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Influence of adsorbent-arak ratio and distillation period in bioethanol purification process using Balinese liquor as a raw material

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Abstract

Arak is one of traditional Balinese drink that has alcohol content used as a potential raw material in making bioethanol. The aim of this research was to study the influence of adsorbent-arak ratio and distillation period on the characteristics of bioethanol and to identify the right method to produce bioethanol with the best characteristics using distillation-adsorptive purification method. This research used completely randomized design with factorial experiment. The first factor was adsorbent-arak ratio, *i.e.*, 1:2, 1:3 and 1:4. The second factor was the distillation period, *i.e.,* 1, 2, 3 h. Each factor was grouped into two groups based on the two times of arak production so that there were 18 experimental units. The data was analysed using Analysis of Variance (ANOVA) and means were compared by Duncan's multiple range test. The result showed that the ratio of adsorbent-arak and distillation period had a significant effect on ethanol content, density, specific gravity, API gravity, and heating value. The combination of 1:3 adsorbent-arak ratio and one-hour distillation period produced the best characteristic *viz*., ethanol content 91.86 %, density 0.83 kg/L, specific gravity 0.81, API gravity 42.31, and heating value 11081.9 kcal/kg.

Key words: Arak, adsorbent ratio, distillation period, bioethanol

Introduction

There is an increase in energy needs, to keep up with recent development; fossil fuels that exist today cannot be expected to last for a long time. New alternative energy sources that are sufficient and can save energy from fossil fuels are required (Jhonprimen *et al.*, 2012). Bioethanol is a biochemical liquid derived from the fermentation of sugar from carbohydrate sources with the assistance of microorganisms. Bioethanol has been identified as a significant renewable and sustainable fuel source for the future (Yang *et al.*, 2012; Gunam *et al.*, 2019)

Arak is one of the traditional Balinese liquor from traditional distillation of palm juice and coconut. Arak has the potential for high alcohol content (±30 %) which is suitable to be used as a raw material for the production of bioethanol (Sukadana and Tenaya, 2014). To improve the quality of bioethanol from Arak, further purification is necessary to increase the concentration of alcohol content. One of the purification methods which is commonly done is by distillation (Goering and Schrader, 1988).

Distillation is a method for separating two types of solutions based on the boiling point differences. However, according to Onuki *et al.* (2008), this method has several deficiencies. First, it is estimated that there are impurities with similar boiling point to ethanol mixed with ethanol after the distillation process so that the purity is low. Second, distillation which is a process of repetition of evaporation and condensation, requires high costs. To overcome this, a purification process is needed by means of adsorption as a continuation of the distillation process, to achieve a purity of around (90-95 % v/v). This process is known as the distillation-adsorption process (Mujiburohman *et al.,* 2006; Patil and Patil, 2017).

Distillation-adsorption is a combination of two separation processes, namely distillation and adsorption. In this method, the distillation and adsorption processes are carried out simultaneously, that is, by means of the adsorbent column arranged together with a distillation tool. The distillate vapor will pass through the adsorbent column. The adsorbent will absorb water vapor so that the purity of ethanol rises. The difference between distillation-adsorption and other distillation methods is the addition of the additives substance which does not mix together with the solution but in a separate column. Thus, the separation between the additives and the solutions is not required (Silviana and Purbasari, 2008; Tang *et al.*, 2013; Chopade *et al.*, 2015). The adsorbent chosen in the bioethanol distillation-adsorption process is a hydrophilic adsorbent because it has the ability to absorb water. Silica gel is used because it has several advantages including being very inert, hydrophilic and the costs of synthesis are quite low. In addition, this material is relatively stable thermally and does not expand in organic solvents when compared to organic polymer solids (Purwaningsih, 2009).

