Influence of Crosslinking Agent Concentration on The Microencapsulation Parameters of Collagen Hydrolysate-Walled Microcapsules

Ahmet Aslan
Department of Leather Engineering
Ege University, Faculty of Engineering
Izmir, Turkey
ahmet.aslan@gmail.com

Bugra Ocak
Department of Leather Engineering
Ege University, Faculty of Engineering
Izmir, Turkey

***Abstract*—Despite being a lot of discussion, a large amount of chrome tanned leather is still being tanned in the world. After tanning with chrome, chrome solid leather wastes occurred. These chrome wastes arise during mechanical processes such as trimming, shaving, buffing and splitting. These solid wastes containing trivalent chromium and may be oxidized to hexavalent chromium by the appropriate conditions in the landfill areas. Increasing environmental awareness and regulations have been making great pressure on the leather industry. Nowadays, solid leather wastes tanned with chromium are assessed by hydrolysis with the enzyme in high alkaline media and can subsequently be used for several applications. However, there is still no definitive solution to these chromed solid leather wastes. In this study, microcapsules of collagen hydrolysate cross-linked with glutaraldehyde containing Lavender essential oil were prepared by coacervation method. The influence of crosslinking agent concentration on the oil load (%), oil content (%) and encapsulation efficiency (%) of lavender oil microcapsules were investigated. As the crosslinking agent concentration increased, the oil load (%) decreased but oil content (%) and encapsulation efficiency (%) of microcapsules increased. The best result obtained by using 5 g of collagen hydrolysate, 7 ml of glutaraldehyde and 7 ml of lavender oil.**

***Keywords*—Chrome, Collagen hydrolysate, lavender oil, leather, microencapsulation**

I. INTRODUCTION

Chrome-tanned leather accounts for over 90% of world leather production [1]. Treatment of chromium-tanned leathers by mechanical processes such as shaving, buffing, splitting and trimming results in chromium-tanned leather solid wastes (so-called shavings) [2]. There are many studies concerning modified or alternative leather processing techniques or processes to reduce the solid wastes from the leather industry [3,4]. However, these studies have not provided a viable solution because they are not economical and cannot achieve the desired properties for leather in the same way as chromium, and for this reason chromium tanning still keeps its place in the leather industry [5].

Studies on lavender essential oil have been reported due to its applicability in odor industry. In addition, it can be used as a pesticidal and an antimicrobial agent [6]. Essential oil, like lavender oil, are instable compounds. They can suffer volatilization and environmental damages. Microencapsulation of essential oils is a feasible alternative to increase the stability of these compounds for improving their efficiency. The studies regard the microencapsulation of volatile essential oils used for the purpose of durable aroma finishing in the textile industry especially [7].

In this study, we investigated the possibility of using collagen hydrolysate obtained from shavings, as a polymeric wall material to microencapsulate lavender oil using the technique of coacervation and the effects of variations in concentrations of crosslinking agent on the microencapsulation parameters of collagen hydrolysate-walled microcapsules. In this regard, an alternative field of use of chrome-tanned solid leather wastes would be provided to enable evaluation of these wastes instead of dispose in security landfills.

II. MATERIALS

Magnesium oxide (Merck, Germany), Alcalase (Sigma-Aldrich, USA), glutaraldehyde 25% w/v (Merck, Germany), anhydrous sodium sulphate (Merck, Germany), silicone antifoam 30% w/v (Sigma-Aldrich, USA), Tween 80 (Sigma-Aldrich, USA), lavender oil (Sigma-Aldrich, USA), acetic acid (Merck, Germany) were used as received, without further purification. Shavings were obtained from a local tannery in Izmir, Turkey. Besides this, other reagents used (Sigma-Aldrich, USA), were of analytical grade. Ultra-pure water was used throughout the study.

III. METHODS

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A. Preparation of Collagen Hydrolysate

Chromium-tanned leather wastes were soaked in water five times their weight and treated with 4% MgO at 65°C for 30 min. In the following step, the enzyme Alcalase was added to this solution at a concentration of 0.1% (w/v) and the mixture was digested for four hours. This mixture was then filtered hot through Whatman # 1 filter paper to separate the chrome cake, and the liquid collagen hydrolysate was dried to powder form in a spray drier (Niro Atomizer brand) [8].

B. Phase Separation Behaviour of Collagen Hydrolysate

In order to determine the phase separation temperature reached with the addition of the anhydrous sodium sulphate, several experiments were performed with collagen hydrolysate. To that end, an Erlenmeyer flask containing a known quantity of collagen hydrolysate was immersed in a temperature-adjusted water bath kept at 5°C. Anhydrous sodium sulphate (10% w/v) was added under continuous mixing gradually. The temperature of the water bath was then increased by 1 ºC per minute, and the temperature at which the phase separation began and the proportion of sodium sulphate solution to collagen hydrolysate were recorded. The minimum temperature and ratio of collagen hydrolysate to sodium sulphate at which phase separation occurred were 40 °C and 1:6.

