American Journal of

Food Technology

ISSN 1557-4571

Academic
Journals Inc.

www.academicjournals.com

OPEN ACCESS American Journal of Food Technology

ISSN 1557-4571
DOI: 10.3923/ajft.2017.311.321

Research Article
Optimization of the Degree of Toasting, Concentration and Aging Time of *Quercus mongolica* (Chinese Oak) for Jujube Brandy Aging

[1,2]Ya-nan Xia, [1]Ran Suo, [1]Haoran Wang , [3]Stefan Cerbin and [1]Jie Wang

[1]College of Food Science and Technology, Agricultural University of Hebei, 071000 Baoding, China
[2]College of Food Science and Engineering, Inner Mongolia Agricultural University, 010000 Hohhot, China
[3]Department of Horticulture, Michigan State University, 48864 Lansing, USA

Abstract
Background and Objective: Faced with the insufficient resources and high cost of French and American oak, new and innovative aging methods for brandy aging should be explored. The effect of the aging of *Quercus mongolica* (Chinese oak) on jujube brandy was determined. Aging parameters were optimized to provide a new aging method with Chinese oak. **Methodology:** Degree of toasting, concentration and aging time of Chinese oak was tested by evaluating color, acids, esters, phenols and sensory characteristics by significance difference analysis and the aging effect of Chinese oak was evaluated by flavor compounds analysis. Data were analyzed by SPSS. **Results:** Esters and phenols (gallic and benzoic acids) were advantageous and superior to other parameters in Chinese oak aging and changed from 0.178 g L^{-1} and 13.4 mg L^{-1} to 0.487 g L^{-1} and 132.2 mg L^{-1}, respectively. Phenols (peak values of 169.824 mg L^{-1}) of jujube brandy increased with increasing degree of toasting of Chinese oak. Heavy toasting of with Chinese oak is not suitable for the esterification of jujube brandy. Esters and phenols increased first and then peaked at 15 g L^{-1} Chinese oak after 45 days. Under optimal conditions, 62 kinds of aroma components were detected in jujube brandy aged with Chinese oak (91.022 mg L^{-1}), which included 34 esters (80.209 mg L^{-1}). A total of 23 aroma compounds were added to the brandy after aging with Chinese oak. Chinese oak remarkably contributed in improving the aroma composition of jujube brandy. **Conclusion:** Optimized aging conditions of Chinese oak for jujube brandy were 15 g L^{-1} of medium-toasted oak aged for at least 45 days.

Key words: Chinese oak, jujube brandy, optimization, toasting, concentration, aging time

Citation: Ya-nan Xia, Ran Suo, Haoran Wang , Stefan Cerbin and Jie Wang, 2017. Optimization of the degree of toasting, concentration and aging time of *Quercus mongolica* (Chinese oak) for jujube brandy aging. Am. J. Food Technol., 12: 311-321.

Corresponding Author: Jie Wang, College of Food Science and Technology, Agricultural University of Hebei, 071000 Baoding, China Tel: +86-13131262819

Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The aging of distillates in oak barrels was critical to the development of quality spirits[1]. During aging, Brandy de Jerez and the wood of the cask undergo slow physicochemical changes[2]. Unique oak aroma can be formed in brandy during oak aging process, flavor and color can be enhanced due to the reaction with the tannin, lignin and other ingredients in oak, taste can also be improved by increased phenolic content and reduced astringency[1]. Several reactions result in considerably increased polyphenolic[3,4] and volatile[5,6] contents of the brandy during aging.

American and French oak are the commonly used types of wood for brandy aging[7,8]. However, new choices and innovative aging methods for brandy aging should be explored because of the insufficient resources and high cost of the aforementioned wood[9,10].

Quercus mongolica, also known as Chinese oak, is distributed in Dongbei province, Eastern Inner Mongolia and North Hebei. Chinese oak grows under bright light, which does not impose serious demand for survival. This species is one of the main broad-leaved tree species in Northeast China. Chinese oak belongs to the same family as French oak. Thus, these types of oak are remarkably similar in appearance, fruit, texture and aroma. These characteristics indicate that Chinese oak can be used to age brandy. The appearance of Chinese oak provides a new pathway for brandy aging. This process has significant great market prospect because of the moderate price of Chinese oak, which was important for the development of brandy.

This study was performed to provide a new choice of wood, in particular, Chinese oak, to age brandy. Thus, the effect of aging of Chinese oak was tested first and then the aging parameters, namely, degree of toasting, concentration and aging time, were optimized. The results may provide useful practical information to the jujube brandy industry to accelerate brandy aging.

MATERIALS AND METHODS

Samples

Oak block: The blocks (18 mm long, 8 mm wide and 3-4 mm thick) of Chinese oak and French oak were provided by Jinzi Wood Company (Chengde, Hebei, China, 117°51' E, 40°57' N). Four degrees of toasting, namely, untoasted, slightly toasted, medium toasted and heavily toasted, were considered. Toasting was performed at 160-170°C for 0, 20-25, 45-50 and 85-90 min, respectively.

Jujube brandy: Commercial jujube brandy produced in 2014 using jujube from Fuping (Hebei, China), which lies at 38°9' N and 113°45' E, was used for the present study. Jujube brandy was produced using the usual wine-making process in China, that was, by solid fermentation, solid distillation and aging (average alcohol content of 50%)[11,12]. Wine samples (100 mL) were bottled in glass bottles (250 mL) and stored at 15°C in the laboratory in September 2015. Oak (9 g L^{-1}) was added and contact time was 2 weeks.

Experimental designing: The study was performed in 2015. The aging effects of Chinese oak on color, acids, esters, phenols and sensory characteristics were evaluated. Then, degree of toasting (untoasted, lightly toasted, medium toasted and heavily toasted), amount of oak additive (0, 6, 9, 12, 15 and 18 g L^{-1}), aging time (15, 30, 45, 60, 75 and 90 days) and ultrasound treatment time (0, 10, 20, 30, 40, 50 and 60 min) were optimized with the evaluated values of color, acids, esters, phenols and sensory characteristics. Finally, the aging effect of Chinese oak was evaluated by flavor compounds analysis.

Standard sample: Twelve monophenols, namely, gallic acid (98%), protocatechuic acid (97%), catechin ($\geq$97%), vanillic acid ($\geq$97%), syringic acid ($\geq$95%), 4-coumaric ($\geq$98%), syringaldehyde (98%), ferulic acid (99%), guaiacol (98%), benzoic acid (98%), salicylic ($\geq$99%) and quercetin ($\geq$95%), were used in the study. The alcohols included methanol ($\geq$99.9%), isopropanol ($\geq$99.5%), 1-propanol ($\geq$999.9%), ethyl acetate ($\geq$99.8%), isobutanol ($\geq$99.5%), 1-butanol ($\geq$99%), isoamyl alcohol ($\geq$98%), 1-pentanol ($\geq$99%), furfural ($\geq$98%) and 2-phenethyl alcohol ($\geq$99%). All chemicals were purchased from Sigma (USA).

