

Study of ultrasonic wave treatments for accelerating the aging process in a rice alcoholic beverage

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Abstract

This research focussed on the accelerated aging of a rice alcoholic beverage by applying two different levels of ultrasonic power. The rice alcoholic beverage was fermented with *Saccharomyces sake*. The control treatment was aged for one year in fired clay containers (standard aging) while two other treatments simulated aging by using 20 kHz and 1.6 MHz ultrasonic waves, respectively (accelerated aging). Comparisons were made of alcohol content, titratable acidity value, gas chromatography measurements of volatile aroma compounds, sensory evaluation and time of aging. Results showed that the 20 kHz treatment influences rice alcoholic beverage aging better than the 1.6 MHz treatment. The 20 kHz ultrasonic wave treatment has potential as a good alternative method for aging the alcoholic beverage. Further studies are needed to elucidate the best power range of ultrasonic waves for aging alcoholic beverages made of various materials.

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1. Introduction

Moderate consumption of alcoholic beverages plays a role in life, inducing a mild euphoria and increasing longevity. Alcoholic beverages can be made from various materials, using several processes. Alcoholic beverage making, however, takes time, for fermenting and for aging. Fermenting produces alcohol, flavor, and taste; aging improves the sensory properties that make alcoholic beverages pleasurable (Sato, 1984). Alcoholic beverages usually have many flavors and other properties which each play a subtle, intertwined, yet important role in the tastes that humans prefer. Much research is published on the flavors, physical properties, chemical properties and constituents of alcoholic beverages made from various materials using different processes and aging

techniques (Chang, 2003; Chang & Chen, 2002; Cocito, Gaetano, & Delfini, 1995; Ho, Hogg, & Silva, 1999; Huang, 1987; Maduagwu, 1982; Masuda, Yamamoto, & Asakura, 1985; Simpson & Miller, 1983). There have been studies of alcoholic beverages aging using different chemical methods (Castellari, Matricardi, Arfelli, & Amati, 2000; Huang, 1980; Jan & Hsu, 1984; Maduagwu, 1982), but very little work was found (and/or was not written in English) dealing with accelerating alcoholic beverage aging by applying physical methods. There is a lack of verifiable data in this field. Therefore, a research study of ultrasonic wave treatments for reducing the time of the aging process is an important new contribution to the field.

In an ultrasonic wave process, high temperature and high pressure can be generated from circulated ultrasonic waves, causing chemical polymers to be broken into numerous mist particles (subparticles) and then recombined. In a review of sonoelectroanalysis, Saterlay and Compton (2000) concluded that ultrasonic

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equipment, working in the range of 20–100 kHz, is relatively inexpensive, readily available and is an enabling technology for electroanalysis. Lindley and Mason (1987) reported the types of ultrasound which are used in chemistry for effecting chemical reactivity, which were broadly divided in power between 20 and 100 kHz. They concluded that chemists are interested in ultrasound power because it provides a form of energy for the modification of chemical reactivity which is different from those normally used, such as heat, light and pressure. Furthermore, they indicated that ultrasound power produces its effects via cavitation bubbles. These bubbles are generated during the rarefaction cycle of the wave when the liquid structure is literally torn apart to form microbubbles which collapse in the compression cycle. Lindley and Mason (1987) further noted that pressures of hundreds of atmospheres and temperatures of thousands of degrees are generated during the collapse of the bubbles. Aroma extraction (Cocito et al., 1995), cell growth and ester formation (Matsuura, Hirotsune, Nunokawa, Satoh, & Honda, 1994), associated with ultrasound treatment, have been previously studied.

