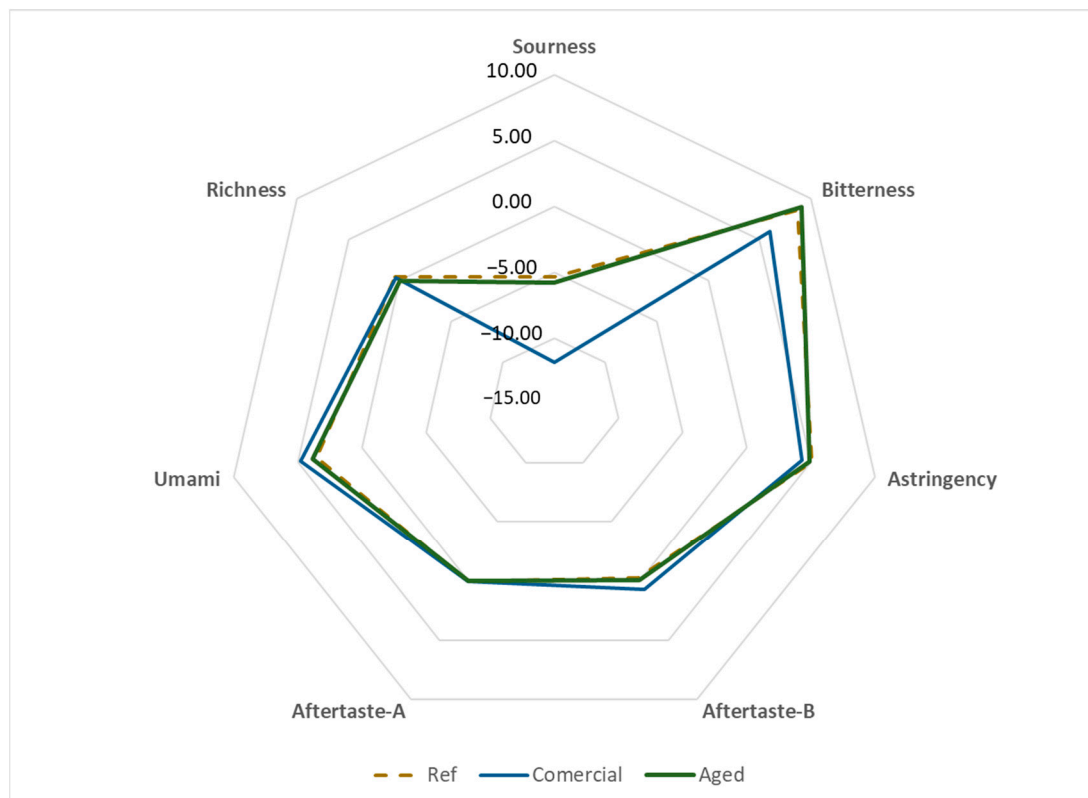


Figure 5. Radar chart of E-tongue responses for the unaged sample (Ref), average aged samples (3, 5, and 7), and average commercial samples.

The average aged sample (3, 5, and 7) exhibited comparable values for the taste indicators compared to the untreated reference sample. The taste profile for both samples exhibited sourness values below the taste threshold. The reported bitter aftertaste (aftertaste-B) and umami aftertaste (richness) values are below the taste threshold. Additionally, the values for the remaining taste indicators surpassed the taste threshold, serving as effective taste indicators. Both samples demonstrated strong bitterness, astringency, and umami, from stronger to less strong, respectively, and showed a slight indication of astringency aftertaste (aftertaste-A). Compared to the average aged sample, the commercially aged rum sample presented similar values for the umami aftertaste (richness), astringency, and astringency aftertaste (aftertaste-A) taste indicators. Also, the umami and bitter aftertaste (aftertaste-B) values were slightly higher for the commercial samples studied. In contrast, the results for bitterness and sourness are significantly lower in the average commercial sample.

3.2.5. Electronic Nose Evaluation

An electronic nose was used to evaluate the aroma profile characteristics of the aged samples against the unaged reference. The radar chart of the E-nose measurements of the ten sensors for the studied samples is presented in Figure 6. Appendix D presents the aroma characteristics data, and Table A5 presents the average response values and standard deviations for the measured aged spirit samples and untreated reference. All results were normalized to the most significant value response for each sensor. Response values for the sensors W1C (sensitive to aromatic constituents, benzene), W3C (sensitive to ammonia), W6S (sensitive to hydrides), W5C (sensitive to olefin, short-chain aromatic compounds), W2W (sensitive to organic sulfides), and W3S (sensitive to long-chain alkanes) presented moderate standard deviations (<15%) when compared among aged samples. The remaining response values differed significantly among the treated samples. Samples 1, 3, and 4 have the most significant overall differences when compared to the reference