Analysis of pH, chroma(OD), acids, esters and phenols: Total acidity, pH (Mettler Toledo Sevencompac pH/ion S220, Santiago, Chile) and esters were evaluated following the OIV official analytical methods[13]. Chroma (OD) was evaluated using the absorbance measured at 420 nm. Total phenolic contents (TP) were determined by the Folin-Ciocalteu method. With hydration gallic acid as criterion, linear equation was $Y = 0.0125C+0.02$, $R^2 = 0.9999$, with unit of mg L^{-1}.

HPLC analysis of monophenols: Twelve monophenols of jujube brandy were detected by HPLC (Waters HPLC, UV detector) and quantified using an external standard. A ZORBAX Eclipse XDB-C18 column (250×4.6 mm, 5 µm) was used for separation with flow rate of 1.0 mL min^{-1}, column temperature of 30°C, detection wavelength of 280 nm, injection volume of 20 µL and acetonitrile-acetic acid-water solution as mobile phase. Mobile phase proportions were as follows: 0-5 min: 97% A (acetic acid-water) and 3% B (acetonitrile), 5-15min: 90% A and 10% B, 15-25 min: 85% A and 15% B, 25-35 min: 75% A and 25% B, 35-40 min: 60% A and 30% B and after 40 min: 100% A and 0% B. The samples were prepared as follows. Samples of 25 mL were concentrated by a rotary evaporator and then dissolved by chromatographically pure methanol to 10 mL. Linear range and detection limit standards of the 12 monophenols are shown in Table 1.

GC-FID analysis of higher alcohols: Higher alcohols, methanol and ethyl acetate of jujube brandy were detected by GC-FID (Agilent 7890A Gas Chromatograph, Santa Clara, USA) and quantified using an external standard. A DB-FFAP column (60 m×0.25 mm ID and 0.25 µm film thickness) was used for the separation. The working parameters were injector, temperature of 220°C and FID temperature of 230°C. The initial temperature was 40°C for 6 min, which was increased to 120°C at 5°C min^{-1}. The temperature was further raised to 210°C at 50°C min^{-1}. The carrier gas had a flow rate of 2.0 mL min^{-1}. Samples of 1.0 µL were injected using the split mode of 25:1.

Sensory analysis: Ten trained judges were selected for the descriptive analysis. Quantitative descriptive analysis was conducted using five sensory terms, namely, color, clarity, aroma, taste and specificity. The selected attributes were written on tasting cards and panelists were asked to rank each descriptor on a 15 cm unstructured scale (from unnoticeable to very strong). Average scores for all descriptors were calculated. Discriminant sensory evaluation was performed through triangular tests to assess any significant differences caused by the varied treatments in each wine.

SPME-GC-MS parameters: Jujube brandy was diluted by distilled water (10% alcohol content). Sodium chloride (1 g) was added to 7.5 mL of sample solution in a 20 mL volume sealed glass vial. The sample was extracted at 40°C for 40 min with 50/30 µm DVB/CAR/PDMS fiber and then analyzed using GC-MS.

Flavor compounds of jujube brandy were detected by GC-MS. The contents of these compounds were quantified using an internal standard (3-octanol, 99%, Sigma-Aldrich). Wine volatile compounds were analyzed using the Agilent 5975 mass spectrometer coupled to an Agilent 7890A gas chromatograph (Agilent, Santa Clara, USA). A DB-WAX column (60 m×0.25 mm ID and 0.25 µm film thickness) was used for separation. The working parameters were injector temperature of 250°C, EI source of 230°C, MS Quad of 150°C and transfer line of 250°C. The initial temperature was 50°C for 3 min, which was increased to 80°C at 3°C min^{-1}. The temperature was further increased to 230°C at 5°C min^{-1} and maintained at 230°C for 6 min. The carrier gas had a flow rate of 1.0 mL min^{-1}. Samples were injected using the splitless mode. A mass range of 50-550 m/z was recorded at one scan/sec.

Statistical analysis: Each sample was tested thrice. Data were analyzed by Statistical Package for the Social Science (SPSS) version 17.0 for windows (SPSS Inc., Chicago, IL). Prominent difference levels included 0.05(a) for significant differences ($p<0.05$) and 0.01 (A) as highly significant differences ($p<0.01$).

RESULTS AND DISCUSSION

Analysis of the aging effect of Chinese oak

Analysis of pH, chroma (OD), acids, esters and phenols: Several important differences were found among jujube brandies matured with and without Chinese oak. Remarkable increase in concentration was found in jujube brandies matured with Chinese oak. The brandies changed from colorless to golden yellow. Acids and esters are important flavor contents of jujube brandy such that increasing these contents can effectively improve the flavor quality of jujube brandy. Phenol is closely related to human health[14,15]. Therefore, considerable increase in phenols can effectively improve the nutritional value of jujube brandy[15,16] (Table 2).

Analysis of monophenols: Phenols have a significant role in the sensory and nutritional characteristics of wines and these characteristics affect the organoleptic profile and exhibits positive effects on health[17]. Remarkable increase in the number of types of monophenols, except for TP, was found. Gallic and benzoic acids had the highest contents at 27 and 28% of TP, respectively. Similar findings have been reported in previous studies[18]. Syringic acid had the lowest content of only 4.4% (Table 1). Compared with previous study, the concentrations of ferulic (7.05 µg mL^{-1}) and salicylic acid (3.87 µg mL^{-1}) of jujube brandy were obviously higher than corresponding values (0.310 and 0.116 µg mL^{-1}) in Apulian Italian wines. Phenols apparently cause the advantage of Chinese oak aging[19].

Table 1: Regression equation, linear range and detection limit of 12 monophenols

Monophenols	Standard curves	Correlation coefficient (r)	Linear range (mg kg^{-1})	Retention time (min^{-1})	Detection limit (mg kg^{-1})
Gallic acid	Y = 5.90e+004X-1.03e+005	0.999587	5.02-100.33	4.620	0.084
Protocatechuic acid	Y = 3.34e+004X-4.05e+004	0.999841	4.98-99.67	7.564	0.032
Catechin	Y = 1.41e+004X-2.78e+004	0.999642	5.04-100.83	10.584	0.035
Vanillic acid	Y = 4.71e+004X-3.84e+004	0.999747	4.96-99.50	13.146	0.028
Syringic acid	Y = 7.17e+004X-6.11e+004	0.999744	5.075-99.5	14.122	0.052
4-coumaric	Y = 1.51e+005X-1.28e+005	0.999772	5.03-100.67	19.009	0.032
Syringaldehyde	Y = 5.39e+004X-4.95e+004	0.999793	5.07-101.33	20.074	0.187
Ferulic acid	Y = 8.08e+004X-9.32e+004	0.999834	4.99-99.83	21.882	0.094
Guaiacol	Y = 3.25e+004X-9.70e+003	0.999453	4.99-99.83	24.101	0.098
Benzoic acid	Y = 1.06e+004X-8.67e+003	0.999704	5.03-100.5	26.167	0.075
Salicylic	Y = 1.47e+004X-1.38e+004	0.999770	4.98-99.50	27.967	0.216
Quercetin	Y = 2.79e+00X-2.99e+004	0.999762	5.01-100.17	33.086	0.046