We hypothesized that the high temperature and high pressure generated from circulated ultrasonic waves might help speed up the aging process and give more flavor and/or taste to wine. By applying this physical acceleration aging method using ultrasonic waves, aging would be fulfilled within a shortened time and space would be saved as well. Our paper reports the effects of different powers (20 kHz and 1.6 MHz) of ultrasonic wave treatments on rice alcoholic beverage aging. The power range of 20 kHz, selected, was based on previously published data (Lindley & Mason, 1987; Saterlay & Compton, 2000) and a 1.6 MHz treatment as an additional variant with higher energy. We also hypothesized that ultrasound treatments by a power higher than 20 kHz would shorten the process of aging alcoholic beverage more than a lower power. Rice was chosen because it is relatively inexpensive, rich in starch (essential for alcohol fermentation techniques) and has fewer complex flavors and other properties than alcoholic beverages made from other materials which would complicate experimental analysis. In addition to sonication, rice alcoholic beverage was also aged for one year in fired clay containers as the comparative standard aging method or control. In this study, the length of time used for the standard aging method was set at one year because one year is the aging time for the market product of grain alcoholic beverage. In Asia, most grain alcoholic beverages sold in the market are conventionally aged only for one year (or less). This study should indicate whether the ultrasonic waves aging process has potential to produce an alcoholic beverage from rice comparable in quality to a conventionally aged alcoholic beverage from rice and whether a higher power is better than a lower power in accelerating aging.

2. Materials and methods

2.1. General

In this research, there were two ultrasonic wave aging treatments, one conventional aging group and one unaged (untreated) group. The two ultrasonic wave treatments on rice alcoholic beverages were: 20 kHz ultrasonic wave-treated, and 1.6 MHz ultrasonic wave-treated. The conventionally aged group was aged for one year.

2.2. Sample preparation of rice alcoholic beverage for ultrasonic wave treatments, for one year conventionally aged treatment and for unaged treatment

All the rice alcoholic beverage was made with a 1:1 ratio (by volume) of whole dry grain rice (Taiwanese PonLai, 14% water, dry basis) and water. Four litres of a 1:1 rice/water mixture were cooked at 125 °C for about 1 h and cooled to room temperature (25 °C). When this cooked rice was cooled to room temperature, four extra litres of water (pre-boiled and cooled to room temperature) were added and mixed well into the rice to liquefy the rice. This process was repeated 9 times in our experiment for accelerating the aging process and was repeated 3 times for the conventional aging process. *Aspergillus awamori* (CCRC 30891) was then inoculated into the rice in order to produce amyloglucosidase, glucoamylase, α -amylase, and β -glucuronidase to break down the starch polymers into smaller starch oligo-carbohydrate polymers and sugar monomers and also to liquefy the rice for further yet more complete, fermentation. About 1 h later, with the temperature remaining around 25 °C, *Saccharomyces sake* (CCRC 20262) was inoculated into the liquefied rice and kept at 25 °C for fermentation. The fermentation took place in six closed fired clay containers; three of the six were 30 l containers for the accelerated aging process and the other three were 10 l containers for the conventional one year aging process, which were vibrated 2–3 times a day. After the fermentation was completed (14–16 days, depending on the alcohol content, 18–20% alcohol in our experiment), the rice alcoholic beverage was collected using the 80 °C distillation method and was ready for the ultrasonic wave treatments of the accelerated aging process or for the one year standard aging process.

2.3. Additional sample preparation of one year conventionally aged rice alcoholic beverage

The standard, or conventionally aged, rice alcoholic beverage was aged by placing it in three 3-l fired clay containers for one year prior to conducting the accelerated aging. An aging time of one year is the standard for conventional aging of Asian market grain alcoholic

beverages, and analysis at that aging time was done. About one year later, the alcoholic beverage to be aged by accelerated methods, was aged by the two ultrasonic processes. Analysis was done immediately after the accelerated aging processes was completed.

