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Effects of degree of milling on japonica and glutinous rice used for *huangjiu* brewing physicochemical properties

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ABSTRACT

Rice is one of the main ingredients in *huangjiu* brewing. Studying the effects of dgree of milling (DOM) and variety on the nutritional composition, physical and chemical properties, and brewing characteristics of rice will help improve the quality of *huangjiu* brewing. In this study, the nutrient composition, gelatinization properties, cooking properties and brewing characteristics of *huangjiu* of japonica rice (JR) and glutinous rice (GR) with different DOM were studied using a fast viscosity analyzer, differential scanning calorimeter, and fast gas chromatography electron nose Heracles II combined with principal component analysis. The results showed that when the DOM is in the range of 5%–15%, the quality of rice wine was relatively superior and stable. And the flavour compounds of *huangjiu* brewed from JR and GR could be well clustered. The liquor, reducing sugar and amino acid nitrogen contents of *huangjiu* made from JR and GR were relatively high. The flavour compounds and content were relatively similar. This study effectively avoided the problems of low grain utilization and high energy consumption caused by excessive milling are, which caters to the industrial production of *huangjiu*. The results help to explore the relationship between DOM and *huangjiu* quality, and provide theoretical foundation and scientific guidance for *huangjiu* production.

1. Introduction

Huangjiu is known as one of the world's three ancient alcoholic beverages, in addition to beer and wine. Rice is the main raw material for *huangjiu* brewing and can be divided into three categories: indica rice (IR), japonica rice (JR), and glutinous rice (GR) (Gong et al., 2020). There are significant differences in the appearance quality and content of internal nutritional components among different rice varieties, which substantially effect the processing characteristics of rice for the brewing quality of *huangjiu* (Jasim et al., 2015). Most premium *huangjiu* utilize GR as the raw material, and JR and IR are the main raw materials for normal *huangjiu* (Jiao et al., 2017). However, due to its high amylose content, difficulties in cooking and squeezing, and low wine yield, IR has been gradually eliminated by the *huangjiu* brewery industry. Therefore, only JR and GR have been studied in this paper.

JR, and GR have similar structures but different nutrient components. Brown rice (i.e., paddy after hulling) has four layers from the outside to the inside: bran, aleurone, embryo, and endosperm. Starch is predominantly present in the inner layer (i.e., endosperm), while outer layers (such as bran, aleurone, and subaleurone) are rich in nutrients such as protein, lipid and minerals. Although organic acids and amino acids are key contributors to the flavour and quality of *huangjiu*, protein and lipids of the bran layer will introduce an odd flavour to it and reduce the quality of the finished products. In addition, the existence of the bran layer impedes the absorption and expansion of water during rice cooking, making it difficult for steam to pass through and further affecting the saccharification and fermentation process. The rich nutrients in the bran layer will promote the vigorous fermentation of microorganisms, hindering control of the fermentation mash temperature. Moreover, high temperature will facilitate the reproduction of acid-producing

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bacteria and raise the acidity of the fermentation mash (Qi et al., 2018). Therefore, appropriate milling of brown rice will help eliminate the abovementioned adverse effects and improve the quality of *huangjiu*.

Rice milling has been used in the brewing of Japanese sake for millennia. When producing sake, the average rice polishing rate will be controlled at approximately 73% (Iwano et al., 2004). Rice milling affects not only the whiteness of rice but also its nutrient components and physicochemical properties. With increasing DOM, the structure of rice gradually approaches the endosperm, and the proportion of starch increases, while nutrients such as protein and lipids are lost to varying degrees (Paiva et al., 2016). Roberts (2010) investigated the protein, lipid, dietary fibre, ash, and iron contents of four DOM samples of 0%, 3%, 6%, and 10% and demonstrated that nutrients in rice had a negative correlation with the DOM.

Nowadays, investment in high-end *huangjiu* is gradually increasing as the *huangjiu* industry is innovating in technology and production concepts. As one of the key ingredients in *huangjiu* brewing, the quality of rice plays a decisive role in the color, aroma and taste of *huangjiu*, as well as the size of the yield. Currently, there have been a large number of systematic studies on the development of food using different varieties of rice as raw materials (convenience rice, rice flour, rice cake, etc.), but there are few relevant studies on the production and brewing of huangjiu. Moreover, there are numerous studies on the effects of milling on the edible quality of rice, but few related studies on the production and brewing of *huangjiu*. Understanding the effects of DOM on the nutritional composition and physicochemical properties of rice, as well as the physicochemical index and flavor components of *huangjiu*, will help to explore the relationship between milling and *huangjiu* quality and provide ideas and references for *huangjiu* production.

2. Materials and methods

2.1. Materials and sample preparation

JR and GR (both brown rice) and wheat qu were provided by Jinfeng Wine Co., Ltd. in 2022. (Shanghai, China). α-amylase (44 000 U/g) and glucoamylase (130 000 U/g) were purchased from Suzhou Hongda Enzyme Co., Ltd. (Suzhou China). Total starch and Amylose/Amylopectin detection kits were purchased from Megazyme Co., Ltd. (Shanghai, China). Reagents such as sodium chloride, sodium hydroxide, dimethyl sulfoxide (DMSO), glacial acetic acid, sulfuric acid, 4-methyl-2-pentanol and ethanol were used as received. All the chemicals, reagents, and solvents were of analytical grade. The DOM of rice was determined based on the following equation:

$$\mathrm{DOM}(\%) = \left(1 - \frac{m_1}{m_2}\right) \times 100$$

where m_1 refers to the weight of the milled rice (g) and m_2 refers to the weight of the brown rice (g) (Sandhu et al., 2018). JR and GR with relatively uniform sizes were selected for milling, and the DOM value of the brown rice was recorded as 0%. One hundred grams of brown rice samples were weighed and milled by a rice polisher (RSKM5B, Suzhou, China). When the weight of the samples after milling was 98 g, the DOM value of the samples was recorded as 2%. Using the same method, experimental rice samples with DOM contents of 5%, 10%, 15%, 20%, 25%, 30%, 40%, 50%, 60%, 70%, 80% and 90% were prepared, as shown in Table S1. The ground nutrients are collected and used to make the mash (2.2.5). Samples were crushed by a high-speed grinder, collected using a 100-mesh filter, and stored in a dryer at room temperature.

2.2. Methods

2.2.1. Analysis of macronutrients components of rice samples

Standard AOAC methods (AOAC, 1998) were selected for the measurement of moisture, protein, and lipid content. The starch and amylose content (%, dry basis) was analysed using the Amylose/Amylopectin detection kit according to the assay procedure described in a previous report (Hao et al., 2018). Briefly, glucose oxidase/peroxidase (GOPOD) reagent was added to the samples after a series of pretreatments, and the samples were incubated at 40 °C for 20 min. The absorbance at 510 nm was measured by a UV spectrophotometer (A560, Shanghai, China), which was proportional to the total starch and amylose content in the samples.