Table 2: Changes of jujube brandy with Chinese oak blocks

Sample	Chroma (OD)	pH	Acid (g L^{-1})	Esters (g L^{-1})	Phenols (mg L^{-1})	-	-
1	0.682	4.30	0.821	0.487	132.2	-	-
2	0.529	4.15	0.815	0.178	13.4	-	-
Phenol acids	Gallic acid	Vanillic acid	Syringic acid	Syringaldehyde	Ferulic acid	Benzoic acid	Salicylic
1	11.13	3.33	1.78	2.29	7.05	11.27	3.87
2	-	-	-	-	-	-	-
Higher alcohols	Methanol	N-propanol	Isobutanol	N-butanol	Isoamyl alcohol	N-pentanol	Ethyl acetate
1	1.10	0.24	0.19	0.02	0.66	0.13	0.24
2	1.12	0.24	0.19	0.02	0.64	0.13	0.21
Sensory	Color (5)	Clarity (5)	Aroma (30)	Taste (40)	Specificity (20)	Total (100)	
1	4	5	25	36	19	89	
2	3	5	23	30	18	79	

1: Jujube brandy matures with Chinese oak, 2: Blank jujube brandy, -: Means not found

Analysis of higher alcohol: No obvious change appeared in the concentration of methanol and higher alcohols, except for ethyl acetate, with 17% increase with Chinese oak blocks. This result was consistent with the above total esters. Therefore, aging with Chinese oak blocks had no effect on the alcohol content of jujube brandy. Given that ethyl acetate is an important flavor composition of jujube brandy, aging with Chinese oak blocks had important effect on enhancing the flavor of jujube brandy (Table 2).

Analysis of sensory characteristics: Color, clarity, aroma, taste and specificity improved in the brandy aged with Chinese oak blocks, with higher sensory evaluation score of 89, compared with wine matured without blocks. Aging with Chinese oak blocks resulted in jujube brandy with mellow and sweet taste, no alcoholic irritation and beautiful color (light golden yellow), light wood flavor and harmonious palate (Table 2).

Optimization of aging parameters on jujube brandy with Chinese Oak

Selection of toast degree on jujube brandy with Chinese Oak: The pH of jujube brandy decreased as the degree of toast of Chinese oak increased ($p<0.01$). Chroma and phenols of jujube brandy increased with increasing degree of toasting of Chinese oak ($p<0.01$), that is, from 0.627 (untoasted) to 1.088 (heavily toasted, 173%) and from 82.696-169.824 mg L^{-1} (205%), respectively. Esters decreased from 1.095-0.850 g L^{-1}, which indicated that heavy toasting is not suitable for Chinese oak to mature jujube brandy (Fig. 1a-b). Medium toasted Chinese oak had the highest content of acids (0.9 g L^{-1}, $p<0.05$). Therefore, medium toasted Chinese oak was the best choice to mature jujube brandy based on pH, chroma (OD), acids, esters and phenols.

Total content of higher alcohols increased first, peaked (1.255 g L^{-1}) at light toast ($p<0.01$) and then declined. Methanol, isoamyl alcohol and N-pentanol followed the same trend. Ethyl acetate also followed a similar trend but peaked (0.547 g L^{-1}) at medium toast ($p<0.01$). No significant relations were found between the degree of toasting of Chinese oak and content of N-propanol, isobutanol and N-butanol (Table 3).

TP increased with increasing degree of toasting ($p<0.01$). Monophenols, except for vanillic and ferulic acids, followed the same trend. Heavily toasted and medium toasted Chinese oak obviously showed higher content of these phenols ($p<0.05$). Vanillic and ferulic acids increased first,

Table 3: Effects on methanol, higher alcohols and ethyl acetate of jujube brandy with different roast degree of Chinese oak blocks

Sample	Non roast	Light roast	Medium roast	Heavy roast
Methanol	1.006 ± 0.005^{Aa}	1.024 ± 0.004^{Ab}	1.008 ± 0.007^{Aa}	1.000 ± 0.012^{Aa}
N-propanol	0.234 ± 0.001^{Aa}	0.238 ± 0.004^{Aa}	0.244 ± 0.001^{B}	0.232 ± 0.001^{Aa}
Isobutanol	0.186 ± 0.002^{Aa}	0.199 ± 0.010^{Aa}	0.189 ± 0.001^{Aa}	0.183 ± 0.007^{Aa}
N-butanol	0.020 ± 0.001^{Aa}	0.021 ± 0.002^{Aa}	0.021 ± 0.001^{Aa}	0.021 ± 0.001^{Aa}
Isoamyl alcohol	0.600 ± 0.004^{Aa}	0.674 ± 0.020^{B}	0.607 ± 0.004^{Aa}	0.601 ± 0.001^{Aa}
N-pentanol	0.1115 ± 0.001^{Aa}	0.122 ± 0.004^{Ab}	0.115 ± 0.001^{Aa}	0.114 ± 0.001^{Aa}
Total	1.157 ± 0.001^{Aa}	1.254 ± 0.009^{B}	1.176 ± 0.010^{Ab}	1.152 ± 0.006^{Aa}
Ethyl acetate	0.479 ± 0.007^{Aa}	0.493 ± 0.008^{Aab}	0.547 ± 0.007^{B}	0.505 ± 0.005^{Ab}
Color (5)	3	3	5	4
Clarity (5)	3	5	5	5
Aroma (30)	22	23	26	25
Taste (40)	32	34	37	35
Specificity (20)	16	18	18	17
Total (100)	76	83	91	86

Values are Mean±SD, ^{A,B,C,D}Means high significance difference ($p<0.01$), ^{a,b,c,d}Means significance difference ($p<0.05$), Unit: g L^{-1}

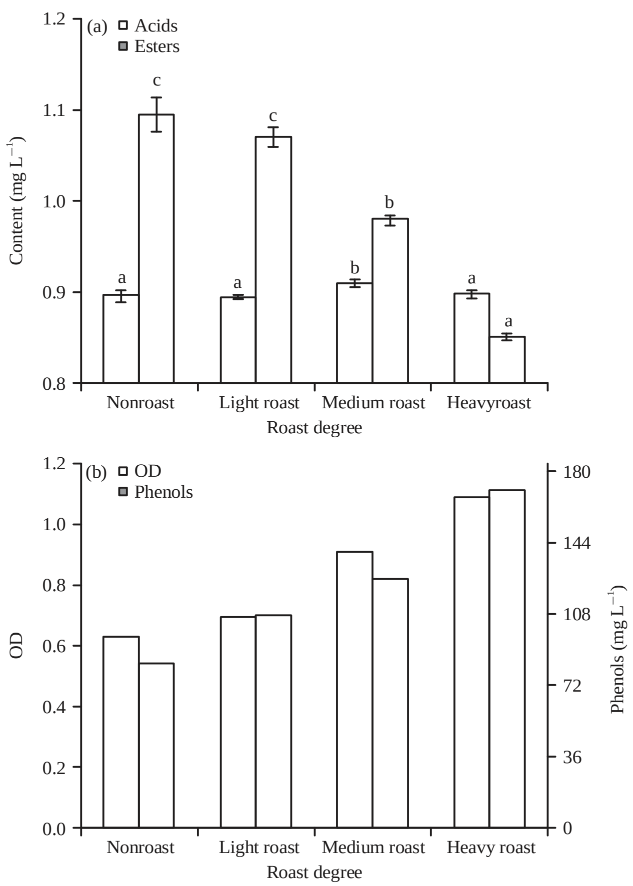


Fig. 1(a-b): Effects on (a) Acids, esters and (b) OD, phenols of jujube brandy with different toast degree of Chinese oak blocks

Values are Mean±SD, ^{a,b,c,d}Means significance difference ($p<0.05$)

peaked at medium toast ($p<0.05$) at 5.229 and 11.851 mg L^{-1}, respectively and then declined (Fig. 2a-d).