2.4. Accelerated aging process for the 20 kHz and 1.6 MHz ultrasonic wave treatments

The accelerated-aged rice alcoholic beverage was aged about one year after the standard aging rice alcoholic beverage, so that analysis of the conventionally aged alcoholic beverage and accelerated-aged alcoholic beverage could take place concurrently. Moreover, the rice alcoholic beverage-making techniques for both conventional standard aging and accelerated aging, were as close to exactly the same as possible for all tested samples in our study in order to produce similar rice alcoholic beverage sample quality and to minimize bias. Analysis of the untreated rice alcoholic beverage (for accelerating aging process) was done immediately after the fermentations were completed, for which the rice alcoholic beverage samples were collected by using the 80 °C distillation method. This test was used as a check index of the start quality of the two different aging processes to assure the same quality level at commencement. Both the 20 kHz and 1.6 MHz ultrasonic wave setups were designed and self-made by a Professor of the Mechanical Engineering Department in our school. 20 kHz and 1.6 MHz ultrasonic waves generators were used to apply ultrasound energy to the rice alcoholic beverage. The two ultrasonic wave generators both had piezoelectric chips, which generated electrons. When the electrons accumulated they vibrated in terms of ultrasonic waves. The ultrasonic nozzle orifice for 20 kHz was 10 mm while that for 1.6 MHz was 4 mm. We chose 10 and 4 mm orifice nozzle because these nozzle amplitude energies were found, by trial, to fit our experimental needs in terms of atomizing the bulk alcoholic beverage liquids into a spray of smoke-like mist droplets to fulfill the ultrasonic process. The rice alcoholic beverage volume for one ultrasonic treatment run was 2 l and was triplicated. All the ultrasonic aging treatments were at 25 °C and were in a closed chamber to prevent evaporation loss of the rice alcoholic beverage and to assure the best collection of the misted rice alcoholic beverage. The rice alcoholic beverage was repeatedly treated, up to 16 times, for the 20 kHz process and up to 4 times for the 1.6 MHz ultrasonic wave process. The whole processing time of each was considered as the time of aging and was used for comparison to conventional aging. Samples of the treated rice alcoholic beverage were collected after 0 (untreated, immediately after evaporation), 4, 8, 12, and 16 treatments (repetitions) for the 20 kHz level of ultrasonic waves and were collected after 0 (untreated, immediately after

evaporation), 1, 2, 3, and 4 treatments (repetitions) for the 1.6 MHz level of ultrasonic waves.

2.5. Tests

2.5.1. Types of tests

Each sample was analyzed for alcohol content, titratable acidity, gas chromatography measurements of volatile aroma compounds, sensory evaluation and the time of aging. Each of the experimental treatments was replicated 3 times (e.g. three one year-aged alcoholic beverage samples were obtained from three different containers) for analysis, and the results were the averages of the three samples.

2.5.2. Titratable acidity

The titratable acidity was represented as acetic acid due to its domination in the rice alcoholic beverage. The titratable acidity was measured by the AOAC (1995) from the 1 N NaOH used. The titratable acidities of rice alcoholic beverage (fresh, immediately after distillation, for the accelerating aging process), one year conventionally aged rice alcoholic beverage (similar to the market aging technique) and each sample of all accelerated aging treatments (repetitions) 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 throughout the entire study.

2.5.3. Alcohol content

Alcohol was measured by a KYOTO, DA-310, electric specific gravity meter. Untreated rice alcoholic beverage (fresh, immediately after distillation, for accelerating aging process), one year conventionally aged rice alcoholic beverage (similar to the market aging technique) and each sample of all accelerated aging treatments (repetitions) were measured. Alcohol was then calculated from the gravity measurements by the AOAC method (1995). The alcohol content check gave an indication of possible alcohol loss in the process.

2.5.4. Gas chromatography measurements of volatile aroma compounds

Flavor compounds were analyzed by a WHIRLPOOL 5790 gas chromatograph with a glass column that was 1.8 m long, 2 mm thick, packed with 6.6% carbowax and 20 M/80–120 mesh carbopack B Aw using a flame ionization detector (FID). The carrier gas was nitrogen with 20 ml/min flow rate. The injection temperature was 150 °C and the FID temperature was 200 °C. The gas chromatograph temperature gradient started at 60 °C and remained at that level for 1 min. It then increased at 5 °C/min and reached 160 °C in 20 min where it stayed for 2 min. Each sample injection was 2 µl. Standards for the gas chromatograph were prepared as

follows: 0.5 ml of 5.14% acetaldehyde, 0.4 ml of 5.01% methanol, 2 ml of 5.0% ethyl acetate, 0.2 ml of 5% 1-propanol, 0.4 ml of 5% 2-methyl-1-propanol, and 1 ml of 2% 2,3-dimethyl-butanol were mixed with 40% ethanol to reach 100 ml in total.