Studies on the use of adsorption distillation methods in the purification process have been carried out and one of which is on the process of separating isopropyl alcohol-water mixtures (Silviana and Purbasari, 2008; Mujiburohman *et al.*, 2006; Banat *et al.,* 2003). The adsorption distillation method was also investigated by Rizki *et al.* (2012), based on the results of the study it was found that the most effective process for ethanol purification is the adsorption distillation process with the ethanol content produced at 98 % (v/v). Furthermore, Yuliana *et al.* (2015) reported that with purification process based on the adsorbent-ethanol ratio of 1:2, the purity of bioethanol obtained was 99.7 % (v/v). Based on this, a study was conducted to identify the effect of adsorbent-arak ratio and distillation period in order to obtain the best bioethanol characteristics in the bioethanol purification process using distillation-adsorption method.

Materials and methods

Material: The arak was obtained from Duda village in Karangasem and Dawan village in Klungkung with ±30 % alcohol content (v/v), and granular white silica gel adsorbent (non-food grade) obtained from Bratachem.

Research design: This research used a two factor completely randomized design with factorial experiment. The first factor was the weight ratio between adsorbent and arak which consisted of three levels, namely, 1:2, 1:3, and 1:4. The second factor was the distillation time consisting of three levels, namely, 1, 2, and 3 h grouped into two groups based on times of arak production so that 18 experimental units were obtained.

Research implementation: Initially, silica gel adsorbent was physically activated; activation was carried out by heating the adsorbent on the furnace at a temperature of 200±2 °C for 2 hours. Physical activation is carried out to increase the absorption ability of silica gel adsorbents. Then, the activated adsorbent was inserted into the adsorbent column contained in the distillation according to the adsorbent-arak ratio (1:2, 1:3, 1:4). The volume of arak used for each ratio was 1; 1.5; and 2 L, while an adsorbent in arak was used as much as 0.5 kg for each ratio. The alcohol content was measured first using an alcohol meter and entered into the feed column according to the adsorbent: arak ratio (1:2, 1:3, 1:4). Arak was heated until the temperature reached 80 °C and then kept constant. The distillation process was carried out at 80 °C, because ethanol boils at 78.4 °C while water at 100 °C. The steam produced by the arak was passed through the adsorbent column and the water vapor produced from the heating process was adsorbed by the silica adsorbent gel. The ethanol vapor was passed through the cooling column and accumulated in the distillate tank. The period of distillation-adsorption process was carried out according to the experimental variables (1, 2, 3 h). The distillate obtained from the distillation process was then analysed according to the observed parameters. Meanwhile, the used silica gel adsorbent was removed from the distillation tool and then reactivated for further use.

The variables observed: Ethanol content (SNI: Indonesian National Standard, 3565:2009), density, specific gravity, API gravity and heating value (Tjokrowisastro and Widodo, 1990).

Data analysis: The data were analyzed according to the variance (ANOVA) and means compared with Duncan's multiple comparison test using SPSS 25.

Results

Bioethanol content: Based on the results of the ANOVA, it was found that the adsorbent-arak ratio, duration of distillation and interaction between treatments, had a very significant effect on the ethanol content obtained from the purification process by the adsorption distillation method (Table 1). The highest level of bioethanol (92.65 %) was produced with adsorbent-arak in the 1:2 ratio with distillation period for one hour and with longer distillation period, the bioethanol content decreased.

Table 1. Average value of bioethanol (v/v)

Adsorbent-arak ratio	Distillation period (hour)		
	1	2	3
1:2	92.65±0.54[a]	87.71±0.24[d]	85.05±1.38[e]
1:3	91.86±1.05[a]	89.87±0.02[b]	88.24±0.68[c]
1:4	90.79±0.90[b]	88.89±0.70[c]	87.64±0.14[d]

Same letter after the average value shows non significant difference at *P*=0.05.