Sensory characteristics of jujube brandy increased first, peaked (91) at medium toast and then declined with increasing degree of toasting of Chinese oak. Brandy with medium toasted oak was better than other brandies based on the different sensory criteria Brandy with fresh oak showed the lowest score (76) and evaluation (Table 3).

Therefore, medium toasted Chinese oak is suitable for jujube brandy aging. Prior to toasting, each wood showed different and specific polyphenolic profiles, with qualitative and quantitative differences[20]. Toasting notably changed these profiles proportional to the toasting intensity and resulted in minor differences among species in toasted woods but phenolic markers were also found in the toasted woods[21].

Selection of the amount of Chinese oak additive: The pH of jujube brandy with different degrees of toasted oak showed no clear regularity but the highest pH level (4.48) was found when 15 g L^{-1} Chinese oak was used ($p<0.05$). Remarkable increase in chroma and phenols were observed until 15 g L^{-1} Chinese oak ($p<0.05$), then a slight increase in these parameters was observed. Almost thrice as much phenol was observed at 15 g L^{-1} Chinese oak, compared with that at 6 g L^{-1} Chinese oak. Acids basically remained stable (approximately 0.9 g L^{-1}) with increase in amount of Chinese oak additive. Esters increased first, peaked (1.030 g L^{-1}) at amount of 15 g L^{-1} Chinese oak ($p<0.05$) and then declined (Fig. 3a-b). Therefore, 15 g L^{-1} was the suitable amount of Chinese oak additive for jujube brandy.

The amount of Chinese oak showed no remarkable relation with methanol and higher alcohols of jujube brandy (Fig. 4a-d). Ethyl acetate showed the highest content (0.407 g L^{-1}, $p<0.05$) at 15 g L^{-1} Chinese oak.

The types of monophenols increased with increase in Chinese oak. No monophenols could be found without oak. Catechins and quercetin were not detected at 6 and 9 g L^{-1} Chinese oak. Vanillic acid and 4-coumaric increased first,

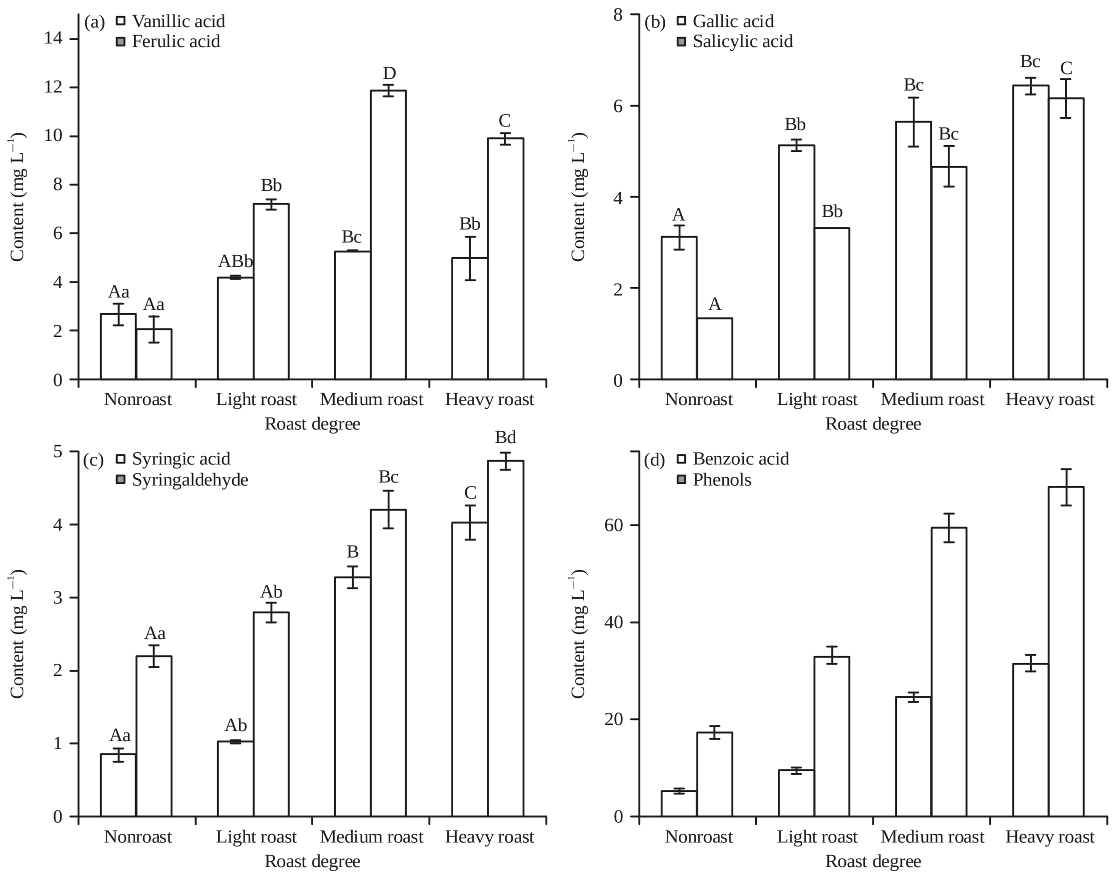


Fig. 2(a-d): Effects on (a) Vanillic acid, ferulic acid, (b) Gallic acid, salicylic acid, (c) Syringic acid, syringaldehyde and (d) Benzoic acid, phenols of jujube brandy with different toast degree of Chinese oak blocks

Values are Mean±SD, ^{a,b,c,d}Means significance difference ($p<0.05$), ^{A,B,C,D}Means significance difference ($p<0.01$)

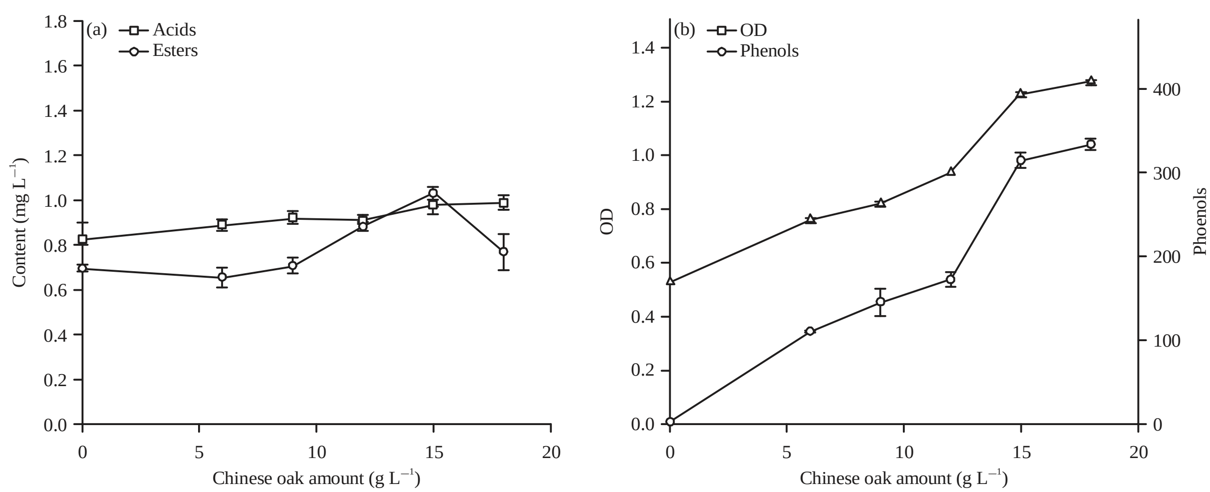


Fig. 3(a-b): Effects on (a) Acids, esters and (b) OD and phenols of jujube brandy with different amount of Chinese oak