2.5.5. Sensory evaluation

Sensory evaluations were done by 12 qualified and experienced alcoholic beverage sensory tasters using the Kramer preference priority organoleptic method which has a statistical significance of $\alpha = 5\%$ and had been applied widely for the purpose of alcoholic beverage tasting (Huang, 1984; Sato, 1988; Wang, 1980; Zhai, 2000). Each of the 12 tasters tasted the rice alcoholic beverage before any treatments (repetitions) and after each of the 4 (for 1.6 MHz) and each of the 16 (for 20 kHz) accelerated aging treatments (repetitions). The tasters ranked the treated rice alcoholic beverage against the rice alcoholic beverage conventionally aged for one year. They gave the treated rice alcoholic beverage a ranking of 1 (best) to 9 (worst). Generally, a quality conventionally aged rice alcoholic beverage would be ranked 1 on this scale. The results were then calculated and analyzed from the preference priorities of each taster and ranked from best to worst flavor.

2.5.6. Time of aging

Time of aging for the alcoholic beverages aged by the accelerated process was counted as the time needed to complete the whole ultrasonic waves aging process vs. the one year conventional aging process.

3. Results and discussion

3.1. Titratable acidity volume

The titratable acidity value in 20 kHz ultrasonic wave-treated rice alcoholic beverage decreased slightly as the number of treatments increased, thus giving a less sour taste (Table 1) and was preferred by the sensory

evaluators (Table 3). The titratable acidity in 1.6 MHz ultrasonic wave-treated rice alcoholic beverage remained the same as that of the untreated rice alcoholic beverage as the number of treatments (repetitions) increased (Table 2). The titratable acidity check of all studied samples also showed that it was in the normal range of rice alcoholic beverage, which indicated the right fermentation process and no contamination in our work over this entire study.

3.2. Alcohol content

The alcohol content in the rice alcoholic beverage aged with 20 kHz and 1.6 MHz ultrasonic waves decreased as treatment time increased, resulting in less burning flavor (Tables 1 and 2). The alcohol content (52%) of the samples treated 16 times by 20 kHz ultrasonic waves was a little lower than the alcohol content (54%) of samples aged by the standard one-year process. The alcohol content (45%) of the samples treated 4 times by 1.6 MHz ultrasonic waves was much lower than the alcohol content (52%) of the samples treated (repeated) 16 times by 20 kHz ultrasonic waves and the alcohol content (54%) of samples aged by the standard one-year process. The large quantity of alcohol lost in the rice alcoholic beverage aged by 1.6 MHz ultrasonic waves may be a problem.

3.3. Gas chromatography measurements of volatile aroma compounds

The gas chromatography analyses of volatile aroma compounds showed that the evaporating flavor compounds generally decreased slightly as the number of 20 kHz ultrasonic waves process treatments (repetitions) increased, but decreased to a lower level as the number of 1.6 MHz ultrasonic waves process treatments (repetitions) increased (Tables 1 and 2). Acetaldehyde has the lowest boiling point of rice alcoholic beverage flavor compounds and has an unpleasant offensive flavor. Thus, by decreasing the amount of acetaldehyde of 20

Table 1

The titratable acidity, alcohol content, acetaldehyde content, ethyl acetate content, and alcohol contents and standard deviations of 20 kHz ultrasonic wave-treated rice alcoholic beverage with different repetitions vs. one year conventionally aged rice alcoholic beverage, and fresh rice alcoholic beverage (untreated)