2.2.2. Analysis of pasting properties

The pasting properties of rice were measured by a rapid viscosity analyser (RVA, RVA 4500, Hägersten, Sweden). According to the actual moisture content of the rice sample, a 2 g sample and a certain amount of water were weighed according to 14% moisture correction. The mixed samples were kept at 50 °C for 1 min, then heated from 50 °C to 95 °C at a rate of 12 °C/min, held at 95 °C for 2.5 min, and then cooled to 50 °C at the same rate. The heating process was accompanied by rotating stirring at a constant rate of 960 rpm for the first 10 s, followed by rotating stirring at a constant rate of 160 rpm until the end of the analysis. Each sample was tested three times.

2.2.3. Analysis of thermal properties

The thermal properties of the rice were measured according to the method described by Sandhu and Siroha (2017) using a differential scanning calorimeter (DSC, DSC3, Greifensee, Switzerland), which was equipped with a thermal analysis data station. A2 mg (dry basis) sample was accurately weighed and placed into a DSC aluminium tray, and deionized water was added at a ratio of 1:3 (W/W, sample/water). The DSC aluminium pan was then sealed and balanced overnight at room temperature, and an empty pan served as a reference. The pan was heated from 20 to 100 °C at a scanning rate of 10 °C/min. The reported transition temperatures reported were the onset temperature (T_0), peak temperature (T_P), conclusion temperature (T_c), and gelatinization enthalpy (ΔH).

2.2.4. Analysis of the cooking properties

The amount of rice used for analytics was 20 kg for each sample.

2.2.4.1. Determination of the delivery rate of rice. Rice (m_0) after soaking (3 d) was spread on a rice plant and steamed in a rice cabinet (CH-VI-6, Shanghai, China). The temperature of the rice cabinet was set to approximately 105 °C, and the time was controlled at approximately 30 min. After cooking, the rice was reweighed (m_1). The delivery rate of rice (%) = (m_1/m_0) × 100%.

2.2.4.2. Determination of volatile components of the cooked rice. The volatile components of the cooked rice were analysed using a previous method with modifications (Ma et al., 2020). The electronic nose (E-nose, Heracles II, Toulouse, France) was used in this method; it consisted of an automatic sampling device, a detector unit containing the sensor arrays, and the hardware and software required for pattern recognition. The weight of the samples required for analysis was 3 g. Hydrogen was applied as the carrier gas at a flow rate of 10 mL/min. The chromatographic programs are described as follows: initial temperature 50 °C (kept for 2 s), increased to 80 °C at 1 °C/s, and then ramped at 2 °C/s to 250 °C (kept for 60 s). The injected volume was 5 mL, and the injector temperature was 200 °C. The temperature of the two flame ionization detectors was 260 °C. For calibration, an alkane solution (C8–C40) was used to convert the retention time in Kovats indices and to identify the volatile compounds using AromaChemBase software (Alpha MOS S.A. Inc., France). Each sample was tested three times.

2.2.5. Process of huangjiu fermentation

JR and GR with different DOMs were soaked in water for 4 days.

After soaking, the soaked rice was steamed for subsequent *huangjiu* brewing processes, as described by Wei et al. (2017). At the end of fermentation, the whole mash was mixed and filtered, centrifuged (8000 rpm, 10 min) and stored at −20 °C for further analysis.

2.2.6. Analysis of physicochemical properties of huangjiu samples

The general physicochemical properties of *huangjiu*, such as alcohol content, pH, total acid and amino nitrogen, were determined according to the method described by Liu et al. (2019). Reducing sugars were determined by using the dinitro salicylic acid (DNS) method.

2.2.7. Determination of volatile flavour compounds of huangjiu samples

The main volatile flavour compounds in *huangjiu* were analysed by headspace-solid phase microextraction (HS-SPME) combined with gas chromatography−mass spectrometry (GC−MS) (Agilent 8890-7000D, Agilent, America). The main volatile flavour compounds were quantified using external standards, and 4-methyl-2-pentanol was selected as the internal standard for semiquantitative analysis to eliminate variations in extraction efficiency (Liu et al., 2019). The free amino acids were analysed with an automatic amino acid analyser (S7130; Sykam Company, Germany). The organic acids were detected by HPLC, as described by Gong et al. (2020).