Values are Mean±SD

peaked (11.236 and 1.848 g L^{-1}, respectively) at 15 g L^{-1} Chinese oak and then declined. Other monophenols increased with the amount of oak additive and then especially sharply increased after 12 g L^{-1} (Table 4).

Evaluation score of jujube brandy increased, with highest scores of 95 and 93 were obtained at 12 and 15 g L^{-1} Chinese oak and then declined. At 12 and 15 g L^{-1} Chinese oak, brandy was the best at all evaluation respects, while 6 and 9 g L^{-1}

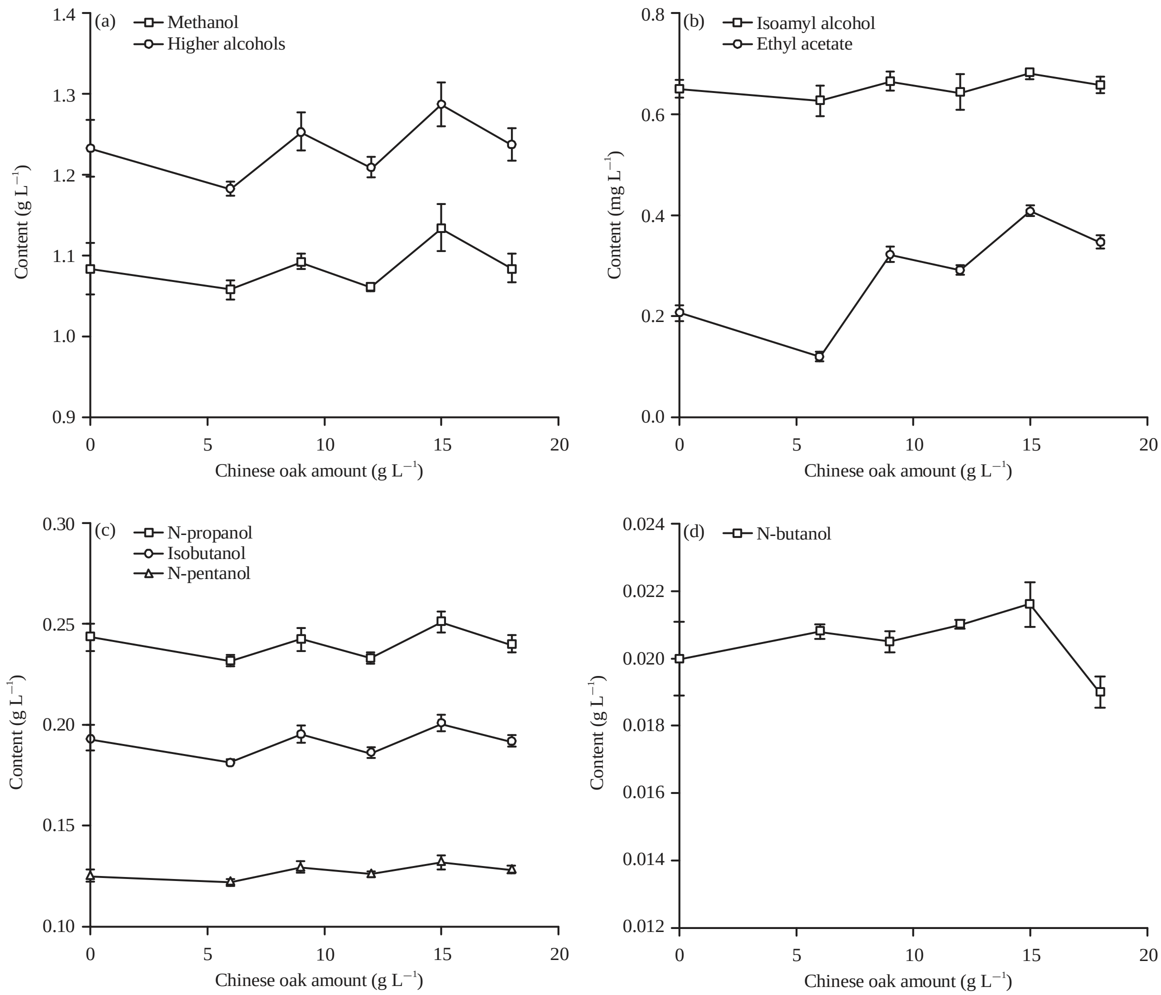


Fig. 4(a-d): Effects on (a) Methanol, higher alcohols, (b) Isoamyl alcohol, ethyl acetate, (c) N-propanol, Isobutanol, N-pentanol and (d) N-butanol of jujube brandy with different amount of Chinese oak
Values are Mean±SD

Table 4: Effects on monophenol of jujube brandy with different amount of Chinese oak