sample for the sensors W5S (susceptible to nitrogen oxides), W1S (sensitive to methane), W1W (sensitive to sulfides), and W2S (sensitive to alcohols, aldehydes, and ketones). The average response of samples 1, 3, and 4 is used for further analysis, as the significant differences when compared to the reference sample indicate that the ultrasound aging treatment influences the aroma of the sugar cane spirit samples. The highest value for the sensor W5S, which is highly sensitive to nitrogen oxides, is presented by sample 8, and the lowest value is presented by sample 1. The reference sample shows the most significant value for sensor W1S, which denotes sensitivity to methane in the headspace environment; the lowest value for this sensor is reported by sample 1. For the sensor W1W, the most significant value is presented by sample 8 (this sensor is related to sensitivity to sulfides) and the lowest value is shown by sample 3. Finally, the unaged sample measured the most significant value for sensor W2S, which is related to sensitivity to alcohols, aldehydes, and ketones; the lowest value for sensor W2S is associated with sample 1.

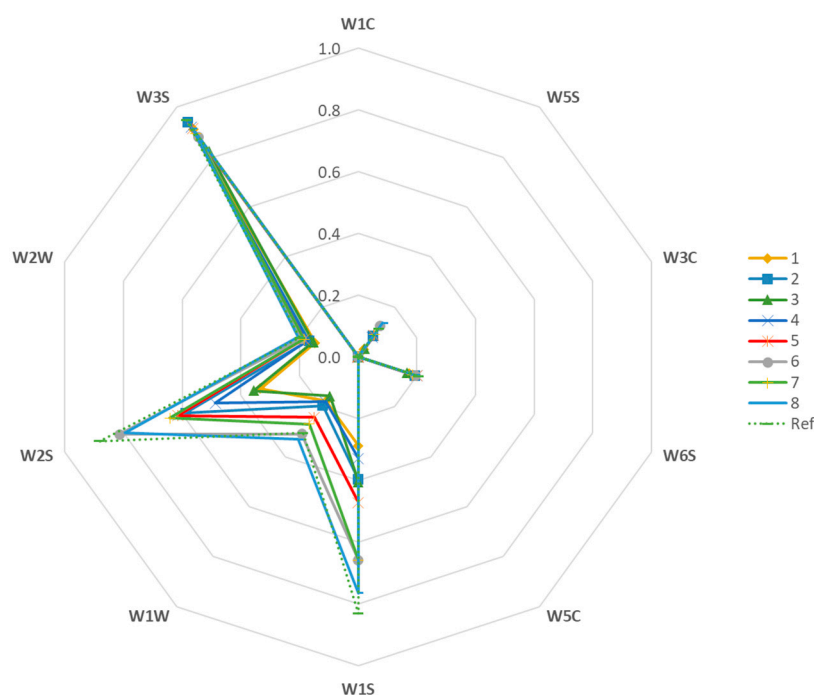


Figure 6. Radar chart of E-nose responses of unaged sample (Ref) and average aged samples (1–8).

The aroma profile of the average aged sample is compared to the average commercial data. The aroma profile results for three commercial traditionally aged rum brands studied are reported in Table A6. The results for the reference unaged sample are shown in Figure 7. The average ultrasound-treated sample has the narrowest difference for sensor W3C compared to the average commercial sample (the sensitivity of this sensor is related to ammonia), followed by sensors W5C (sensitive to olefin, short-chain aromatic compounds) and W1C (sensitive to aromatic constituents, benzene), respectively. The sensors with the most significant difference when the average aged sample is compared to the commercial rum samples are W1S (sensitive to methane), W2W (sensitive to organic sulfides), and W2S (sensitive to alcohols, aldehydes, and ketones), respectively. The results show a significant diminishing in the intensity of methane (W1S), organic sulfides (W2W), and alcohols, aldehydes, and ketones (W2S) compounds in both the traditional and ultrasound-aged samples studied.