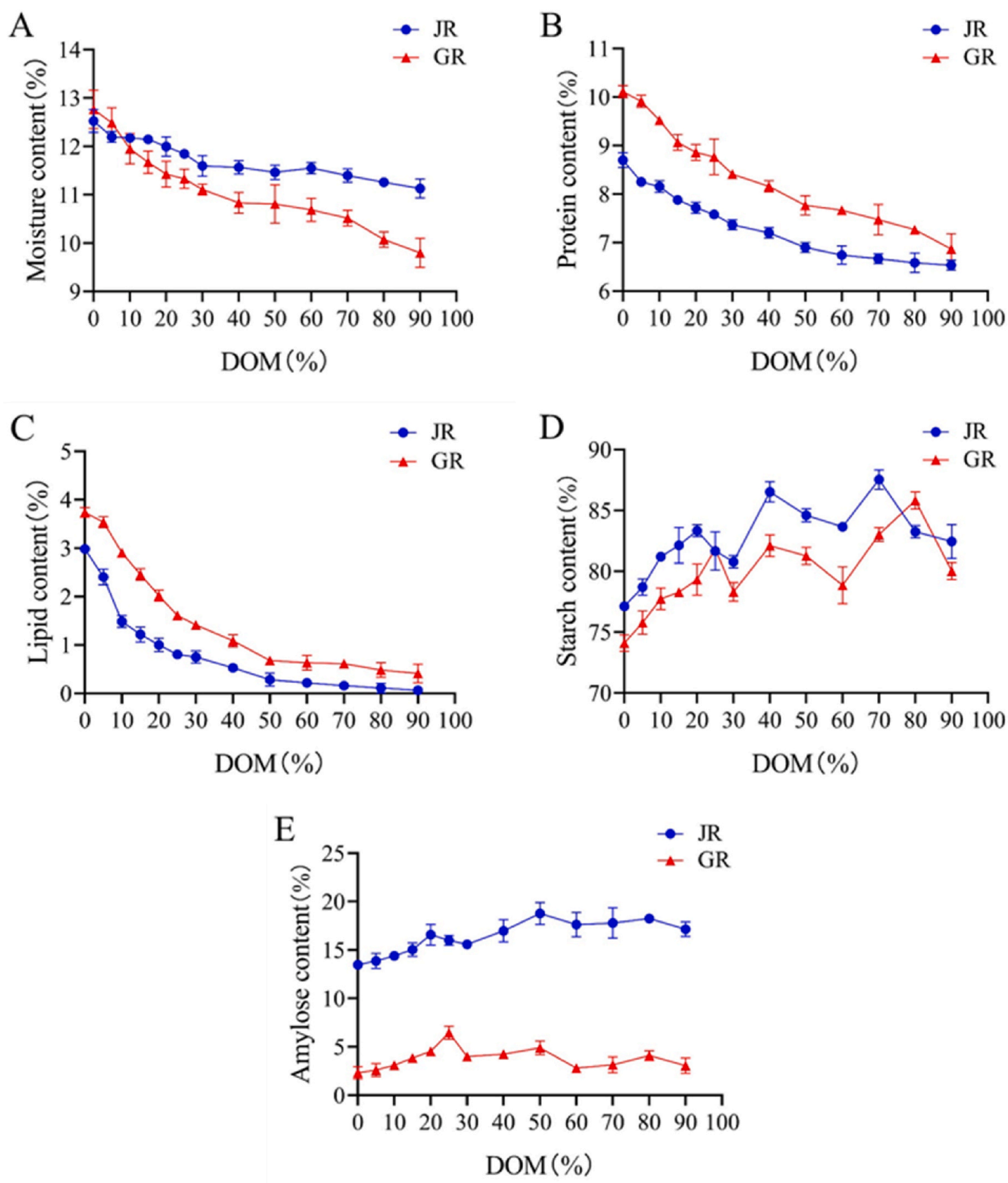

Fig. 1. Nutrient components of rice with different DOM, A: moisture content; B: protein content; C: lipid content; D: starch content; E: amylose content (based on dry basis).

2.3. Statistical analysis

The data reported in the tables were collected in triplicate, and the mean values and standard errors were determined for these experiments. Statistical significance was assessed with one-way analysis of variance using SPSS 19.0 software (Cary, NC). The results were considered statistically significant at $p < 0.05$. The PCA were carried out using SIMCA 15 (Umetrics, Sweden) choosed k to be 2 or 3. The data matrix of PCA has 7 columns and 43 rows, and the input matrix is automatically scaled.

3. Results and discussion

3.1. Nutrient components of rice with different DOMs

The nutrient components of rice with different DOMs (including the moisture, protein, lipid, total starch, and amylose contents) are shown in Fig. 1. The moisture, protein, and lipid contents of both JR and GR decreased with increasing DOM, which showed agreement with previous reports (Wang et al., 2021). The gradual decrease in moisture of rice might be attributed to the friction between the rice grains and the inner wall of the machine, which caused the evaporation of water inside the rice. The decrease in moisture content of GR was 1.5-fold higher than that of JR (2.97% and 1.17%, respectively) (Fig. 1A), which might be attributed to the relatively loose structure of the bran and aleurone layer in GR, facilitating the evaporation of water. The protein content rapidly decreased with the removal of the aleurone layer as the concentration of DOM increased. The protein content of GR was higher than that of JR without milling (10.11% and 8.70%, respectively), and both had a similar protein content (6.87% and 6.53%, respectively) when the DOM was 90% (Fig. 1B).

The lipid content for both JR and GR decreased with increasing DOM, which was reduced by 97.7% and 88.8%, respectively (Fig. 1C). In particular, the lipid contents of GR and JR sharply decreased from 0% DOM to 10% DOM, which might be attributed to the lipid mainly being distributed in the bran and aleurone layer, and they were gradually removed during the milling processes. Sandhu et al. (2018) discovered that the highest concentration of lipids was present in the pericarp followed by the testa and aleurone layers in short- and long-grain Indica rice cultivars. A thin layer corresponding to the layer removed by a DOM ranging from 0% to approximately 5% contained more than half of the lipid in the rice (Monks et al., 2013). This finding reflected that lipids were mainly distributed in the bran and aleurone layers in both cultivars, which were gradually removed from 0% to 10% DOM.

The total starch and amylose contents significantly varied for both cultivars. With the same DOM, the content of total starch and amylose in JR, especially the content of amylose, was generally higher than that in GR. The total starch content of JR varied from 77.12% to 87.53%, and the amylose content varied from 13.48% to 18.77%, among which DOM from 0 to 20% DOM gradually increased. Similarly, the total starch content of GR varied from 74.11% to 85.83%, and the amylose content varied from 2.34% to 4.35%, among which the DOM from 0 to 25% gradually increased (Fig. 1E). Studies have shown that starch is not vegetally present in the outer layer of the grain husk but is attributed to the breakdown of the endosperm during the milling process. With increasing milling, the bran and aleurone layers of rice, as well as the protein and lipids, were gradually removed (Wang et al., 2021), which promoted the dissolution of starch and amylose to varying degrees. Subsequently, the total starch and amylose content increased and decreased without significant regularity with a high DOM, which might be attributed to the uneven distribution of starch content in the endosperm of different cultivars of rice.

3.2. Pasting properties of rice with different DOMs

RVA was used to analyse the pasting properties of JR and GR; the pasting properties with different DOMs are shown in Table 1. The results showed that different DOM and cultivars of rice exhibited significant differences in pasting properties, indicating that pasting properties will also be greatly affected by their different compositions and structures, especially the size of starch granules and the proportion of amylose. Peak viscosity reflects the ability of starch granules to swell and combine with water in the cooking process. Breakdown is usually related to the tendency of starch granules to swell and rupture under high-temperature shear. In general, brown rice (DOM = 0%) from GR and JR showed lower peak viscosity, trough viscosity, breakdown, and final viscosity than milled rice from both cultivars. Moreover, these properties of both cultivars were highly significantly positively correlated with DOM (Tables S2 and S3) and significantly increased with increasing DOM. This increase might be attributed to the uneven removal of protein and lipids with increasing DOM, which promoted the dissolution of starch and amylose and increased the content of starch and amylose (Xu et al., 2021).

The pasting temperature of JR gradually decreased with increasing DOM, especially when the DOM was between 0% and 10% (Singh et al., 2014), which might be attributed to the increase in amylose and the decrease in nonstarch components such as protein and lipids with increasing DOM (Fig. 1). A recent study showed that the protein and lipid in rice and other grains would increase the resistance of starch granules to gelatinization and swelling (Rahimi et al., 2020). Furthermore, amylose-lipid complexes also enhanced the integrity of starch granules and protected them from destruction, which is attributed to the higher pasting temperature. The decrease in lipids resulted in the formation of amylose-lipid complexes, which correspondingly decreased the pasting temperature. However, the pasting temperature of GR remained unchanged at approximately 82.55 °C with increasing DOM, which might be attributed to a slight increase in the amylose content of GR, while the protein and lipid contents of GR gradually decreased.