Phenols	0 (g L^{-1})	6 (g L^{-1})	9 (g L^{-1})	12 (g L^{-1})	15 (g L^{-1})	18 (g L^{-1})
Gallic acid	-	11.93 ± 1.12^{Bb}	5.53 ± 0.09^{Aa}	5.53 ± 0.01^{Aa}	18.90 ± 0.04^{C}	11.34 ± 1.07^{Bb}
Protocatechuic acid	-	2.62 ± 0.15^{Aa}	2.70 ± 0.01^{Aa}	3.00 ± 0.13^{ABa}	3.69 ± 0.36^{BCb}	4.29 ± 0.10^{Cc}
Catechin	-	-	-	6.52 ± 0.19^{A}	11.56 ± 0.60^{Bb}	10.58 ± 0.07^{Bb}
Vanillic acid	-	3.77 ± 0.62^{Aa}	3.88 ± 0.33^{Aa}	4.61 ± 0.90^{Aa}	11.24 ± 0.36^{Ab}	10.99 ± 0.99^{Ab}
Syringic acid	-	1.95 ± 0.23^{Aa}	1.98 ± 0.10^{Aa}	2.60 ± 0.22^{Ab}	3.98 ± 0.17^{Bc}	4.29 ± 0.13^{Bc}
4-coumaric	-	1.15 ± 0.18^{Aa}	1.50 ± 0.01^{Aab}	1.73 ± 0.08^{Aab}	1.85 ± 0.45^{Ab}	1.24 ± 0.19^{Aab}
Syringaldehyde	-	2.49 ± 0.28^{ABa}	2.09 ± 0.01^{Aa}	3.03 ± 0.19^{ABab}	4.05 ± 0.04^{BCb}	5.55 ± 0.86^{Cc}
Ferulic acid	-	7.28 ± 0.32^{ABa}	3.44 ± 0.22^{Aa}	18.96 ± 0.96^{ABa}	13.80 ± 1.09^{ABa}	37.93 ± 2.09^{Bb}
Guaiacol	-	2.36 ± 0.13^{Ab}	2.09 ± 0.01^{Aab}	1.56 ± 0.18^{Aa}	3.85 ± 0.06^{Bc}	3.67 ± 0.39^{Bc}
Benzoic acid	-	11.58 ± 0.43^{ABab}	9.16 ± 0.10^{Aa}	13.98 ± 1.02^{Bb}	24.22 ± 0.19^{C}	31.74 ± 1.48^{D}
Salicylic	-	4.42 ± 0.78^{Ab}	3.31 ± 0.01^{Aa}	4.33 ± 0.11^{Aab}	4.30 ± 0.09^{Aab}	6.14 ± 0.42^{B}
Quercetin	-	-	-	2.24 ± 0.15^{Aa}	2.30 ± 0.01^{Aa}	2.42 ± 0.04^{Aa}
Total	-	49.53 ± 4.23^{ABa}	35.67 ± 1.31^{Aa}	68.07 ± 2.32^{Bb}	103.73 ± 5.97^{Cc}	130.17 ± 3.26^{Cd}
Color (5)	2	4.00	4.00	5.00	5.00	4.00
Clarity (5)	5	5.00	5.00	5.00	5.00	5.00
Aroma (30)	23	25.00	26.00	27.00	27.00	24.00
Taste (40)	32	35.00	36.00	37.00	38.00	36.00
Specificity (20)	17	17.00	17.00	18.00	18.00	16.00
Total (100)	79	86.00	88.00	92.00	93.00	85.00

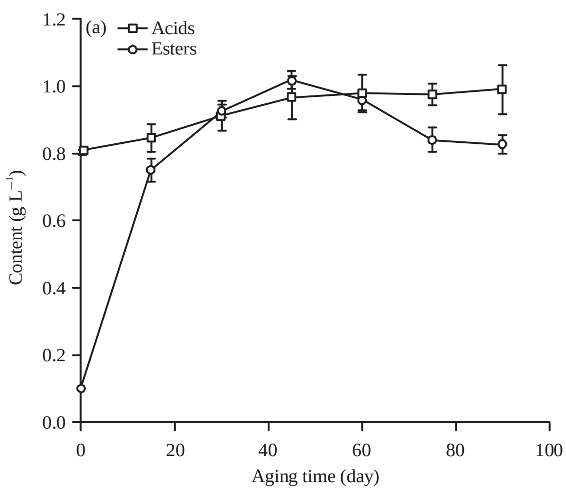
Values are Mean±SD, ^{A,B,C,D}Means highly significance difference ($p<0.01$), ^{a,b,c,d}Means significance difference ($p<0.05$)

Chinese oak resulted in light yellow color, disharmonious flavor and spicy taste, while 18 g L^{-1} Chinese oak resulted in heavy yellow color, heavy wood taste and after-taste (Table 4). The increasing demand for wood for barrel-making, in addition to the rapid extension of alternative aging system, have led to the exploration of utilizing Chinese oak. Oak chips, segments, staves and other oak alternatives have been proposed for wine aging. These materials would be obtained from wooden remnants from barrel-making[22].

Selection of aging time on jujube brandy with Chinese oak: The pH of jujube brandy increased first, then remained stable at approximately 4.6 but declined at 45 days. Chroma and phenols remarkably increased in the first 15 days and subsequently slightly increased. A slight increase in acids, from 0.8-1.0 was observed. Esters increased first, peaked (1.021 g L^{-1}) at 45 days ($p<0.05$) and then declined (Fig. 5a-b).

Evaluation score increased first, peaked (94) at 45 days and then decreased with prolonged aging time. Jujube brandy with aging time of 45 days showed genuine gold yellow, bright attractive, full, harmony taste and wood-flavor. This brandy was better than other brandies (Fig. 6).

Analysis of flavor compounds: Under optimal conditions, 62 kinds of aroma components were detected in jujube brandy aged with Chinese oak (91.022 mg L^{-1}), containing 34 esters (80.209 mg L^{-1}). Then, 39 kinds of aroma components were found in jujube brandy under natural aging (56.671 mg L^{-1}), containing 17 esters (49.094 mg L^{-1}). Chinese oak was found to be very advantageous in improving the flavor composition and content of jujube brandy, especially esters. Except for acids, the concentrations of all aroma compounds of jujube brandy aged with Chinese oak were higher than the naturally aged group (Table 5).



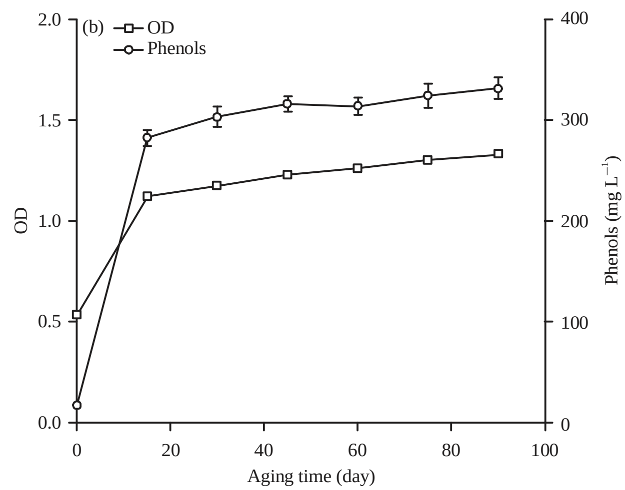


Fig. 5(a-b): Effects on (a) Acids, esters and (b) OD and phenols of jujube brandy during aging
Values are Mean±SD

Table 5: Comparison of aroma compounds of jujube brandy with and without Chinese oak

Time (min^{-1})	Compounds	amu	Chinese oak	Non
Esters				
3.95	Butanoic acid, ethyl ester	116.08	0.488	-
5.81	1-Butanol, 3-methyl-, acetate	130.10	0.260	0.484
6.15	Pentanoic acid, ethyl ester	130.10	0.221	-
9.13	Hexanoic acid, ethyl ester	144.12	4.221	6.631
12.83	Heptanoic acid, ethyl ester	158.13	3.415	2.801
15.73	Hexanoic acid, butyl ester	172.15	0.091	-
16.27	Octanoic acid, ethyl ester	172.15	16.955	7.548
16.82	2-Heptenoic acid, ethyl ester, (E)-	156.12	0.124	-
17.11	Isopentyl hexanoate	186.16	0.468	-
17.89	7-Octenoic acid, ethyl ester	170.13	1.941	-
18.24	3-Octenoic acid, ethyl ester, (Z)-	170.13	0.628	-
19.22	Nonanoic acid, ethyl ester	186.16	4.031	2.282
19.93	Formic acid, octyl ester	158.13	0.487	-
21.69	2-Furancarboxylic acid, ethyl ester	140.05	0.090	-