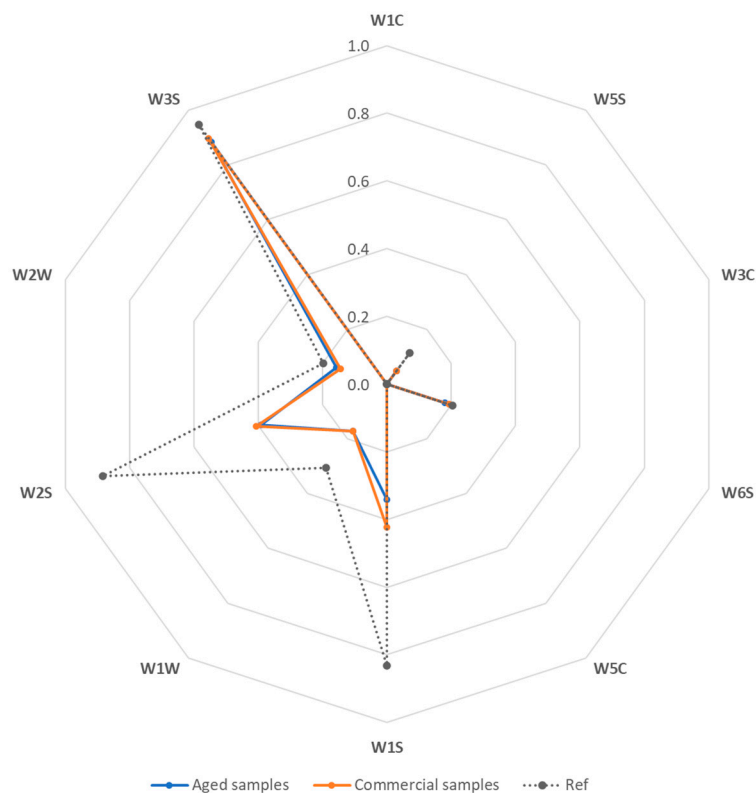


Figure 7. Radar chart of E-nose responses of the unaged sample (Ref), average aged samples (1, 3, and 4), and average commercial samples.

Patrons of 47 aroma descriptors were created using the E-nose system kit and software (PEN3, Aisense Analytics GmbH, Schwerin, MV, Germany). These patrons are compared to the average ultrasound-aged samples and the commercial rum brand samples to identify their aroma profiles regarding the aroma descriptors. These aroma profiles are shown in Table A7. Furthermore, the commercial brand samples exhibited 33 aromas of the 47 studied, while the ultrasound-treated samples contained 24. Additionally, all aromas measured in the ultrasound-aged samples are within the aromas presented in the commercial brand samples in various intensities. Therefore, 73% of the aroma profile obtained for a commercial rum brand was obtained through the ultrasound treatment.

4. Discussion

In general, the methods used allowed us to produce and accurately characterize an aged sugar cane honey spirit. The organoleptic profile obtained by extracting the compounds from the oak chips yielded a distinct overall color change. Similar flavor and aroma profiles were achieved compared to a commercial sample.

The chromatic characteristics of ultrasound-aged spirit samples and the reference samples allow the proposition of visible spectrophotometry and the CIELab method to estimate the total color change. Overall, the color changes increased as the extraction time increased. Most treatments with a higher ultrasound duration (20 min) showed higher differences in total color regardless of the ethanol concentration or the use of successive cycles of treatment. Therefore, the ultrasound treatment facilitates the extraction of color pigments from the wood chips. When analyzing the data obtained for total color change, it is shown that the results for most of the samples are towards a value of 10. The box plot analysis shows that the data are not symmetrical, with a tendency towards the higher range of the spectrum, and also presents sample 2 as an outlier. A one-sample *t*-test was performed with a test value of 1.5, since this is the boundary for detecting the color difference according to the ranges studied. The results showed that the mean significantly

differs (p -value < 0.001) from the test value and, thus, the aging treatment, regardless of the factor and level, noticeably changes the color of the spirit [27].

Additionally, the overall flavor profile of the sugar cane honey spirits could be determined using e-tongue sensing. The results showed the flavor profiles for ultrasound-aged, untreated reference, and commercial brand samples. Although the flavor profiles for the aged and unaged samples are comparable, a more robust method is needed to identify the specific flavor compounds in each sample. The attenuated values of the bitterness and sourness taste indicators in the commercial sample could be linked to a maturation period after the aging treatment. Furthermore, a slow extraction process, similar to the traditional aging method, could yield an attenuated flavor profile for the bitterness and sourness indicators. These taste indicators may be related to the descriptors, chocolate, smoke, toast, and coffee identified in the aroma profile studied. The attenuation of these taste indicators could be linked to a maturation period after the aging treatment. Some compounds, such as *p*-cymene (roasted) [28], 2-methoxyphenol (smoky) [23], caffeic acid (coffee and smoky) [29], and diacetyl (cacao) [30], could be linked to the previous descriptors found in the flavor wheel of rum [21]. These compounds are induced into the spirit during the production process.

The aroma profiles of all samples studied were obtained through e-nose sensing regarding aroma descriptors. This proved to be an effective technique for obtaining an accurate aroma profile. Most of the aroma profile of the commercial rum was also present in the ultrasound-aged samples. Additionally, the ultrasound aging process may induce changes in the aroma profile of the sugar cane spirit, presenting higher variability in compounds related to nitrogen oxide, methane, sulfide, alcohol, aldehyde, and ketone aromas. These compounds are related to flavor descriptors in a sugar cane spirit, such as floral, fruity, spicy, and other vital odorants. Previous authors have reported important flavor volatiles in rum, relating the compounds responsible for the aroma to their corresponding sensory attributes [23]. The key odorants in aged rum are highlighted. These results are shown in Table A8. The essential aromas in rum reported previously have been identified via E-nose sensing for the ultrasound-aged samples, such as dried apricot, pineapple, apple, honey, vanilla, and smoke. Additionally, more general descriptors can be related to the results found in this study, such as fruity, floral, spicy, sweet, and alcoholic.