The setback of starch reflects the difference in short-term ageing regeneration, which is not only closely related to the proportion and molecular structure of amylose and amylopectin but also influenced by other compounds, such as protein, lipid and nonstarch polysaccharides (Wang et al., 2021). The setback of both cultivars showed an opposite trend with increasing DOM. Setback for JR gradually decreased, which was possibly related to the gradual decrease in protein content (Likitwattanasade et al., 2010). While the setback for GR gradually increased and remained unchanged after 50% DOM, which might be attributed to the relatively low content of amylose in GR, the amylose content exhibited minimal change with increasing milling, especially when the grain husk and aleurone layers were completely removed at low DOM.

3.3. Thermal properties of rice with different DOMs

The thermal properties of JR and GR with different DOMs include T_0, T_P, Tc, and ΔH, as shown in Fig. 2. GR showed a higher T_0, T_P, Tc, and ΔH than JR with the same DOM, which showed that different cultivars of rice had a substantial influence on its thermal properties. The variation in thermal properties might be attributed to differences in nutrient composition (such as contents of starch, protein, and lipid) and granule structure (crystalline and amorphous content) (Bao et al., 2004). Varavinit et al. (2010) discovered that the positive correlation (r) between amylose content and T_0, T_P, and Tc was 0.84, 0.88, and 0.85, respectively. Correspondingly, the T_0, T_P, and Tc of high amylose were higher than those of middle, low, and glutinous rice starch, which is not consistent with our study (Table S2 and Table S3). This result might be attributed to the variety of protein and lipid contents with increasing DOM. Protein in rice showed a significant negative correlation with ΔH since protein is tightly packed around the starch, preventing water from entering. More energy is required to start starch gelatinization (Zhu et al., 2020). Lipids had a positive correlation with ΔH since lipids formed complexes with the amylose in the starch granules, which accelerated the disintegration of the amorphous zone of the starch

Table 1
Pasting properties of JR with different DOM.

	DOM (%)	DOM (%)	DOM (%)	DOM (%)	DOM (%)	DOM (%)	DOM (%)
JR	0	1427.50 ± 14.85^{k}	1039.00 ± 18.38^{k}	388.50 ± 3.54^{f}	92.70 ± 0.57^{a}	2483.00 ± 31.11^{k}	1444.00 ± 12.73^{a}
	5	1650.50 ± 9.19^{j}	1214.00 ± 16.97^{j}	436.50 ± 7.78^{e}	90.73 ± 0.04^{b}	2543.00 ± 8.49^{j}	1329.00 ± 8.49^{b}
	10	1845.00 ± 18.38^{i}	1397.50 ± 26.16^{i}	447.50 ± 7.78^{e}	73.05 ± 1.20^{c}	2595.00 ± 25.46^{i}	1197.50 ± 0.71^{c}
	15	1987.00 ± 2.83^{h}	1479.50 ± 7.78^{h}	507.50 ± 4.95^{d}	72.28 ± 0.04^{cd}	2642.50 ± 20.51^{h}	1163.00 ± 12.73^{c}
	20	2073.50 ± 4.95^{g}	1570.50 ± 30.41^{g}	503.00 ± 25.46^{d}	72.35 ± 0.00^{cd}	2686.50 ± 7.78^{g}	1116.00 ± 22.63^{d}
	25	2094.50 ± 16.26^{g}	1594.00 ± 12.73^{fg}	500.50 ± 28.99^{d}	72.33 ± 0.04^{cd}	2680.50 ± 2.12^{g}	1086.50 ± 10.61^{de}
	30	2159.50 ± 17.68^{f}	1632.00 ± 36.77^{f}	527.50 ± 27.58^{cd}	71.90 ± 0.57^{d}	2713.50 ± 16.26^{g}	1081.50 ± 20.51^{de}
	40	2250.00 ± 2.83^{e}	1712.50 ± 24.75^{e}	537.50 ± 27.58^{bcd}	71.48 ± 0.04^{de}	2762.50 ± 0.71^{f}	1050.00 ± 25.46^{ef}
	50	2302.50 ± 26.16^{d}	1740.50 ± 0.71^{de}	562.00 ± 25.46^{bc}	70.63 ± 0.04^{ef}	2796.00 ± 11.31^{e}	1055.50 ± 10.61^{ef}
	60	2407.00 ± 14.14^{c}	1779.50 ± 27.58^{d}	627.50 ± 13.44^{a}	70.63 ± 0.04^{ef}	2858.00 ± 12.73^{d}	1078.50 ± 14.85^{de}
	70	2433.00 ± 7.07^{c}	1855.00 ± 25.46^{c}	578.00 ± 18.38^{b}	69.83 ± 0.04^{f}	2891.00 ± 0.00^{c}	1036.00 ± 25.46^{f}
	80	2727.00 ± 8.49^{b}	2184.00 ± 15.56^{b}	543.00 ± 24.04^{bcd}	71.90 ± 0.64^{d}	3129.50 ± 0.71^{b}	945.50 ± 14.85^{g}
	90	2815.00 ± 21.21^{a}	2277.50 ± 43.13^{a}	537.50 ± 21.92^{bcd}	71.48 ± 0.04^{de}	3196.00 ± 16.97^{a}	918.50 ± 26.16^{g}
GR	0	1788.50 ± 12.02^{k}	1001.50 ± 0.71^{k}	787.00 ± 11.31^{j}	82.75 ± 0.00^{ab}	1233.00 ± 11.31^{l}	231.50 ± 10.61^{d}
	5	1955.00 ± 2.83^{j}	1105.00 ± 2.83^{j}	850.00 ± 0.00^{i}	82.70 ± 0.07^{ab}	1329.50 ± 6.36^{k}	224.50 ± 9.19^{d}
	10	2120.00 ± 1.41^{i}	1193.50 ± 6.37^{i}	926.50 ± 4.95^{h}	83.13 ± 0.53^{a}	1426.50 ± 3.54^{j}	233.00 ± 9.90^{d}
	15	2244.00 ± 12.76^{h}	1268.50 ± 10.61^{h}	975.50 ± 23.33^{g}	82.25 ± 0.64^{ab}	1521.50 ± 6.36^{i}	253.00 ± 4.24^{c}
GR	20	2352.00 ± 1.41^{g}	1323.00 ± 4.24^{g}	1029.00 ± 5.66^{f}	82.68 ± 0.04^{ab}	1584.00 ± 2.83^{h}	261.00 ± 7.07^{c}
	25	2443.00 ± 8.49^{f}	1375.50 ± 4.95^{f}	1067.50 ± 13.44^{ef}	82.35 ± 0.57^{ab}	1641.50 ± 0.71^{g}	266.00 ± 4.24^{c}
	30	2531.50 ± 13.44^{e}	1423.50 ± 0.71^{e}	1108.00 ± 14.14^{e}	82.28 ± 0.67^{ab}	1721.50 ± 0.71^{f}	298.00 ± 1.41^{b}
	40	2616.50 ± 30.41^{d}	1463.50 ± 3.54^{d}	1153.00 ± 26.87^{d}	82.68 ± 0.04^{ab}	1780.00 ± 12.73^{e}	316.50 ± 9.19^{b}
	50	2767.50 ± 44.55^{c}	1520.50 ± 6.36^{c}	1247.00 ± 38.18^{c}	81.85 ± 1.13^{b}	1883.50 ± 27.58^{d}	363.00 ± 21.21^{a}
	60	2893.50 ± 27.58^{b}	1592.50 ± 2.12^{b}	1301.00 ± 29.70^{b}	82.75 ± 0.00^{ab}	1960.50 ± 3.53^{c}	368.00 ± 5.66^{a}
	70	3016.50 ± 9.19^{a}	1645.00 ± 8.49^{a}	1371.50 ± 0.71^{a}	82.68 ± 0.04^{ab}	2008.50 ± 6.36^{b}	363.50 ± 2.12^{a}
	80	2866.50 ± 31.82^{b}	1579.00 ± 16.97^{b}	1287.50 ± 14.85^{bc}	82.70 ± 0.00^{ab}	1941.50 ± 9.19^{c}	362.50 ± 7.78^{a}
	90	3061.00 ± 35.36^{a}	1655.50 ± 12.02^{a}	1405.50 ± 23.33^{a}	82.33 ± 0.60^{ab}	2031.00 ± 5.66^{a}	375.50 ± 6.36^{a}