Table 5: Continue

Time (min^{-1})	Compounds	amu	Chinese oak	Non
21.92	Decanoic acid, ethyl ester	200.18	25.771	17.132
22.52	Chloroacetic acid, nonyl ester	220.12	0.848	-
22.63	Benzoic acid, ethyl ester	150.07	3.514	1.398
23.18	Ethyl 9-decenoate	198.16	1.980	1.019
24.19	Acetic acid, phenylmethyl ester	150.07	0.071	0.115
24.37	Undecanoic acid, ethyl ester	214.19	1.108	0.867
25.19	Benzoic acid, 2-hydroxy-, methyl ester	152.05	0.151	-
25.48	Benzeneacetic acid, ethyl ester	164.08	0.258	0.222
25.75	Dodecanoic acid, methyl ester	214.19	0.571	0.208
26.13	Acetic acid, 2-phenylethyl ester	164.08	1.152	-
26.67	Dodecanoic acid, ethyl ester	228.21	8.405	7.411
27.02	Pentadecanoic acid, 3-methylbutyl ester	242.23	0.272	-
27.59	Benzenepropanoic acid, ethyl ester	178.10	1.227	0.803
27.78	Ethyl 9-hexadecenoate	282.26	0.697	0.039
28.70	Ethyl 5-methyl hexanoate	158.13	0.177	-
28.77	Ethyl tridecanoate	242.23	0.028	-
30.05	Methyl tetradecanoate	242.23	0.118	-
30.81	Tetradecanoic acid, ethyl ester	256.24	0.283	0.107
32.46	2-Propenoic acid, 3-phenyl-, ethyl ester, (E)-	176.08	0.036	-
34.62	Hexadecanoic acid, ethyl ester	284.27	0.122	0.027
Alcohols				
17.04	1-Heptanol	116.12	-	0.284
23.91	2-Dodecanol	186.20	0.664	0.486
25.01	1-Octanol, 2-butyl-	186.20	0.127	-
27.12	1-Undecanol	172.18	0.200	0.108
27.48	Benzyl Alcohol	108.06	-	0.125
28.17	Phenylethyl Alcohol	122.07	1.515	1.104
Acids				
17.22	Acetic acid	60.02	0.290	1.337
26.87	Heptanoic acid	130.10	-	0.529
31.19	Octanoic Acid	144.12	0.202	0.102
35.09	n-Decanoic acid	172.15	0.586	0.278
36.92	Undecanoic acid	186.16	0.026	-
38.67	Dodecanoic acid	200.18	0.730	0.294
Aldehyde and ketone				
14.78	2-Nonanone	142.14	0.257	0.130
15.11	Nonanal	142.14	0.753	-
16.20	2-Octenal, (E)-	126.10	0.130	-
17.43	Furfural	96.02	1.576	1.489
18.54	Ethanone, 1-(2-furanyl)-	110.04	0.098	-
18.92	Benzaldehyde	106.04	0.754	0.358
20.42	2-Furancarboxaldehyde, 5-methyl-	110.04	0.168	0.244
20.93	2-Undecanone	170.17	1.157	0.372
21.42	4-Hydroxy-2,4,5-trimethyl-2,5-cyclohexadien-1-one	152.08	0.112	-
21.57	Benzaldehyde, 4-methyl-	120.06	0.123	-
25.13	2H-1-Benzopyran-2-one, 3,4-dihydro-	148.05	0.129	-
26.91	5,9-Undecadien-2-one, 6,10-dimethyl-, (E)-	194.17	0.482	-
28.82	2(1H)-Naphthalenone, octahydro-4a,7,7-trimethyl-, cis-	194.17	0.076	0.074
30.49	2(3H)-Furanone, dihydro-5-pentyl-	156.12	0.059	0.052
Others				
26.35	Naphthalene, 1,2,3,4-tetrahydro-1,6-dimethyl-4-(1-methylethyl)-, (1S-cis)-	202.17	0.151	0.145
29.85	Naphthalene, 1,7-dimethyl-	156.09	0.166	-
34.09	Naphthalene, 1,6-dimethyl-4-(1-methylethyl)-	198.14	0.175	0.086
35.81	Phenol, 2,4-bis(1,1-dimethylethyl)-	206.17	0.092	0.064
43.80	Quinoline, 3-(methylthio)-	175.05	0.015	-

Unit: mg L^{-1}

The main aroma components of jujube brandy were decanoic acid ethyl ester, octanoic acid ethyl ester, dodecanoic acid ethyl ester, hexanoic acid ethyl ester, phenylethyl alcohol, acetic acid, n-decanoic acid, dodecanoic acid, furfural, benzaldehyde and 2-undecanone. After aging with Chinese oak, 23 aroma compounds were added,

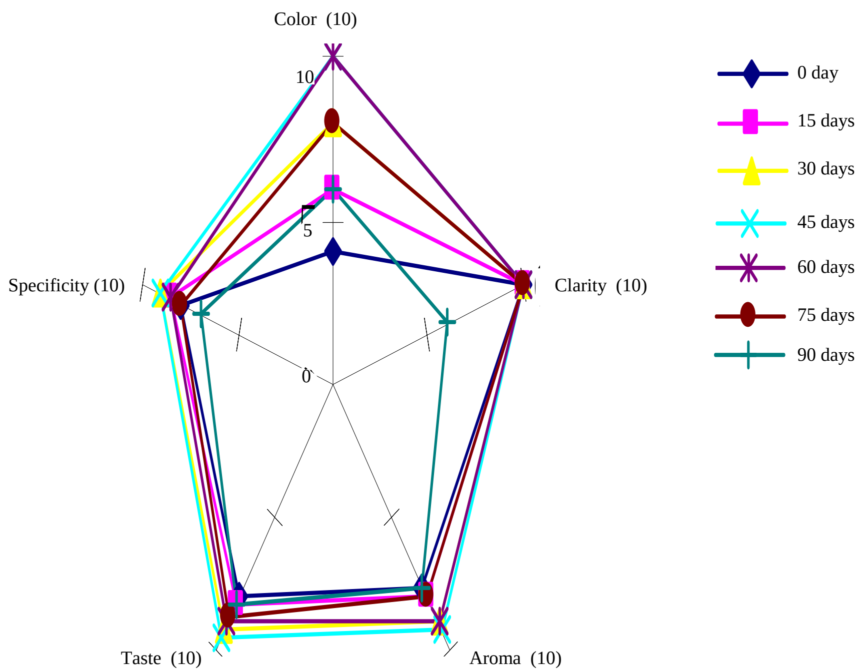


Fig. 6: Sensory evaluation result in optimized test of Chinese oak for aging

including several less common components, such as hexanoic acid butyl ester, formic acid octyl ester, 2-furancarboxylic acid ethyl ester, 1-(2-furanyl)-ethanone and 3-(methylthio)-quinoline. In addition to 1-butanol, 3-methyl-, acetate, hexanoic acid ethyl ester, acetic acid phenylmethyl ester and acetic acid, all aroma components of jujube brandy aged with Chinese oak were higher than those of the naturally aged group. Therefore, Chinese oak greatly contributed in improving the aroma composition of jujube brandy.