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Appendix A. Calculation Scheme on Rum Fermentation and Distillation [21]

A.1. Pre-Fermentation Dilution of Sugarcane Honey

Outline of sugarcane honey dilution for the onset of fermentation:

The initial concentration of sugarcane honey is 70 °Bx.

Dilution to 12 L of mash at 23 °Bx.

To obtain the specific gravity (SG) of the sugarcane honey at 23 °Bx, the formulas presented in Table A1 were used:

Table A1. Formulas to obtain specific gravity (SG), alcohol by volume percentage (%ABV), and Brix degrees (°Bx).

Table A1. Chromatic characteristics of the aged samples of sugar cane spirit and total color difference against unaged sample (REF).

SG	% ABV	°Bx
$SG = (0.99958251 - 0.0037958995 \cdot ^\circ Bx)^{-1}$	$\%ABV = [0.88411955 \cdot ^\circ Bx^{0.5} - 0.61771014]^2$ $\%ABV = 134.65985 \cdot SG \cdot \ln(SG) - 0.60945368$	$^\circ Bx = 263.15882 - 263.25586 \cdot SG^{-1}$

A temperature correction factor is necessary when the temperature of sugarcane honey is not 20 °C, using the following equation:

$$SG_c = SG_m \cdot \left(-1.35666446 \cdot 10^{-8} T^3 + 5.88216338 \cdot 10^{-6} T^2 - 2.02881424 \cdot 10^{-5} T + 9.98161431 \cdot 10^{-1} \right)$$

where T is the temperature in Celsius and SG_m and SG_c are the measured and the corrected SG, respectively.

The SG is determined to be 1.096. To obtain the mass of the honey, the following equation was used:

$$12 \text{ L} \cdot 1.096 = 13.15 \text{ kg}$$

The following formula is used to determine the final volume of the solution:

$$C_1 \cdot V_1 = C_2 \cdot V_2$$

$$70 \text{ }^\circ Bx \cdot M1 = 23 \text{ }^\circ Bx \cdot 13.15 \text{ kg}$$

$M1 = 4.34 \text{ kg}$ of sugarcane honey at 70 °Bx

To convert the mass to volume using the SG of sugarcane honey at 70 °Bx (1.355):

$$V = \frac{4.34}{1.355} = 3.2 \text{ L of sugarcane honey at } 70 \text{ }^\circ Bx$$

Therefore, 8.8 L of water was added (12–3.2), which is necessary to obtain 12 L of mash at 23 °Bx.

A.2. Fermentation Efficiency Calculation

Fermentation efficiency (E) directly influences the ethanol yield. It is calculated by comparing the actual ethanol produced (% v/v) to the theoretical ethanol (% v/v), expressed as a percentage. The equation for fermentation efficiency is:

$$E(\%) = \frac{\text{Real Volume}}{\text{Theoretical Volume}} \times 100$$

Consider the stoichiometry of sucrose's ($C_{12}H_{22}O_{11}$, 342 g) conversion to ethanol ($4 \times C_2H_6O$, 184 g) to calculate the theoretical yield. The theoretical yield is 0.538 g of ethanol per gram of sucrose (184 g ethanol/342 g sucrose).

The density of sugarcane honey with 23 °Bx is 1.096, resulting in 13.15 kg of mash for a 12 L volume. With a 0.23 fraction of sucrose in the must, we have 3.02 kg of sucrose. Assuming 100% of soluble solids as sucrose, the theoretical volume of ethanol can be calculated as follows:

$$\text{Theoretical Volume} = \frac{(3.02 \text{ kg sucrose} \times 0.538)}{0.7893 \text{ (ethanol SG)}} = 2.06 \text{ L of ethanol (1.63 kg)}$$

To calculate the real volume, consider that the fermentation process yielded 10.99% ethanol:

$$\text{Real Volume} = \frac{10.99\% \text{ ethanol}}{100} \times 12 \text{ L honey} = 1.32 \text{ L of ethanol}$$

Finally, the fermentation efficiency is calculated as follows:

$$E(\%) = \frac{1.32 \text{ L}}{2.06 \text{ L}} \times 100 = 63.98\%$$

For the fermentation process of sugarcane honey, the efficiency of 63.98% indicates that 63.98% of the sugars were converted into alcohol. A higher efficiency represents a smaller gap between the theoretical and real volumes and a higher sugar-to-alcohol conversion rate.