Results are expressed as mean values ± standard deviations of three independent experiments.
Means in a column with the different letter are significantly different ($P < 0.05$).

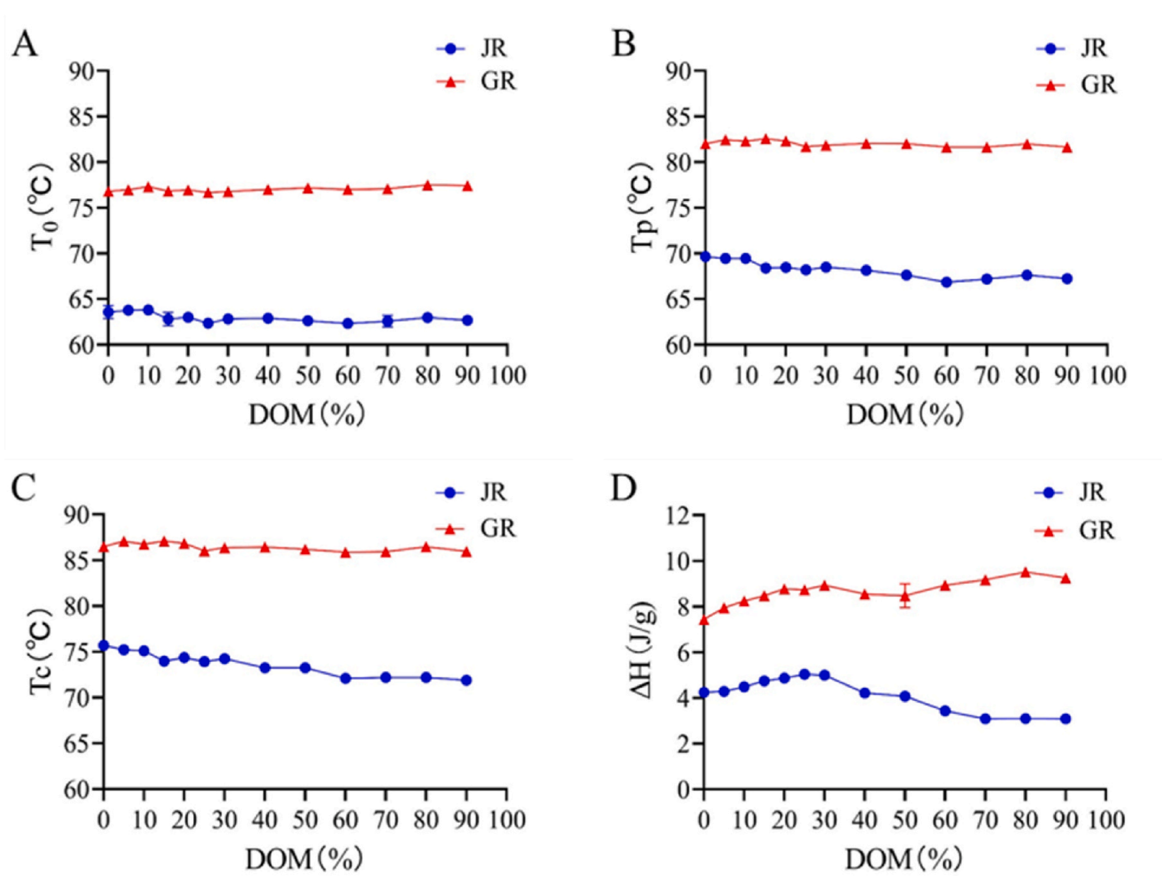

Fig. 2. Thermal properties of rice with different DOM, A: T_0; B: T_p; C: T_c; D: ΔH.

granules (Ohishi et al., 2007). In addition, the gelatinization of starch is accompanied by the formation of an amylose-lipid complex (exothermic).

With increasing DOM, the T_0, T_P, and T_c of JR gradually decreased, and ΔH increased in the first 30% DOM, decreased after 30% DOM, and reached the maximum value at 25% DOM. However, the T_0, T_P, and Tc of GR showed no obvious change, whereas ΔH gradually increased in the first 30% DOM, decreased after 30% DOM, and gradually increased at 50% DOM, showing an overall upwards trend and reaching the maximum value at 80% DOM. The starch with the higher T_0 requires more energy to start the gelatinization process, and ΔH represents the amount of heat absorbed by the starch for the crystal structure to be destroyed or for the double helix to be unwound. The amylose content of JR gradually increased from 0% to 20% DOM, and that of GR gradually increased from 0% to 25% DOM with increasing DOM, while the protein and lipid contents of JR and GR showed a decreasing trend (Fig. 1), which might be related to the increase in ΔH at low DOM (0%–30%). In addition, the effect of protein and lipid on ΔH was less than that of amylose, although lipid had a negative correlation with ΔH. With increasing DOM, the protein and lipid contents gradually decreased, and the starch content increased or decreased due to its uneven distribution in the endosperm, which resulted in different ΔH values of JR and GR at high DOM concentrations.