C. Preparation of Microcapsules

After inserting 50 ml CH solution (6-10% w/v) into the reaction vessel placed in a temperature-controlled water bath, the internal temperature of the reaction vessel was adjusted to 30°C. The CH solution in the reaction vessel was stirred at 1200 rpm with the help of a mechanical stirrer. One drop of silicon-based antifoaming agent was added, and the internal temperature of the reaction vessel was raised to 40°C. Then, the lavender oil used as the active substance was added into the reaction vessel. Coacervation was achieved by gradual addition of aqueous sodium sulphate solution (10% w/v) for about 90 minutes.

The internal temperature of the reaction vessel was kept at 40°C for 30 minutes more, after which it was decreased to 12°C. The collagen hydrolysate-walled microcapsules were crosslinked through the slow addition of glutaraldehyde solution containing 25% glutaraldehyde at the stirring speed of 800 rpm with the mechanical stirrer and the internal temperature of the reaction vessel was raised to 40°C again, and after continuous stirring for 6-8 hours, was decreased to room temperature. Then, the microcapsules were filtered and rinsed in 0.1% Tween 80 solution.

The rinsed microcapsules were transferred to stainless steel trays and frozen at -30ºC for 24 hours, after which they were dried for 24 hours in a freeze dryer (Christ ALPHA 1-2 LD) at -30°C with 100 mTorr vacuum, and stored at 4°C in an amber-coloured glass bottle [9].

D. Calibration of Curve of Lavender Oil

A known concentration of essential oil in distilled water containing 0.3% Tween 80 was scanned in the range of 200–400 nm by using a UV-VIS spectrophotometer. The sharp peaks caused by the sequential concentrations of 0.05–5 g/100 ml lavender oil in 0.3% Tween 80 at 341 nm (λ_{max}) were recorded. Unknown concentration of lavender oil were determined by extrapolating the absorbance values from the calibration curve prepared by plotting the known LO concentration versus its corresponding absorbance values.

E. Determination of the Oil Load, Oil Content and Encapsulation Efficiency

After crushing the microcapsules completely in a porcelain mortar, known concentration of crushed microcapsules was placed in a volumetric flask. 100 ml of 0.3% Tween 80 (in distilled water) was poured into the volumetric flask, and the flask was shaken all night in a shaker. The encapsulation efficiency (%), oil content (%), and the oil load (%) were determined using the calibration curve and the following Equation 1, 2 and 3.

$$\text{Oil load (\%)} = w_2/w_3 \times 100 \quad (1)$$

$$\text{Oil content (\%)} = w_1/w \times 100 \quad (2)$$

$$\text{Encapsulation efficiency (\%)} = w_1/w_2 \times 100 \quad (3)$$

where w=weight of microcapsules; w_1=actual amount of oil encapsulated in a known amount of microcapsules; w_2=amount of oil introduced in the same amount of microcapsules; w_3=total amount of polymer used including crosslinker.

IV. RESULTS

A. Effects of Variations of Crosslinking Agent Concentaration

It was found that the oil load (%) and oil content (%) values of the microcapsules decreased as the collagen hydrolysate concentration rose (Table 1).

It was observed that, as the crosslinking agent concentration increased, the oil content (%) of the microcapsules increase from 41.54 to 48.37 and encapsulation efficiency (%) of the microcapsules increased from 62.10 to 73.46. However, the oil load (%) of the microcapsules decreased from 200.41 to 174.86 (Table 1). Cross-linking agent reinforce the wall of microcapsules by developing the strong covalent crosslinks [10]. And the cross-linking degree can affect the microencapsulation parameters of microcapsules [11].

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TABLE I EFFECTS OF VARIATION OF GLUTARALDEHYDE CONCENTRATION ON THE OIL LOAD (%), OIL CONTENT (%) AND ENCAPSULATION EFFICIENCY (%) OF MICROCAPSULES

Sample Particulars			Oil load (%)	Oil content (%)	Encapsulation Efficiency (%)
CH (g)	*GA (ml)*	*LO (ml)*	*Mean*	*Mean±SD*	*Mean±SD*
5	3	7	200.41	41.54 ± 0.90	62.10 ± 1.37
5	5	7	177.98	44.29 ± 1.02	68.95 ± 1.58
5	7	7	174.86	48.37 ± 1.06	73.46 ± 1.65

V. CONCLUSION

It was possible to encapsulate lavender essential oil successfully in a collagen hydrolysate-wall with the help of glutaraldehyde. As the amount of glutaraldehyde used as a crosslinker increased, the oil load (%) decreased while the oil content (%) and encapsulation efficiency (%) increased.

Consequently, the use of collagen hydrolysate obtained from solid wastes of chromium-tanned leather in the production of microcapsules containing lavender essential oil has transformed a substance into a substrate meeting a different ecological use.

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