The theoretical ethanol yield can be obtained using the theoretical ethanol mass obtained using the reaction stoichiometry and the total mass of the mash as follows:

$$\text{Theoretical\% ABV} = \frac{1.63 \text{ kg}}{13.15 \text{ kg}} \times 100 = 12.37\%$$

The ethanol yield efficiency can be obtained by comparing the theoretical and real ethanol yield as follows:

$$\text{Ethanol Yield Efficiency (\%)} = \frac{10.99\%}{12.37\%} \times 100 = 88.85\%$$

This value indicates that 88.85% of the ethanol that could be produced in this fermentation, according to the fermentable sugar content, was made during this process.

A.3. Distillation Efficiency Calculation

The alcohol by volume percentage in the wash was obtained using the following expression based on the initial (SG_i) and final (SG_f) specific gravity:

$$\%ABV = 95.819 \cdot SG_f \cdot \left(\frac{SG_i - SG_f}{1.775 - SG_i} \right) = 10.99\% \text{ ABV}$$

Distillation efficiency (DE) expresses ethanol recovery from the wash. For this purpose, the volume of absolute ethanol distilled (% v/v), as compared to the ethanol present in the fermented must (% v/v), is expressed as a percentage. The equation for distillation efficiency is:

$$DE(\%) = \frac{\text{Volume of ethanol distilled}}{\text{Volume of ethanol in the fermented wash}} \times 100$$

We have 12 L of fermented must with 10.99% ABV:

$$12 \text{ L} \cdot 10.99\% = 1.31 \text{ L (1.04 kg) of ethanol in the fermented wash}$$

The wash was placed in the distillation column and all the distillate was collected until the instantaneous alcohol value read 1% ABV. Table 3, presented in the distillation efficiency calculation section, reports the results of the collection of the head, heart, and tail.

Based on Table 3, it is necessary to determine the total ethanol distilled and the ethanol content as follows:

$$\text{Total volume} = \text{head} + \text{heart} + \text{tail} = 0.18 + 1.56 + 0.35 = 2.09 \text{ L}$$

$$\text{Ethanol content} = \frac{\text{head} + \text{heart} + \text{tail}}{\text{Total volume}} = \frac{0.18 \cdot 0.85 + 1.56 \cdot 0.65 + 0.35 \cdot 0.21}{2.09} \cdot 100 = 59.35\% \text{ ABV}$$

Therefore, after distillation, 2.09 L at 59.35% ABV.

$$2.09 \text{ L} \cdot 59.35\% = 1.24 \text{ L of ethanol distilled}$$

The efficiency of the distillation is:

$$DE = \frac{1.24 \text{ L}}{1.32 \text{ L}} \cdot 100 = 94.03\%$$

The distillation efficiency of 94.03% indicates that 94.03% of the ethanol present in the fermented wash was recovered during the distillation process. A higher distillation efficiency represents a more efficient separation of the ethanol from the fermented wash and a higher yield.

Overall Yield:

The overall yield of sugarcane honey spirit production is the product of fermentation and distillation efficiency. In this example, the overall yield (OY) is calculated as follows:

$$OY(\%) = E \times DE$$

$$OY = 63.98\% \times 94.03\% = 60.16\%$$

The overall yield of 60.16% indicates that 60.16% of the initial sugar content in the sugarcane juice was converted into ethanol and recovered during the distillation process. A higher overall yield represents a more efficient use of the sugar content in the sugarcane juice and a higher ethanol yield.

Appendix B. Chromatic Characteristics Data

B.1. Absorbance Readings for Unaged and Aged Sugar Cane Spirit Samples

The mean values and standard deviations for the absorbance readings of the aged samples and untreated reference at wavelengths of 450, 520, 570, and 630 nm are presented in Table A2. The CIELab method was used to determine the color coordinates of the samples.

Table A2. Mean values and standard deviations (STDs) of the absorbance readings for aged samples and unaged reference (REF) sample at 450, 520, 570, and 630 nm.

Sample	450 nm	STD	520 nm	STD	570 nm	STD	630 nm	STD
Ref	0.014	0.001	0.008	0.001	0.006	0.001	0.003	0.001
1	0.118	0.047	0.049	0.02	0.03	0.012	0.02	0.007
2	0.062	0.01	0.027	0.011	0.02	0.012	0.015	0.012
3	0.125	0.012	0.058	0.012	0.04	0.016	0.027	0.015
4	0.131	0.039	0.054	0.019	0.03	0.011	0.016	0.006
5	0.101	0.049	0.038	0.018	0.021	0.011	0.012	0.004
6	0.12	0.011	0.048	0.013	0.028	0.015	0.016	0.015
7	0.078	0.016	0.027	0.006	0.016	0.004	0.01	0.002
8	0.102	0.011	0.035	0.002	0.016	0.003	0.007	0.006

B.2. Total Color Difference among Aged Sugar Cane Spirit Samples

The total color difference (ΔE) among aged samples is presented in Table A3, obtained by Equation (1), which uses the following chromatic parameters for both samples in each case: clarity (L^*), red/green attribute (a^*), and yellow/blue attribute (b^*).