3.4. Cooking properties with different DOMs

Rice soaking is an important part of raw material processing in *huangjiu* brewing. During this process, rice absorbs water and expands, which is helpful for its further cooking. Rice soaking also involves the growth of lactic acid bacteria and other microorganisms, which decompose and use nutrients (starch and protein, etc.) and produce volatile flavour compounds (Kim et al., 2015). Rice soaking affects not only affects the delivery rate of rice and the volatile components of cooked rice but also the flavour and taste of *huangjiu*. Rice cultivars, growing conditions, milling degree, soaking, and cooking methods also have significant effects on the delivery rate of rice and the volatile components of cooked rice (Shinoda et al., 2020). Investigations were conducted to evaluate the delivery rate of rice and the volatile components of cooked rice with different DOMs, which is the most direct manifestation of the difference in cooking properties during *huangjiu* brewing.

3.4.1. Delivery rate of rice with different DOMs

The delivery rate of rice was determined by the ratio of the weight of cooked rice to the original weight of rice. As an important index in the production of *huangjiu*, the delivery rate of rice reflects the water absorption of rice in the process of rice soaking, which affects the ratio of raw materials in *huangjiu* brewing, especially the amount of water added. In conclusion, the delivery rate of rice of JR ranged from 145.45% to 178.40%, and the DOM value of the maximum delivery rate of rice was 80%. The delivery rate of rice for GR ranged from 146.39% to 188.85%, and the DOM with the maximum delivery rate of rice was 30%, as shown in Fig. 3A. In particular, with the same DOM, the delivery rate of rice of GR was higher than that of JR, while for the same cultivar of rice, the delivery rate of rice showed an upwards trend with increasing milling at low DOM.

In the commercial production of *huangjiu*, the rice delivery rate of rice fell between 1:1.3 and 1:1.5 by using the rice steamer machine with the exudation of water vapour. At the bench scale, the delivery rate of rice increased as the moisture content loss decreased compared with commercial production. A layer of bran fibre on the surface of brown rice hindered the ability of water to enter the inside of brown rice, which affects the rice's ability to absorb water and gelatinize and may explain why the lowest rice delivery rate of rice of both JR and GR appeared at 0% DOM. Rice cooks upon heating due to water absorption and expansion, and the delivery rate of rice is determined by the content and proportion of each component and the water absorption ability of the rice. Starch and protein are the two main nutrients of rice, and their sum accounts for approximately 96% of its dry weight. Both nutrients have a water absorption capacity, and the absorption capacity of protein is much higher than that of starch, which is consistent with a study by Gong et al. (2020). Combined with the total starch content (Fig. 1D), the rice delivery rate of rice was significantly correlated with the starch content at low DOM. Almost all the starch in GR is amylopectin, which is located in the centre of GR. A recent study showed that the delivery rate of rice was highly positively correlated with the content of amylopectin (or the molecular weight of amylopectin) in rice, which affects the ability to combine with and absorb more water (Yang et al., 2020).

3.4.2. Volatile components of the cooked rice with different DOMs

The E-nose was used to effectively distinguish between the volatile components of the cooked rice of JR and GR with different DOMs. The PCA results of the volatile components of the cooked rice with different DOMs are shown in Fig. 3B. PC1 explained most of the variance at 53.6%, while PC2 captured only 24.7% of the overall variance. The total

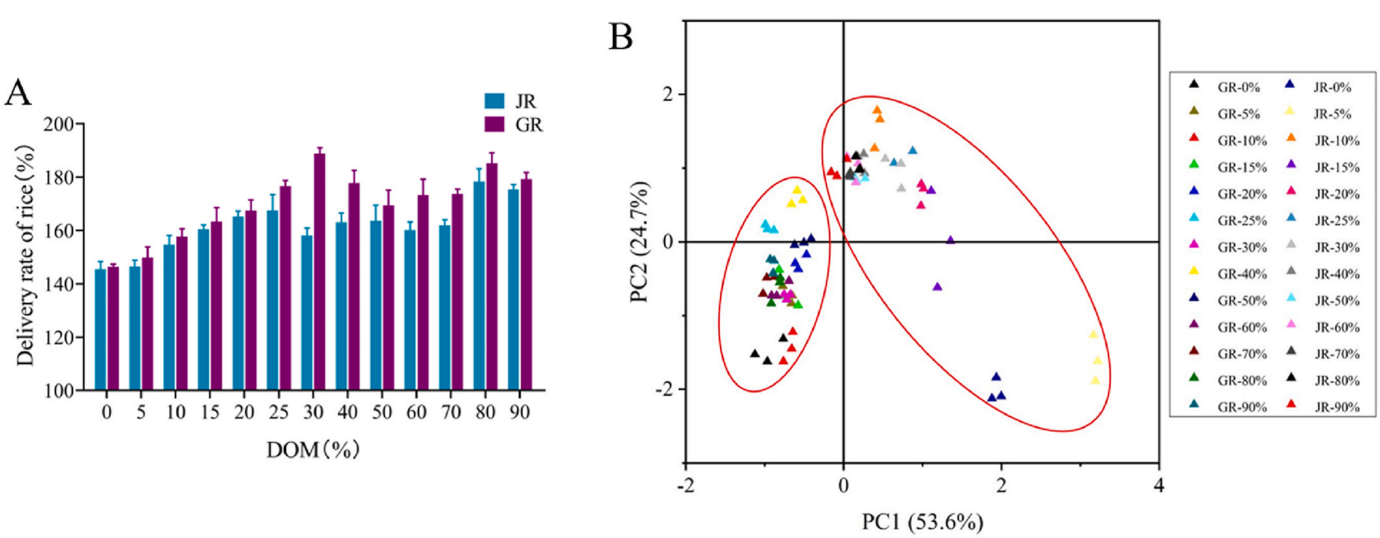

Fig. 3. Delivery rate of rice and volatile components of the cooked rice with different DOM, A: the delivery rate of rice with different DOM; B: PCA analysis of the cooked rice from JR and GR with different DOM.

contribution rate of these two PCs of 78.3% showed that PCA could well reflect the information of the cultivars. All the samples were mainly divided into two parts on the PCA, namely, JR and GR, with different DOMs, showing that different cultivars of rice exhibited significant differences in the volatile components of the cooked rice. The dispersion degree of JR with different DOMs was higher than that of GR in the PCA, showing that the influence of milling on the volatile components of JR was greater than that of GR, which might be attributed to the differences in the structure and nutrient contents of JR and GR (Fig. 1). Notably, the volatile components of the JR at DOMs of 0% and 5% significantly differed from the remainder of the JR, while the distribution of different DOMs of GR was more concentrated and overlapped. Rice soaking for 3–5 days could promote the growth and proliferation of lactic acid bacteria and other microorganisms in large quantities in rice soaking water and the rice surface, producing volatile compounds representing different smells. These compounds will be added to the ingredients (rice soaking water) or retained in the cooked rice and combined with the fermented mash and cooked rice, which further affects the flavour and quality of the *huangjiu* (Gong et al., 2020). Nutrients of JR and GR were used by microbes to different degrees because of their different structures and nutrient contents, especially in brown rice and low DOM. Brown rice and low-DOM rice are rich in many nutrients, such as protein and lipids, microbial growth and metabolism are more vigorous, and numerous volatile components are produced. With increasing DOM, the nutrients available to microorganisms gradually decreased, with fewer naturally occurring volatile components. With the progress of rice soaking, volatile components are produced, which are kept in the rice and participate in fermentation, further contributing to the flavour quality of *huangjiu* (Yang et al., 2020).

