

# The Effect of Alcohol on Bead Performance of Encapsulated Iron Using Deacetylated Glucomannan

Dyah H. Wardhani<sup>1\*</sup> and Heri Cahyono<sup>2</sup>

<sup>1</sup>Chemical Engineering Department, Faculty of Engineering, Diponegoro University, Indonesia

<sup>2</sup>MSc Study Program on Chemical Engineering Department, Faculty of Engineering, Diponegoro University, Indonesia

**Abstract.** The success of encapsulation to protect iron from inhibitor degradation or oxidation depends on many factors including the excipient characteristics. Glucomannan, a neutral heterosaccharide, has a potential for the excipient. To improve the excipient performances, glucomannan is deacetylated to remove the acetyl groups by reacted with Na<sub>2</sub>CO<sub>3</sub>. This deacetylated glucomannan is subject to bead formation after iron loading. The alcohol solution is commonly used in bead forming as dehydration medium during the encapsulation process. The objective of this work was to study the effect of alcohol on the bead performance of encapsulated iron using deacetylated glucomannan. The bead forming was conducted by dropping the excipient into ethanol and isopropyl alcohol (IPA) solution at various concentrations (50, 60, 70, 80 and 90%) and two condition temperatures (27-30° and 7-10°C). The encapsulation samples were subject to yield (YE) and efficiency of encapsulation (EE). The concentration of alcohol showed a positive impact on the yield and efficiency of encapsulation. Ethanol has a better performance compared with that of IPA regarding yield and efficiency of encapsulation. The optimum of yield bead formation (69.67%) and highest EE (66.80%) were obtained at 90% ethanol. Temperature of dehydration did not affect the YE and EE significantly.

## 1 Introduction

Encapsulation techniques have been developed broadly to accommodate various purposes. Among of the purposes of encapsulation are to protect the sensitive substances from the external environment, to mask the organoleptic properties like color, taste, odor of the substance, to obtain controlled release of the drug substance, for safe handling of the toxic materials, and to get targeted release of the drug [1].

Iron is one of the essential micronutrients that urgently develop into encapsulated form since it is easily oxidized when exposed to air. Its present induces unpleasant organoleptic properties especially odour and metallic taste [2],[3]. Moreover, iron encapsulation leads to increase bioavailability due to excipient protection of the iron from interaction with food components which are inhibitor degradation such as phytates, tannins, and polyphenols [4],[5]. The succeed of encapsulation in protecting the iron from inhibitor degradation or oxidation depends on many factors e.g. the excipient properties and preparation condition [1],[6].

Many compounds have been studied as excipient of iron encapsulation such as modified alginate [3],[7], hydrogenated soybean oil [8], dextrin, cellulose, and sodium hexametaphosphate (SHMP) [9],stearic acid, partially dehydrogenated soybean oil, wax matrix [10].

In recent years, application glucomannan (GM) as an excipient has been explored due to its extraordinary capability in absorbing water and excellent and stable films in both cold and hot water [11],[12]. GM is known as one of the highest viscosity among various gum types [13],[14]. However, the water-absorption of GM which is able to absorb up to 100 g water/g becomes a problem in the excipient application. Hence, this property requires to be controlled. Some methods have been reported to modify the water absorption of GM including thermal treatment [12], high-pressure treatment [15] and chemical modification [16]. Since the presence of acetyl groups in the GM chain inhibits interchain interactions [17], hence deacetylation is considered as one of the most appropriate chemical modification in reducing GM water-solubility. This modification is also expected to minimize iron oxidation due to contact with dissolved O<sub>2</sub> in water [6].

There are various techniques of bead forming in encapsulation process including spray drying, spray chilling or spray cooling, and extrusion coating [18]. An extrusion or dripping method is one of the simple method to obtain the bead [1],[19]. This process requires a dehydrating medium to obtain the desired bead of the encapsulated compound. The bead then is separated from the dehydrating medium [6]. Alcohol is commonly used as dehydrating medium due to its solubility in water

\* Corresponding author: [dhwardhani@che.undip.ac.id](mailto:dhwardhani@che.undip.ac.id)

[20]. With respect to solvation, the similar in chemical structure to each other depends primarily on their polarity [21]. Moreover, alcohol has high volatility which could help in dehydrating process. Zilberboim et al. reported that temperature of dehydrating medium affects the success in bead forming of Paprika oleoresin encapsulation with gum arabic [22]. Gupta et al. suggested to apply lower temperature in iron encapsulation when using alcohol as dehydrating medium [6].