Table A3. Total color difference (ΔE) among aged samples.

Samples	Ref	1	2	3	4	5	6	7	8
Ref	-	10.26	4.68	10.17	11.74	9.22	10.72	7.07	9.84
1	10.26	-	5.62	0.9	1.57	1.17	0.55	3.31	1.21
2	4.68	5.62	-	5.54	7.14	4.62	6.11	2.51	5.32
3	10.17	0.9	5.54	-	1.98	1.68	1.27	3.48	2.02
4	11.74	1.57	7.14	1.98	-	2.6	1.07	4.81	2.19
5	9.22	1.17	4.62	1.68	2.6	-	1.56	2.21	0.83
6	10.72	0.55	6.11	1.27	1.07	1.56	-	3.76	1.29
7	7.07	3.31	2.51	3.48	4.81	2.21	3.76	-	2.84
8	9.84	1.21	5.32	2.02	2.19	0.83	1.29	2.84	-

Appendix C. Flavor Characteristics Data

The E-tongue average response and standard deviation values of the eight sensors for the studied samples are presented in Table A4. The average responses and standard deviations for food grade neutral ethanol 5% ABV (NE), the untreated reference (Ref), and the average commercial sample are also shown.

Table A4. Average response values for E-tongue sensing and the standard deviations for neutral ethanol 5% ABV (NE), the measured aged spirit samples (1–8), untreated reference (Ref), and average commercial sample.

Samples	Sourness	Bitterness	Astringency	Aftertaste-B				
NE	-17.68	2.04	16.46	1.16	8.71	0.83	-1.13	0.40
1	-6.07	0.46	8.70	0.03	5.34	0.17	-0.72	0.02
2	-5.30	0.28	9.19	0.37	4.42	0.35	0.75	0.04
3	-5.79	0.43	9.14	0.22	4.63	0.51	-0.14	0.03
4	-5.54	0.08	9.01	0.06	4.58	0.05	-0.25	0.00
5	-6.40	0.56	9.21	0.31	4.84	0.55	0.09	0.01
6	-8.50	2.39	9.57	0.47	5.77	0.80	0.12	0.03
7	-5.14	0.93	8.76	0.48	5.25	0.75	-0.13	0.03
8	-4.85	0.21	8.52	0.14	5.05	0.33	-0.18	0.02
Average	-5.95	0.67	9.01	0.26	4.99	0.44	-0.06	0.02
Ref	-5.34	0.34	8.66	0.12	5.10	0.34	-0.24	0.07
Commercial	-11.86	0.23	5.98	0.16	4.33	0.44	0.78	0.07

Samples	Aftertaste-A	Umami	Richness	Saltiness				
NE	0.14	0.06	6.59	0.84	-0.34	0.01	22.56	1.43
1	0.17	0.01	3.74	0.21	-0.33	0.03	9.42	0.27
2	0.05	0.01	3.63	0.04	-0.13	0.02	8.77	0.32
3	0.04	0.01	3.82	0.12	-0.12	0.01	9.47	0.40
4	0.02	0.01	3.78	0.16	-0.08	0.01	9.33	0.16
5	0.06	0.01	4.26	0.24	-0.08	0.01	10.03	0.60
6	0.03	0.01	4.99	0.78	-0.11	0.01	12.07	2.12
7	0.08	0.01	3.32	0.42	0.16	0.01	8.23	1.05
8	0.07	0.01	3.30	0.14	0.22	0.01	8.06	0.22
Average	0.07	0.01	3.85	0.26	-0.06	0.01	9.42	0.64
Ref	0.07	0.05	3.52	0.13	0.48	0.14	8.55	0.18
Commercial	0.09	0.01	4.79	0.17	0.41	0.13	2.24	0.26

Appendix D. Aroma Characteristics Data

The E-nose average response values of the ten sensors for the studied samples are presented in Table A5. The standard deviations for the measured aged spirit samples and untreated reference are also exhibited. All results were normalized to the most significant value response for each sensor.

Table A5. Average response values for E-nose sensing and standard deviation for the measured aged spirit samples (1–8) and untreated reference (Ref).