Number of accelerated aging repetitions	Titratable acidity (g/100 ml)	Alcohol (%)	Acetaldehyde (mg/l)	Ethyl acetate (mg/l)	Alcohols			
					Methanol (mg/l)	1-Propanol (mg/l)	2,3-Dimethyl-butanol (mg/l)	2-Methyl-1-propanol (mg/l)
0 (untreated)	0.012	56 ± 0.47	106 ± 12	103 ± 15	58 ± 3	161 ± 8	1130 ± 170	368 ± 27
4	0.011	54 ± 0.41	98 ± 11	104 ± 14	58 ± 3	155 ± 8	1088 ± 180	361 ± 23
8	0.011	53 ± 0.40	89 ± 13	105 ± 13	57 ± 3	150 ± 7	1019 ± 180	355 ± 25
12	0.010	53 ± 0.38	80 ± 9	109 ± 13	55 ± 2.5	147 ± 7	975 ± 176	344 ± 26
16	0.010	52 ± 0.53	70 ± 11	113 ± 13	54 ± 3	144 ± 7	913 ± 154	331 ± 22
One year conventionally aged rice alcoholic beverage	0.010	54 ± 0.41	90 ± 7	115 ± 11	54 ± 3	154 ± 6	933 ± 126	355 ± 24

Table 2

The titratable acidity, alcohol content, acetaldehyde content, ethyl acetate content, and other alcohol contents and standard deviation of 1.6 MHz ultrasonic wave-treated rice alcoholic beverage with different repetitions vs. one year conventionally aged rice alcoholic beverage and fresh rice alcoholic beverage (untreated)

Number of accelerated aging treatments	Titratable acidity (g/100 ml)	Alcohol (%)	Acetaldehyde (mg/l)	Ethyl acetate (mg/l)	Alcohols			
					Methanol (mg/l)	1-Propanol (mg/l)	2,3-Dimethylbutanol (mg/l)	2-Methyl-1-propanol (mg/l)
0 (untreated)	0.012	56 ± 0.47	106 ± 12	103 ± 15	58 ± 3	161 ± 8	1130 ± 170	368 ± 27
1	0.012	50 ± 0.61	88 ± 11	107 ± 14	54 ± 4	155 ± 9	1114 ± 187	358 ± 35
2	0.012	47 ± 0.78	77 ± 13	118 ± 14	51 ± 5	150 ± 8	1069 ± 176	350 ± 38
3	0.012	45 ± 0.56	65 ± 12	130 ± 12	50 ± 4	146 ± 9	1035 ± 190	344 ± 32
4	0.012	45 ± 0.44	54 ± 11	134 ± 12	50 ± 4	142 ± 9	990 ± 160	340 ± 35
One year conventionally aged rice alcoholic beverage	0.010	54 ± 0.41	90 ± 7	115 ± 11	54 ± 3	154 ± 6	933 ± 126	355 ± 24

Table 3

The results of sensory evaluation of ultrasonic wave treatments of rice alcoholic beverage vs. one year conventionally aged rice alcoholic beverage and fresh rice alcoholic beverage (untreated); the smaller the preference number, the better the sensory evaluation

	Untreated	Ultrasonic waves treatments								One year aged
		20 kHz		1.6 MHz		20 kHz		1.6 MHz		
		4 times	1 time	8 times	2 times	12 times	3 times	16 times	4 times	
Rank Summation of all tasters	108	93	15	44	40	26	62	12	84	12
Preference	9	8	2	5	4	3	6	1	7	1