Ethanol have been reported as a dehydration medium in iron encapsulation [19],[22] but no report isopropyl alcohol (IPA) as dehydrating medium, even though IPA has potential for dehydrating medium. However, to the best of our knowledge, there is lack of information in comparing both dehydrating medium in iron encapsulation using deacetylated GM (DGM). Therefore, the objective of this study was to determine the effect of type of alcohol and temperature of dehydrating medium in iron encapsulation with DGM. The samples were subject to yield and efficiency of encapsulation.

## 2. Materials and method

### 2.1. Materials

A. oncophyllus flour was bought from a local farmer at Nganjuk, East Java-Indonesia. GM of A. oncophyllus was extracted following Rahayu method's using IPA [23]. The purified flour contained 76.3% GM.

### 2.2. Deacetylation of Glucomannan

The deacetylation was conducted based on the method of Chen et al [24]. Ethanol (50%, 100 ml) and GM (5 g) were placed in Erlenmeyer which magnetically stirrer at 250 rpm and 50°C. After 30 min, Na<sub>2</sub>CO<sub>3</sub> (0.4M, 25ml) was added to the suspension. The mixture was stirred using a magnetic stirrer (225 rpm) for 24 h. The solid which is separated using filter paper was oven-dried at 40°C for 6 h to obtain DGM.

### 2.3. Bead Forming

The bead forming was conducted following Wardhani et al. [25]. The dried DGM (2 g) was diluted with 100 ml of distilled water in Erlenmeyer and stirred at 250 rpm to make a suspension. One gram of FeSO<sub>4</sub>.7H<sub>2</sub>O was diluted into the suspension. After 30-50 min, the solid was precipitated and the liquid was dropped carefully into ethanol (50%, 100 ml) using 10ml-syringe. The bead which formed after 12h was collected and oven-dried at 40°C for 4 hours to obtain dried-iron encapsulate. The yield (YE) of encapsulation was calculated following Eq.1.

$$YE = \frac{A}{B} \times 100\% \quad [1]$$

where A and B are the weight of final dried-iron bead and dried DGM, respectively.

### 2.3. Determination of Iron

Iron concentration was determined based on Tamura et al [26] procedure. Dried bead (1 g) was diluted in 100 ml distilled water. Place 0.1 ml sample into a 10 ml measuring flask. The solutions which are 1 ml of the hydroxylamine solution (100g/l), 10 ml of the 1,10-phenanthroline (1g/l) solution, and 8 ml of the sodium acetate solution (1.2 M) were added to the flask as the order. Distilled water was added upto the limit. Read the absorbance of the mixture at 520 nm against the absorbance of the iron standard solution (10-500 ppm). The efficiency of encapsulation was defined as Eq. 2:

$$\text{Efficiency of encapsulation} = \frac{\text{Bound iron}}{\text{Total iron}} \times 100\% \quad [2]$$

## 3. Result and Discussion

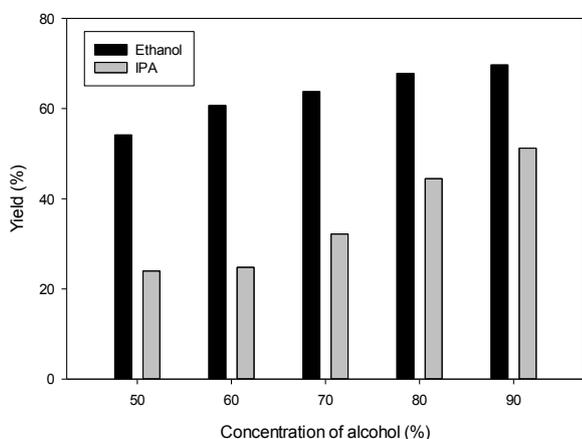
Since iron was sensitive to be oxidized, solvent evaporation was selected as a method to form beads during iron encapsulation using deacetylated glucomannan. Type of alcohol, its concentration and temperature was studied over the yield and encapsulation efficiency.