Density: The highest bioethanol density (0.8463 g/mL) was obtained through purification by the distillation-adsorption method is a combination treatment ratio of 1:2 for three hours (Table 2). The lowest bioethanol density (0.8258 g/mL) was produced by 1:2 ratio treatment for one hour and this treatment combination was significantly not different from the bioethanol density (0.8280 g/mL) produced by the combination of 1:3 ratio treatment for one hour. Based on the average value of bioethanol density, it was found that the longer the distillation process, the higher bioethanol density obtained.

Table 2**.** Average density value of bioethanol (g/mL)

Adsorbent-arak ratio	Distillation period (hour)		
	1	2	3
1:2	0.8258±0.001[e]	0.8393±0.001[b]	0.8463±0.004[a]
1:3	0.8280±0.003[e]	0.8335±0.000[c]	0.8379±0.002[b]
1:4	0.8311±0.003[d]	0.8362±0.002[c]	0.8395±0.000[b]

Same letter after the average value shows non significant difference at *P*=0.05.

Specific gravity: Based on Table 3, the highest specific bioethanol gravity (0.8321)was obtained in the combination of 1:2 ratio treatment for three hours. With the increasing distillation period, an increase in the value of specific gravity of bioethanol was observed.

Table 3. Bioethanol *s*pecific gravity average value

Adsorbent-arak ratio	Distillation period (hour)		
	1	2	3
1:2	0.8120±0.001[e]	0.8253±0.001[b]	0.8321±0.003[a]
1:3	0.8141±0.003[e]	0.8195±0.000[c]	0.8239±0.002[b]
1:4	0.8172±0.003[d]	0.8222±0.002[c]	0.8254±0.000[b]

Same letter after the average value shows non significant difference at *P*=0.05.

API gravity: Based on Table 4, the highest bioethanol API gravity (42.761) was obtained in the combination of 1:2 ratio treatment for one hour distillation period of and the combination of these treatments were significantly not different from the API gravity of bioethanol produced by a treatment combination of 1: 3 ratio, distillation period for one hour, ratio 1:3, distillation period of two hours and ratio of 1:4, distillation period for one hour. The lowest API gravity of bio-ethanol (38.557) was produced by a combination treatment ratio of 1:2, for three hours. The longer the distillation period, the API value was reduced.

Table 4. Average value of API gravity

Adsorbent- arak ratio	Distillation period (hour)		
	1	2	3
1:2	42.761±0.31[a]	39.925±0.13[d]	38.557±0.71[e]
1:3	42.315±0.59[a]	41.163±0.01[a]	40.244±0.38[c]
1:4	41.658±0.58[a]	40.610±0.40[b]	39.557±0.08[d]

Same letter after the average value shows non significant difference at *P*=0.05.

Heating value: The heating value of the produced bioethanol showed a decrease with the longer time of distillation-adsorption and decreasing the adsorbent-arak ratio (Table 5). The heating value was closely related to the bioethanol content; it is known that the longer the distillation-adsorption process, the bioethanol content will decrease due to increased water content in the distillate.

Table 5. Average value of heating value (kcal/kg)

Adsorbent-arak ratio	Distillation period (hour)		
	1	2	3
1:2	11091.8±6.90[a]	11056.3±0.26[b]	11035.9±8.38[c]
1:3	11081.9±13.20[a]	11044.0±8.80[c]	11028.7±1.67[d]
1:4	11067.3±12.97[b]	11029.6±2.83[d]	10998.3±15.79[e]

Same letter after the average value shows non significant difference at *P*=0.05.

Discussion

Bioethanol content: Absorbant-arak ratio, and distillation period both had a significant impact on distillate recovery. The distillate was saturated with the water vapour formed during the distillation process as the silica gel adsorbed, resulting in a lower bioethanol content. According to Satria and Bernardi (2009), the higher the purity of ethanol was obtained when the adsorption process was carried out until the adsorbent reaches a saturated condition. Chopade *et al.* (2015), who discovered that adsorbents absorb water rapidly in the first 60 minutes of adsorption. Wirawan *et al.* (2014) also reported a decrease in bioethanol concentration during the distillation process, indicating that the adsorbent was saturated. The longer the distillation process is carried out, the more saturated the adsorbent becomes, resulting in a lower concentration of bioethanol produced. Meanwhile, for the treatment of adsorbent-arak ratio, the greater the adsorbent-arak weight ratio, the higher the bioethanol content obtained due to an increase in the amount of adsorbent used, which provides a larger surface for effective water absorption by silica gel (Qu *et al.*, 2012).