kHz ultrasonic wave-treated rice alcoholic beverage, other pleasant flavor compounds became more prominent and made the 20 kHz ultrasonic wave-treated rice alcoholic beverage taste better (Tables 1 and 3). But, by decreasing the amount of acetaldehyde of the rice alcoholic beverage further than that 20 kHz ultrasonic wave-treated rice alcoholic beverage, both pleasant and unpleasant flavor compounds became more prominent which made the 1.6 MHz ultrasonic wave-treated rice alcoholic beverage have a more complicated taste (Tables 2 and 3). Ethyl acetate, which is one of the fragrant compounds in the alcoholic beverage, slightly increased in 20 kHz ultrasonic wave-treated rice alcoholic beverage but increased to a much higher level in the 1.6 MHz ultrasonic wave-treated rice alcoholic beverage (Tables 1 and 2), and thus made the 1.6 MHz ultrasonic wave-treated rice alcoholic beverage smell (not taste) much more fragrant than the 20 kHz ultrasonic wave-treated rice alcoholic beverage. The high alcohols (polyhydric alcohols) such as 1-propanol, 2-methyl-1-propanol, 2,3-dimethylbutanol and methanol, often have rice-oil flavors and cause a greasy feel in the mouth. When the levels of these decreased with the number of 20 kHz ultrasonic wave treatments (repetitions), combined with the decreased unpleasant acetaldehyde (Table 1), the ethyl acetate contribution to the taste was increased in the 20 kHz accelerated aging rice alcoholic beverage and made the alcoholic beverage

taste smoother and less greasy in the mouth (Table 3). When the polyol contents decreased to a lesser extent with the number of 1.6 MHz ultrasonic wave treatments (repetitions), in combination with the prominent increase of the ethyl acetate and prominent decrease of the acetaldehyde, the rice alcoholic beverage tasted spicy, tart, and unsmooth in the mouth (Table 2). The acceptability decreased as the treatments (repetitions) increased (Table 3).

3.4. Sensory evaluation

Human preference for certain alcoholic beverages is mostly based on odour, taste and tactile properties. Some are not detectable by gas chromatography analysis and thus human sensory evaluations are needed. Results of sensory evaluations showed that the taste of the 20 kHz ultrasonic wave-treated rice alcoholic beverage improved as the number of treatments (repetitions) increased (Table 3); while the taste of the 1.6 MHz ultrasonic wave-treated rice alcoholic beverage grew progressively worse as the number of treatments (repetitions) increased (Table 3). The rice alcoholic beverage treated 16 times by 20 kHz ultrasonic waves had a taste equivalent to conventional one year-aged rice alcoholic beverage (Table 3) while the rice alcoholic beverage that had one treatment by 1.6 MHz ultrasonic wave was rated nearly as good as the conventionally aged rice

alcoholic beverage, though it was rated progressively worse as the number of treatments (repetitions) increased (Table 3). The rice alcoholic beverage that was treated 4 times by 1.6 MHz ultrasonic waves had a taste less preferred than the rice alcoholic beverage that was treated 16 times by 20 kHz ultrasonic waves (Table 3). That is, as the number of the treatments (repetitions) by 1.6 MHz ultrasonic waves increased, the rice alcoholic beverage did not improve to reach the taste that was expected, though it was still preferred (to some extent) to the untreated rice alcoholic beverage. Aging of wine by ultrasonic wave treatment showed potential as an alternative method but was not applicable in all power ranges. However, further studies are needed to elucidate the best power range for alcoholic beverages made from various materials.

3.5. Time of aging

Rice alcoholic beverage could potentially be aged to a quality taste within one week by using 20 kHz ultrasonic waves and nearly as good quality within one day by using 1.6 MHz ultrasonic waves as compared to one year in the conventional process. The 1.6 MHz ultrasonic wave process for aging rice alcoholic beverage did not achieve the taste that was expected, though the beverage was preferred to the untreated rice alcoholic beverage (Table 3).

4. Conclusion

Ultrasonic wave treatment of rice alcoholic beverage was capable of aging alcoholic beverage to a similar quality of taste as the conventional method of maturation. The ultrasonic wave process is a much faster aging process for rice alcoholic beverages (one week or one day vs. one year), and the ultrasonic waves process can save precious room for a better utilization of space. However, not all kinds of ultrasonic waves are feasible. Further studies are needed to elucidate the best power range for aging alcoholic beverages made from various materials.

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