CONCLUSION

Aging with Chinese Oak enhanced the color, acids, esters, phenols and sensory characteristics of the aged brandy. Esters and phenols (gallic and benzoic acids), which were advantageous and superior than other parameters, for Chinese oak aging, changed from 0.178-0.487 g L^{-1} and from 13.4-132.2mg L^{-1}, respectively. Esters (peak value of 169.824 mg L^{-1}) and phenols (peak value of 205%) of jujube brandy increased with increasing degree of toasting Chinese oak. Esters and phenols increased first and peaked at 15 g L^{-1} Chinese oak for 45 days. Thus, optimized aging parameters of Chinese oak for jujube brandy were medium toasted oak at 15 g L^{-1} and at least 45 days of aging. Twenty-three aroma compounds were added after aging with Chinese oak, which remarkably contributed in improving the aroma composition of jujube brandy.

SIGNIFICANCE STATEMENTS

This study revealed that Chinese oak aging can be beneficial to improve the quality of jujube brandy. The results will facilitate the determination of unexplored critical areas in Chinese oak aging for liquors. Thus, a new theory on the aging of Chinese oak may be formulated.

ACKNOWLEDGEMENT

This study was supported by The National Natural Science Foundation of China, under the Project named "Study on the flavor character and its formation mechanism of Chinese date brandy (Founding No. 31371815)".

REFERENCES

1. Montero, C.M., M.D.C.R. Dodero, D.A.G. Sanchez and C.G. Barroso, 2005. Sugar contents of Brandy de Jerez during its aging. J. Agric. Food Chem., 53: 1058-1064.
2. Madrera, R.R., A.G. Hevia and B.S. Valles, 2013. Comparative study of two aging systems for cider brandy making. Changes in chemical composition. LWT-Food Sci. Technol., 54: 513-520.
3. Schwarz, M., M.C. Rodriguez, M. Sanchez, D.A. Guillen and C.G. Barroso, 2014. Development of an accelerated aging method for Brandy. LWT-Food Sci. Technol., 59: 108-114.

4. Rudnitskaya, A., L.M. Schmidtke, I. Delgadillo, A. Legin and G. Scollary, 2009. Study of the influence of micro-oxygenation and oak chip maceration on wine composition using an electronic tongue and chemical analysis. Anal. Chim. Acta, 642: 235-245.
5. Caldeira, I., O. Anjos, V. Portal, A.P. Belchior and S. Canas, 2010. Sensory and chemical modifications of wine-brandy aged with chestnut and oak wood fragments in comparison to wooden barrels. Analytica Chimica Acta, 660: 43-52.
6. Alanon, M.E., R. Schumacher, L. Castro-Vazquez, M.C. Diaz-Maroto, I. Hermosin-Gutierrez and M.S. Perez-Coello, 2013. Enological potential of chestnut wood for aging Tempranillo wines Part II: Phenolic compounds and chromatic characteristics. Food Res. Int., 51: 536-543.
7. Lago-Vanzela, E.S., D.P. Procopio, E.A.F. Fontes, A.M. Ramos and P.C. Stringheta *et al.*, 2014. Aging of red wines made from hybrid grape cv. BRS Violeta: Effects of accelerated aging conditions on phenolic composition, color and antioxidant activity. Food Res. Int., 56: 182-189.
8. Dodero, M.C.R., D.A.G. Sanchez, M.S. Rodriguez and C.G. Barroso, 2009. Phenolic compounds and furanic derivatives in the characterization and quality control of Brandy de Jerez. J. Agric. Food Chem., 58: 990-997.
9. Madrera, R.R., D.B. Gomis and J.J.M. Alonso, 2003. Influence of distillation system, oak wood type and aging time on composition of cider brandy in phenolic and furanic compounds. J. Agric. Food. Chem., 51: 7969-7973.
10. Figueiredo-Gonzalez, M., B. Cancho-Grande, J. Simal-Gandara, N. Teixeira, N. Mateus and V. de Freitas, 2014. The phenolic chemistry and spectrochemistry of red sweet wine-making and oak-aging. Food Chem., 152: 522-530.
11. Caldeira, I., A.P. Belchior, M.C. Clımaco and R.B. de Sousa, 2002. Aroma profile of Portuguese brandies aged in chestnut and oak woods. Anal. Chim. Acta, 458: 55-62.
12. Guerrero, E.D., M.J.C. Bastante, R.C. Mejias, R.N. Marin and C.G. Barroso, 2011. Characterization and differentiation of sherry brandies using their aromatic profile. J. Agric. Food Chem., 59: 2410-2415.
13. Perez-Caballero, V., F. Ayala, J.F. Echavarri and A.I. Negueruela, 2003. Proposal for a new standard OIV method for determination of chromatic characteristics of wine. Am. J. Enol. Vitic., 54: 59-62.
14. Liu, S.X., H.Y. Yang, S.Y. Li, J.Y. Zhang, T. Li, B.Q. Zhu and B.L. Zhang, 2015. Polyphenolic compositions and chromatic characteristics of bog bilberry syrup wines. Molecules, 20: 19865-19877.
15. Del Barrio-Galan, R., M. Medel-Maraboli and A. Pena-Neira, 2015. Effect of different aging techniques on the polysaccharide and phenolic composition and sensory characteristics of Syrah red wines fermented using different yeast strains. Food Chem., 179: 116-126.
16. Tao, Y., Z. Zhang and D.W. Sun, 2014. Experimental and modeling studies of ultrasound-assisted release of phenolics from oak chips into model wine. Ultrason. Sonochem., 21: 1839-1848.
17. Zhang, B., X.A. Zeng, W.T. Lin, D.W. Sun and J.L. Cai, 2013. Effects of electric field treatments on phenol compounds of brandy aging in oak barrels. Innov. Food Sci. Emerging Technol., 20: 106-114.
18. Osrecak, M., M. Karoglan and B. Kozina, 2016. Influence of leaf removal and reflective mulch on phenolic composition and antioxidant activity of Merlot, Teran and Plavac mali wines (*Vitis vinifera* L.). Scient. Hortic., 209: 261-269.
19. Barnaba, C., E. Dellacassa, G. Nicolini, T. Nardin, M. Malacarne and R. Larcher, 2016. Free and glycosylated simple phenol profiling in Apulian Italian wines. Food Chem., 206: 260-266.
20. Cerezo, A.B., M.A. Alvarez-Fernandez, R. Hornedo-Ortega, A.M. Troncoso and M.C. Garcia-Parrilla, 2014. Phenolic composition of vinegars over an accelerated aging process using different wood species (acacia, cherry, chestnut and oak): Effect of wood toasting. J. Agric. Food Chem., 62: 4369-4376.
21. Chatonnet, P. and J. Escobessa, 2007. Impact of toasting oak barrels on the presence of polycyclic aromatic hydrocarbons in wine. J. Agric. Food Chem., 55: 10351-10358.
22. De Simon, B.F., E. Cadahia, M. del Alamo and I. Nevares, 2010. Effect of size, seasoning and toasting in the volatile compounds in toasted oak wood and in a red wine treated with them. Anal. Chim. Acta, 660: 211-220.