### 3.1. Yield of Encapsulation

Alcohol was used in this study since it did not only serve as a precipitate agent for the glucomannan but it also promote dehydration of the forming bead [27]. Figure 1 describes the effect of ethanol and IPA as dehydration medium and its concentration on the yield of encapsulation (YE). In this study, the mixture of DGM-iron sol was syringe-dropped to the alcohol as dehydration medium. As soon as the sol contact with the alcohol, the sol dehydrated and resulted in the beads formation. This bead formation involved dehydration process where water content of the sol reduced. The water of the sol system was relatively in higher concentration toward the water in the alcohol which acted as the desiccant. As a result, dehydration occurred due to water removal of the sol to the alcohol. This principle of phenomenon was similar to the osmotic dehydration in which the water diffused from the the higher osmotic pressure solution to the hypertonic ones through a membrane [28]. In this case the bead of DGM-Iron acted as the membrane while alcohol was the hypertonic solution.

In general, YE of ethanol was higher than that of IPA in the all concentrations (Figure 1). The highest YE (69.67%) obtained using 90% ethanol and the lowest YE (23.97%) obtained from 50% IPA. This phenomenon could be due to different DGM response to the properties of both desiccant solutions including polarity, solubility and volatility.

One of the driving force of this dehydration was solubility of the desiccants. A solute dissolves in a solvent that has most similarity in chemical structure to itself. With respect to solvation, this similar in chemical structure to each other depends primarily on their polarity [21]. This polarity could be expressed with dielectric constant [29]. Dielectric constant of ethanol (25.3) was higher than that of IPA (20.18) [30]. This higher solubility of ethanol led to higher driving force for water transfer between the sol and the desiccant. This condition allowed to gain more beads in using ethanol as desiccant. Yeo and Park suggested higher solubility desiccant allowed relatively faster mass-transfer between the excipient and the desiccant. This led to faster precipitation of the polymer [31].



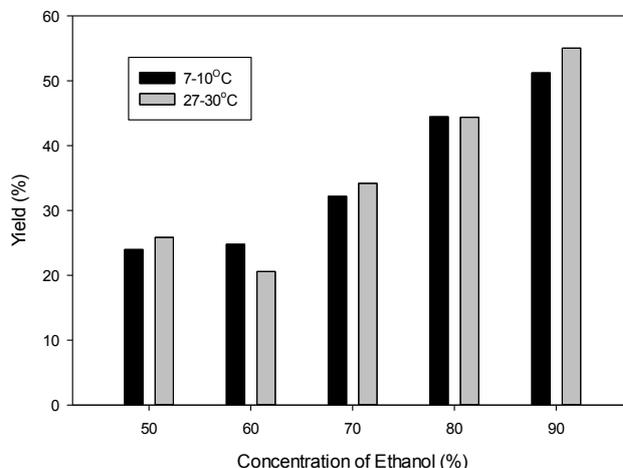
**Fig. 1.** Effect of type and concentration of alcohol on the yield of encapsulation

IPA has more number of carbon than ethanol, hence it is less volatile than ethanol [32]. As consequent, it could require more time to obtain similar amount of beads than that of ethanol.

Increasing alcohol concentration influenced positively on the yield (Figure 1). The water content of high alcohol concentration was low. This led to create high driving force between the system and more beads were formed in shorter time [19]. Similar result was reported by Zilberboim et al. who found higher alcohol concentration as a dehydrating medium increase yield of encapsulation of oleoresin and aromatic esters of paprika. Lower concentration of alcohol caused particle agglomeration which difficult to be filtered [22].

The effect of temperature on YE was conducted in ethanol solution only. Two temperature were studied *i.e.* 7-10°C and 25-27°C. The results is represented at Figure 2. In general lower temperature showed slightly higher yield than higher temperature. The highest YE (55.04%) was obtained at 90% ethanol of low temperatures. Meanwhile, the lowest YE (20.58%) was achieved at

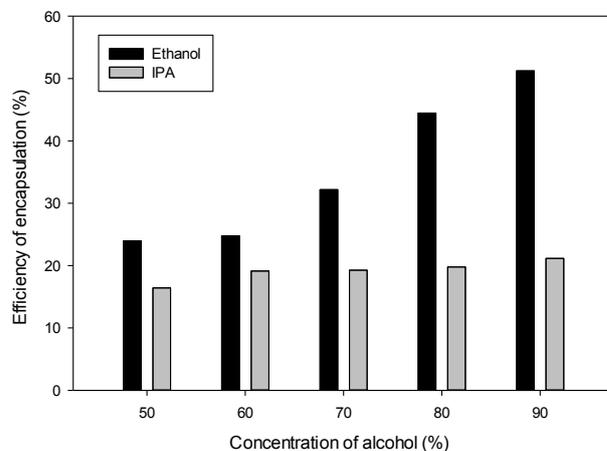
low temperatures using 60% ethanol. Temperature is one of the factors that affect the solubility of the substance. Higher temperature facilitated disruption of the beads and dissolve them [6].