3.5. Physicochemical properties of huangjiu brewed from rice with different DOMs

The physicochemical properties of *huangjiu* brewed from rice with different DOMs, including alcohol content, reducing sugars, total acids, and amino nitrogen, are shown in Fig. 4. With increasing DOM, the alcohol content and reducing sugars showed an overall trend of increasing and then decreasing, especially from 0% to 30% DOM. *Huangjiu* brewed with JR had the highest alcohol content and reducing sugar at 15% DOM, which were 18.10% (v/v) and 3.14 g/L, respectively; *huangjiu* brewed with GR had the highest alcohol content at 10% and 15% DOM, which was 16.15% (v/v); and the reducing sugar was the highest at 15% DOM, which was 2.58 g/L. When DOM = 0%, the alcohol content was lowest, which was 15.65%vol and 13.75%vol, respectively (Fig. 4A and B). A possible reason is that a layer of bran wrapped on the surface of brown rice prevented moisture from entering the interior of brown rice, which affected the water absorption and gelatinization of the rice. Nutrient components such as starch could not be fully enzymatically hydrolysed, and the content of alcohol was relatively low. Total acid is an important indicator of whether the fermentation process of *huangjiu* is normal and of the quality of *huangjiu*. An excessively low acidity could not inhibit the growth of miscellaneous bacteria and ensure the normal growth and reproduction of yeast; an excessively high acidity damages the overall flavour of *huangjiu* (Gong et al., 2020). With increasing DOM, the total acid content showed no obvious regular change. Compared with other DOMs, the total acid content was relatively high, 7.03 g/L and 8.29 g/L when DOM = 0% (Fig. 4C). This result might be attributed to the low alcohol content of *huangjiu* brewed with brown rice, which could not sufficiently inhibit the growth and

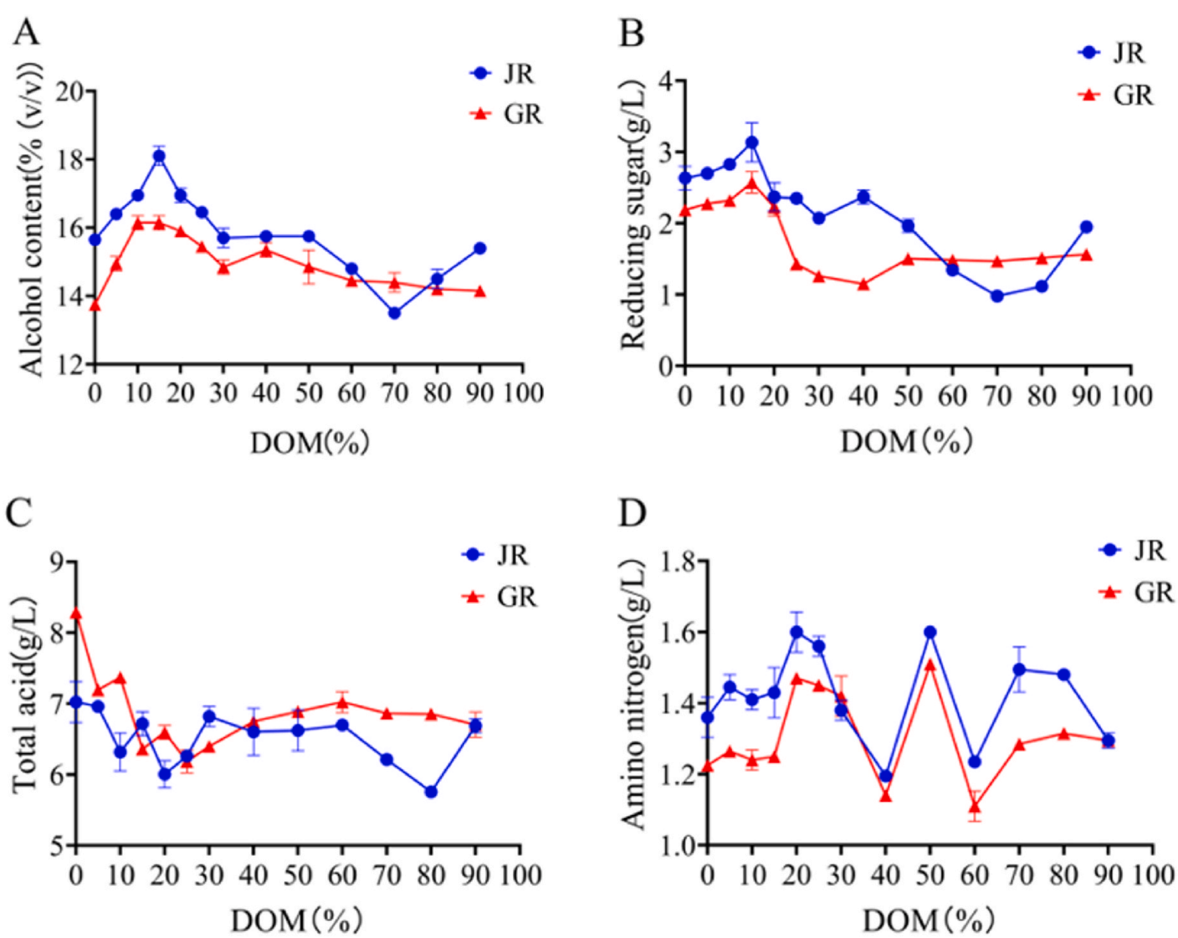

Fig. 4. The physicochemical properties of *huangjiu* brewed from JR and GR with different DOM, A: alcohol content; B: reducing sugar; C:total acid; D: amino nitrogen.

reproduction of bacteria and other miscellaneous bacteria, and the abundance of protein, lipids and other nutrients in brown rice, which is conducive to the growth and metabolism of miscellaneous bacteria to produce acid.