Samples	W1C	W5S	W3C	W6S	W5C
Ref	4.99×10^{-5}	1.13×10^{-1}	5.87×10^{-5}	2.04×10^{-1}	6.20×10^{-5}
1	6.17×10^{-5}	3.11×10^{-2}	6.95×10^{-5}	1.74×10^{-1}	7.31×10^{-5}
2	5.08×10^{-5}	8.27×10^{-2}	6.08×10^{-5}	1.94×10^{-1}	6.41×10^{-5}
3	6.51×10^{-5}	3.31×10^{-2}	7.26×10^{-5}	1.66×10^{-1}	7.67×10^{-5}
4	5.21×10^{-5}	7.79×10^{-2}	6.22×10^{-5}	2.01×10^{-1}	6.53×10^{-5}
5	5.16×10^{-5}	8.66×10^{-2}	6.17×10^{-5}	2.00×10^{-1}	6.56×10^{-5}
6	4.96×10^{-5}	1.22×10^{-1}	5.86×10^{-5}	1.95×10^{-1}	6.11×10^{-5}
7	5.31×10^{-5}	8.99×10^{-2}	6.14×10^{-5}	1.86×10^{-1}	6.44×10^{-5}
8	4.89×10^{-5}	1.36×10^{-1}	5.81×10^{-5}	1.94×10^{-1}	6.09×10^{-5}
Average	5.41×10^{-5}	8.24×10^{-2}	6.31×10^{-5}	1.89×10^{-1}	6.64×10^{-5}
STD	5.95×10^{-6}	3.70×10^{-2}	5.17×10^{-6}	1.25×10^{-2}	5.61×10^{-6}
Samples	W1S	W1W	W2S	W2W	W3S
Ref	8.33×10^{-1}	3.06×10^{-1}	8.83×10^{-1}	1.99×10^{-1}	9.48×10^{-1}
1	2.89×10^{-1}	1.77×10^{-1}	3.34×10^{-1}	1.47×10^{-1}	9.10×10^{-1}
2	3.97×10^{-1}	1.96×10^{-1}	5.89×10^{-1}	1.68×10^{-1}	9.39×10^{-1}
3	4.05×10^{-1}	1.57×10^{-1}	3.56×10^{-1}	1.53×10^{-1}	8.23×10^{-1}
4	3.30×10^{-1}	1.80×10^{-1}	4.88×10^{-1}	1.74×10^{-1}	9.19×10^{-1}
5	4.72×10^{-1}	2.43×10^{-1}	6.15×10^{-1}	1.85×10^{-1}	9.14×10^{-1}
6	6.59×10^{-1}	3.10×10^{-1}	8.14×10^{-1}	1.93×10^{-1}	8.81×10^{-1}
7	6.59×10^{-1}	2.70×10^{-1}	6.42×10^{-1}	1.85×10^{-1}	8.95×10^{-1}
8	7.65×10^{-1}	3.30×10^{-1}	7.95×10^{-1}	2.05×10^{-1}	9.16×10^{-1}
Average	4.97×10^{-1}	2.33×10^{-1}	5.79×10^{-1}	1.76×10^{-1}	9.00×10^{-1}
STD	1.75×10^{-1}	6.54×10^{-2}	1.80×10^{-1}	1.99×10^{-2}	3.55×10^{-2}

The E-nose average response values of the ten sensors for the commercial samples are presented in Table A6. The standard deviation is also shown. All results were normalized to the most significant value response for each sensor.

Table A6. Average response values for E-nose sensing and standard deviation for three commercial rum samples.

Samples	W1C	W5S	W3C	W6S	W5C
Bacardí Añejo	5.26×10^{-5}	1.13×10^{-1}	6.29×10^{-5}	2.27×10^{-1}	6.79×10^{-5}
Ron Medellín Añejo	7.59×10^{-5}	1.45×10^{-2}	8.07×10^{-5}	1.80×10^{-1}	8.45×10^{-5}
Barceló Imperial	7.62×10^{-5}	1.26×10^{-2}	8.13×10^{-5}	1.72×10^{-1}	8.52×10^{-5}
Average	6.82×10^{-5}	4.67×10^{-2}	7.49×10^{-5}	1.93×10^{-1}	7.92×10^{-5}
STD	1.35×10^{-5}	5.74×10^{-2}	1.05×10^{-5}	2.97×10^{-2}	9.81×10^{-6}
Samples	W1S	W1W	W2S	W2W	W3S
Bacardí Añejo	8.54×10^{-1}	2.89×10^{-1}	7.82×10^{-1}	1.76×10^{-1}	9.41×10^{-1}
Ron Medellín Añejo	2.10×10^{-1}	1.30×10^{-1}	2.16×10^{-1}	1.31×10^{-1}	8.73×10^{-1}
Barceló Imperial	2.03×10^{-1}	9.69×10^{-2}	2.20×10^{-1}	1.26×10^{-1}	8.75×10^{-1}
Average	4.23×10^{-1}	1.72×10^{-1}	4.06×10^{-1}	1.44×10^{-1}	8.96×10^{-1}
STD	3.74×10^{-1}	1.03×10^{-1}	3.26×10^{-1}	2.74×10^{-2}	3.87×10^{-2}

Aroma patrons are compared between the average ultrasound-aged samples and commercial rum brand samples to identify their aroma profiles regarding the aroma descriptors. These aroma profiles are shown in Table A7.