Density: The adsorbant-arak ratio had an effect on the density of the bioethanol. It happened because the water vapour formed during the bioethanol distillation process was not completely absorbed by the silica gel adsorbent due to saturation, so the formed water vapour entered the distillate and increased the distillate density. This is consistent with Bahtiar's (2013) study, which claims that the increase in bioethanol density is due to the water content that remains in distilled bioethanol. The higher the density, the lower the gravity, and the higher the quality. According to the findings of Khodijah and Abtokhi (2015), the density level has a significant impact on the value of ethanol. The higher the ethanol content value, the longer the carbon chain, which easily breaks with rising temperature.

Specific gravity: Jannah (2016) obtained similar results and explained the trend as the velocity of the mass transfer of water to the silica adsorbent continues to decrease as the time of adsorption increases, so that the concentration of water from the old column increases as long as the specific gravity value obtained by the distillation-adsorption method increases. The lowest specific gravity of bioethanol was produced by a 1:2 treatment ratio for one hour with a specific gravity of 0.812, and this combination of treatments was not significantly different from the specific gravity bioethanol produced by a 1:3 treatment ratio for one hour with a specific gravity of 0.8141. The higher the adsorbent-arak ratio, the greater the adsorbent surface area and the greater the number of water molecules absorbed in the adsorbent (Dyartanti *et al.*, 2012), resulting in a higher distillate alcohol content. The weight or density of the distillate decreases as the concentration of alcohol increases, resulting in a low value for the mixture's specific gravity. This is due to specific gravity, which is the ratio of the material's density to the density of water. As a result, the density of the substance decreases, and the specific gravity falls as well (Sutanto *et al.*, 2013).

API gravity: Because silica gel's adsorption against water decreases with time, different API gravity was observed in different treatments. Furthermore, the higher the adsorbent-arak ratio in the treatment, the higher the API gravity received. Because more adsorbent is used, the interaction between the adsorbent and the mixture is more uniformly distributed, resulting in more water impurities, increasing bioethanol purity and API gravity value (Hidayat *et al.*, 2015; Al-Saidy and Al-Dokheily, 2014). The reciprocal ratio of liquid fuel to water is known as API gravity, and it is closely related to specific gravity (Bint-E-Naser and Hossain, 2017). The goal of examining API gravity, according to Kholidah (2014), is to indicate fuel quality. If the specific gravity is low, the API gravity is high, indicating that the fuel quality is improving because it contains more fuel than impurities such as water and paraffin.

Heating value: Sutanto *et al.* (2013) reported that higher alcohol content bioethanol appears to emit more heat than lower alcohol content bioethanol. Bioethanol, which is made from organic waste, has a heating value of 10,000-11,000 kcal/kg, whereas liquid fuels have a heating value of 10,160-11,000 kcal/kg. A higher heating value would result in greater flammability, which would result in greater efficiency (Wijaya *et al.*, 2012).

Based on the findings of the research, it can be concluded the adsorbent weight ratio, distillation period, and interaction between treatments all have an impact on the properties of bioethanol obtained from the distillation-adsorption purification process. The combination of adsorbent treatment weight ratio of 1:3 and one hour distillation period yielded the best bioethanol characteristics in the purification of bioethanol from arak by the distillation-adsorption method, with criteria of bioethanol content of 91.86 percent; density 0.8280 kg/L; specific gravity 0.8141; API Gravity 42.315; and heating value 11081.9 kcal/kg.

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