The content of amino nitrogen directly affects the quality of *huangjiu*, and could also be utilized to measure the fermentation degree of raw materials and the degree of protein hydrolysis (Rong et al., 2013). With the same DOM, the content of amino nitrogen in *huangjiu* brewed with JR was generally higher than that in *huangjiu* brewed with GR. When the DOM ranged between 0% and 40%, the content of amino nitrogen in *huangjiu* brewed with JR and GR showed a trend of increasing and then decreasing. When DOM = 50%, the amino nitrogen contents of JR and GR were the highest, 1.60 g/L and 1.47 g/L, respectively. When DOM = 40%, the amino acid nitrogen content in *huangjiu* brewed with JR was the lowest (1.20 g/L), and when DOM = 60%, the amino acid nitrogen content in *huangjiu* brewed with GR was the lowest (1.14 g/L) (Fig. 4D). Rong et al. (2013) discovered that DOM had a significant effect on the content of amino nitrogen. The higher the DOM is, the lower the protein content, and the lower the amino nitrogen content obtained by decomposition, which is not consistent with our study and might be related to the autolysis of yeast.

3.6. PCA of flavour compounds in huangjiu brewed from rice with different DOMs

The volatile flavour compounds, organic acids and free amino acids of JR and GR with different DOMs were analysed by PCA; the results are shown in Fig. 5. PC1 of JR explained most of the variance at 47.4%, while PC2 captured 21.0% of the overall variance, and the total contribution rate of these two PCs was 68.4%. PC1 of GR explained most of the variance at 44.3%, while PC2 captured 33.2% of the overall variance. The total contribution rate of these two PCs was 77.5%, both of which well reflected the sample information. The PCA results showed that when the DOM was in the range of 5%–15%, the flavour compounds of *huangjiu* brewed from JR and GR could be well clustered, indicating that their flavour compounds and contents exhibit a certain similarity.

Based on the physicochemical properties and flavour, when the DOM was in the range of 10%–15%, the alcohol content, reducing sugar and amino acid nitrogen of *huangjiu* brewed from JR and GR were relatively high, and the total acid content also reached the standard of *huangjiu* brewing. In addition, this process avoids the problems of a low grain utilization rate and high energy consumption caused by excessive milling, caters to the current mainstream processing accuracy of rice and is suitable for the industrial production of *huangjiu*. When the DOM was in the range of 60%–90%, the flavour components of *huangjiu* brewed from JR and GR could also be well clustered, which was attributed to the low lipid and protein content in the raw rice (Fig. 1). The flavour of *huangjiu* was purer, but the production cost of rice with a DOM in the range of 60%–90% was much higher than that of rice with a DOM in the range of 5%–15% DOM.

4. Conclusions

In order to explore the effects of milling degree and different varieties on the nutritional components and brewing characteristics of rice for *huangjiu* brewing, on the one hand, different degrees of milling (0%–90%) were carried out for JR and GR, and the nutritional components, gelatinization properties, thermodynamic properties, cooking rate and volatile components of rice with different DOM were analysed. On the other hand, the effects of different DOM and varieties on basic physicochemical indices, volatile flavor compounds, organic acids and amino acids of post-fermented yellow *huangjiu* were studied using the same brewing process. The final result is that when the DOM is in the range of 5%–15%, the flavour compounds of *huangjiu* brewed from JR and GR could be well clustered. The liquor, reducing sugar and amino acid nitrogen contents of *huangjiu* made from JR and GR were relatively high. The flavour compounds and content were relatively similar. The quality of rice wine is relatively stable, and the problems of a low grain utilization rate and high energy consumption caused by excessive milling are avoided, which meets the industrial production of *huangjiu*, and provides theoretical basis and scientific guidance for the processing and selection of raw materials in *huangjiu* production. In the later stage, the conclusions can be applied to the scale-up production of rice wine factories, and the production quality of rice wine can be substantially improved. At the same time, it also needs consumers to conduct sensory appraisal and give the most direct quality evaluation of the upgraded *huangjiu*.

CRediT authorship contribution statement

Shuangping Liu: Formal analysis, Analysis of results, Data curation, data compilation, Writing – original draft. **Hui Zhang:** Experimental supplies sponsorship, results testing. **Jinlong Peng:** Project & writing guidance. **Zhe Yao:** Writing revision. **Xiao Han:** Data collation. **Tiantian Liu:** Writing – review & editing. **Songjing Zhang:** Supervision, Experimental supervision & design. **Jian Mao:** Supervision, Project administration, supervision & administration.

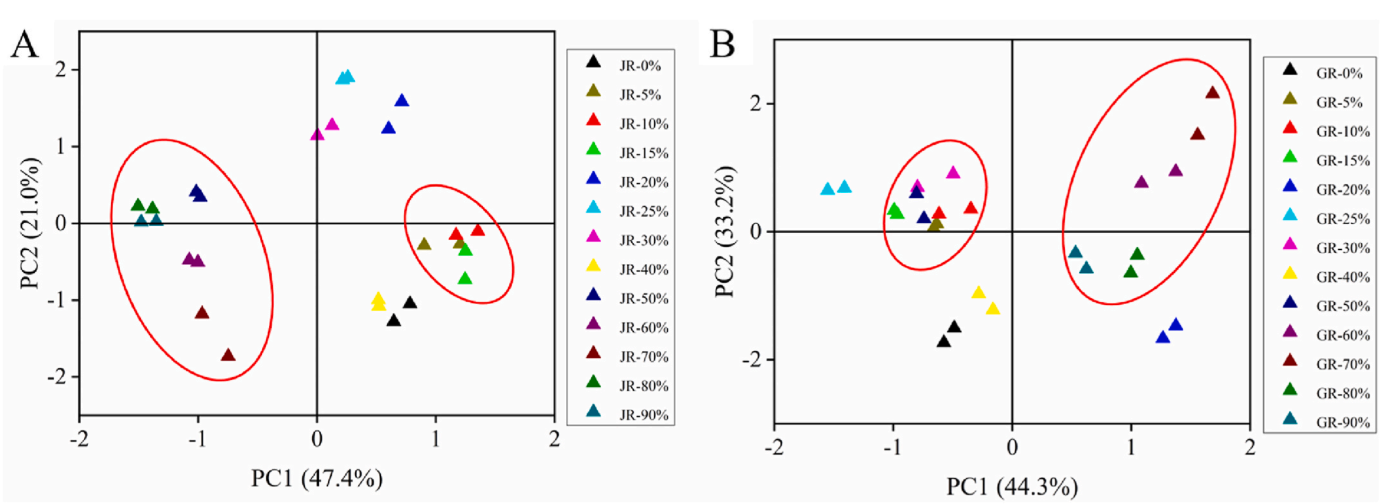

Fig. 5. PCA analysis of volatile components in *huangjiu* brewed from rice with different DOM, A: JR; B: GR.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.lwt.2023.114825.

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