**Fig. 2.** Effect of temperature of dehydrating medium and concentration ethanol on the yield of encapsulation

### 3.2. Efficiency of Encapsulation (EE)

Encapsulation efficiency (EE) is one of the success parameters on the encapsulation process. EE determined by comparing the iron content of the beads and the iron added in the encapsulation process. The iron content was measured by spectrophotometric method. The effect of type and concentration of alcohol on the EE is described at Figure 3. EE has a positive correlation with alcohol concentration in both types of alcohol. Ethanol as a hydrating medium showed a higher EE than IPA. The highest EE (51.23%) was obtained at 90% ethanol, while the lowest level of EE (16.41%) was obtained using 50%.

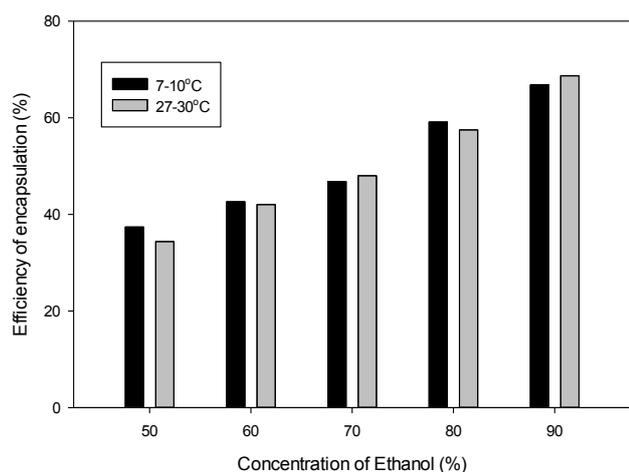


**Fig. 3.** Effect of type and concentration of alcohol on the efficiency of encapsulation (EE)

In this study, ferrous sulphate heptahydrate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) was used as a source of iron. This compound is water soluble (29.51g/100ml, at 25°C) but insoluble in alcohol [33]. Hence, the use of alcohol as dehydration medium in bead forming was an appropriate step.

Ethanol was better in retaining iron in the bead than IPA during the dehydration process. Favourable solubility of iron in the dehydration medium would enhance EE. Low water solubility of desiccant required more time to obtain the beads. However, extended the time allowed to the iron to diffuse to the desiccant [31]. Since IPA has a lower water-solubility than ethanol, hence iron tended to exist on dehydrating medium than bound to DGM excipient.

Alcohol concentration also has an influence on EE on both types of alcohol. Concentration of alcohol showed a positive relation with the EE. High concentration of alcohol led to rapid bead formation due to low water content in the medium. This reduced the possibility of iron loss during beading [19]. This result supported by Gupta et al. which confirmed that the increase in alcohol concentration in bead forming of encapsulated iron could improve the efficiency of iron encapsulation [6].



**Fig. 4.** Effect of temperature of encapsulation and ethanol encapsulation on the efficiency of encapsulation (EE)

Fig. 4 shows increasing ethanol concentration led to increase EE. The highest EE (68.70%) obtained at low temperatures of 90% ethanol. Meanwhile, the lowest EE (34.39%) was found at low temperatures with an ethanol concentration of 50%. The impact of temperature on EE is not significant (Fig.4) which was in line with the yield result. Yeo and Park suggested EE was not affected by solvent evaporation temperature [31].

#### 4. Conclusion

This study confirmed that the ethanol was better as dehydrating medium than IPA in iron encapsulation using deacetylated glucomannan. Higher alcohol

concentration was selected in gaining more bead formation. The optimum bead formation (69.67%) and highest EE (51.24%) were obtained at 90% ethanol. Temperature of dehydration did not affect the YE and EE significantly.

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