Table A7. Average aroma profiles for ultrasound-aged samples and commercial samples.

Category	Aroma	Aged	Commercial
Baking Spices	Nutmeg		
Baking Spices	Vanilla	High	High
Baking Spices	Cinnamon		Medium
Baking Spices	Cloves		Medium
Berry fruit	Blackberry		Medium
Berry fruit	Raspberry		
Berry fruit	Strawberry		Low
Chocolate	Chocolate	High	Medium
Citrus	Lime	High	High
Citrus	Lemon		Medium
Dried fruit	Prune		
Dried fruit	Dried Apricot	Medium	Low
Earthy	Tobacco	Medium	High
Fruity	Cherry	Low	High
Fruity	Plum	High	High
Fruity	Blackcurrant		
Fruity	Apple	Low	Low
Fruity	Green Apple	Medium	High
Fruity	Pear	High	High
Medicinal	Tar	Medium	High
Medicinal	Licorice	Medium	Medium
Nutty	Almond	Low	Medium
Nutty	Hazelnut	Medium	High
Off flavor	Onion		
Off flavor	Nail Polish Remover		
Off flavor	Vinegar	Medium	High
Roasted	Smoke	High	Medium
Roasted	Bacon		Medium
Roasted	Butter		Medium
Roasted	Toast	High	Medium
Spicy	Pepper		
Spicy	Anise		Low
Spicy	Fennel	Low	Low
Spicy	Coffee	Medium	Low
Sugar	Honey	High	Medium
Sugar	Caramel	Medium	Medium
Tropical fruit	Pineapple	Medium	High
Tropical fruit	Banana		
Tropical fruit	Coconut		
Vegetal	Rose		Low
Vegetal	Cut Grass		
Vegetal	Mint		
Vegetal	Black Tea	High	High
Wood	Sandalwood		
Wood	Oak		
Wood	Pine	High	High
Wood	Cedar		

Previous authors have reported important flavor volatiles in rum, relating the compounds responsible for the aroma to their corresponding sensory attributes. The key odorants in aged rum are highlighted. These results are shown in Table A8.

Table A8. Important flavor volatiles in rum. Adapted from [23].

Compound	Sensory Attribute(s)
Esters	
Ethyl acetate	Pineapple, fruity, solvent
Ethyl butanoate *	Pineapple
Ethyl formate	Ethereal, fruity, rum-like
Isoamyl acetate *	Banana, estery
Ethyl propionate	Ethereal, fruity, rum-like
Ethyl butyrate	Ethereal, fruity, apple, buttery
Ethyl valerate	Fruity, apple
Ethyl hexanoate *	Fruity, winey, apple, banana, pineapple
Ethyl octanoate *	Sweet, cognac, apricot
Ethyl decanoate *	Sweet, fatty, nut, winey-cognac
Ethyl dodecanoate	Oily, fatty, floral
Ethyl hexadecanoate	Waxy, fruity, creamy, greasy, oily, balsamic
Ethyl linoleate	Fatty, fruity, oily
Ethyl lactate	Buttery, butterscotch, fruity, artificial strawberry, raspberry, perfumed
Methyl salicylate	Wintergreen
Ethyl 2-methylpropanoate *	Ethereal, sweet, alcoholic
Ethyl 2-methylbutanoate *	Fresh, fruity
2-phenylethyl acetate *	Rose, honey, raspberry
Ketones	
(E)- β -damascenone *	Fruity, apple
Acids	
Acetic acid	Vinegar
Hexanoic acid	Goaty, fatty, vegetable oil
Butyric acid	Rancid
Valeric acid	Strong, pungent, cheesy
Propionic acid	Vinegar, milky
Octanoic acid	Goaty, fatty, vegetable oil, wet dog
Alcohols	
1-propanol	Alcohol
1-butanol	Malty, solvent-like
Isobutanol	Malty, alcohol
2-methyl-1-butanol	Fish oil, green, malt, onion, wine
3-methyl-1-butanol (isoamylacetate)	Fruity at low levels, unpleasant at high
2-phenylethanol *	Floral
Acetals	
1,1-diethoxyethane *	Ethereal, green, nutty, earthy, sweet, vegetable
Phenolic compounds	
2-methoxyphenol (guaiacol) *	Smoky
4-methylguaiacol	Vanilla, clove
4-Ethylguaiacol *	Bacon, clove, phenolic
4-Propylguaiacol *	Spicy, sweet
Eugenol *	Clove-like
Vanillin *	Vanilla
Lactones	
Cis-oak lactone *	Sweet, spicy, coconut, vanilla
γ -nonalactone *	Coconut, creamy, waxy, sweet, buttery, oily

* Key odorants in aged rum.

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