

Synthesis of Mint-flavor Microcapsules and Its Application in Cigarettes

Shujin Cheng^{1,*}, Yiting Han¹, Shuming Zhang², Dongyan Zhao¹

¹GENERAL TOBACCO GROUP Co., Ltd, Jinan, Shandong, 250100, China

²QINGDAO ETSONG TECHNOLOGY Co., Ltd, Qingdao, Shandong, 266011, China

*592315880@qq.com

Abstract. By using microcapsule technology, the components that may react with each other can be made into microcapsule products respectively, so that they can be stabilized in a system, and each effective component can be released in an orderly manner and act at the corresponding time, thus improving the flavor and nutrition of the products. In order to further find an efficient and reasonable cigarette flavoring process, this study used gelatin and Arabic gum as composite wall materials to synthesize mint-flavored microcapsules by complex coacervation. Firstly, gelatin-Arabic gum dispersed peppermint oil in water medium to form micro-droplets, then the pH value of the system was adjusted, the solubility was reduced and condensed into capsules, and then glutaraldehyde was added for curing reaction. The test results show that the embedding efficiency of microcapsules is the highest when the mass ratio of mint essence to two sustained-release materials is 1:9. When the dosage of mint-flavor microcapsules is 10 ~ 15 mg/ cigarette, the aroma of cigarettes increases obviously, the irritation decreases, and the aroma of wild mint essential oil can also be coordinated with the cigarette aroma. The results show that the samples of mint-flavor microcapsules synthesized in this study greatly improve the aroma characteristics and smoking characteristics of cigarettes, which lays a foundation for the brand development of cigarettes and has a good application prospect.

Keywords: Mint-flavor; Microcapsules; Cigarettes.

1. Introduction

Microcapsule is a kind of hollow microcapsule which encapsulates liquid, solid or gas with natural or synthetic polymer materials, and its particle size is micro-nano. As an important core technology of tobacco industry, the material coated outside the microcapsule is called wall material and the encapsulated material is called core material. It has become one of the keys to form the quality characteristics of Chinese cigarettes and build the core technology of Chinese cigarettes [1]. Industry technicians can use different natural or synthetic flavors and fragrances to enhance and supplement cigarettes, thus forming different styles of cigarette products. Microcapsule technology creates a physical barrier between the wall material and the core material, and under appropriate conditions, the core material can be slowly released by heating and extrusion. Therefore, this technology is widely used in textiles, food, cosmetics, medicine, feed, pesticides and other fields [2-3].

By using microcapsule technology, the components that may react with each other can be made into microcapsule products respectively, so that they can be stabilized in a system, and each effective component can be released in an orderly manner and act at the corresponding time, thus improving the flavor and nutrition of the products. Related research shows that the volatile oils of wild mint, wild mint and chrysanthemum morifolium can play a role in modifying smoke, increasing aroma, reducing irritation and improving aftertaste when added to cigarette cut tobacco [4-5]. Studies have shown that [6], at present, microcapsule technology has been widely used in adhesives, sealants, scented paper and pharmaceutical production in the medical field. Using amylose as inclusion wall material to embed some flavor substances can not only effectively protect the embedded substances, but also artificially control the release and reduce unnecessary diffusion and evaporation [7-8]. However, in the smoking process, the first few mouthfuls of mint cigarettes synthesized by the above methods are full of flavor, while the last few mouthfuls are lacking, and the mint flavor is very weak

at the end of the cigarette. In addition, in the process of cigarette storage, mint spices are easy to volatilize and diffuse, which seriously shortens the effective storage period of products.

In order to make the cigarette style more obvious and attract consumers, companies compete to launch cigarettes with different styles and characteristics, among which the most influential one is mint cigarette [9-10]. In order to further find an efficient and reasonable cigarette flavoring process, this study used gelatin and Arabic gum as composite wall materials to synthesize mint-flavor microcapsules by complex coacervation. Through experiments, the application of mint-flavor microcapsules in cigarettes was preliminarily realized, so as to provide some reference for the development of mint-flavor cigarettes.

2. Overview of microcapsules

Microcapsule synthesis technology originated in 1950s. Microencapsulation is to embed solid, liquid and gaseous substances into tiny capsules and release them under certain conditions. The embedded material is called core material, and the embedded material is called wall material [11]. Simply put, the process of microencapsulation is called microencapsulation. After microencapsulating the components that react with each other, they exist stably in the same substance; Controlled release, controlling the slow release of substances such as flavors and fragrances at the most appropriate time and at the most appropriate rate; Masking bad flavor, such as bad smell, spicy taste, bitter taste, peculiar smell, etc. Prevent and delay food spoilage.

Spray drying is a process of dispersing feed liquid into fine droplets by atomizer, and quickly evaporating solvent in hot dry medium to form dry powder products. Generally, spray drying includes four stages: atomization of feed liquid, contact and mixing of mist with hot drying medium, evaporation drying of mist droplets, and separation of dried products from drying medium. Its disadvantage is that the particles are too small, which makes the fluidity poor; When the operation is not controlled properly, more spices will be adsorbed on the surface of the capsule and oxidized, which will affect the flavor; Moreover, in the drying process, in order to evaporate water quickly, the drying temperature will be relatively high, which is easy to cause the loss of highly volatile spices [12].

Extrusion embedding method is a new processing method, which is carried out at low temperature. This method is more suitable for heat-sensitive substances. The process is that the core material is dispersed into the molten carbohydrate as the wall material, and then the mixture is discharged into the water absorbent to dehydrate and harden it, and the product is obtained after separation and drying. Its disadvantage is that the yield is not high, only 70%, while the spray drying method can reach 90% ~ 95%. In addition, its hard candy particle nature also greatly limits its application in some food systems.

The formation process of coagulation method can generally be divided into three stages, namely, the formation of coagulation phase, the deposition of wall film and the curing of wall film. Coagulated phase is to add another substance to the mixture of core material and wall material, or to reduce the solubility of wall material and condense from the mixed solution to produce a new phase; Microcapsules produced by coagulation method have the function of controlled release and are a very promising method.

3. Research method

3.1 Test raw material

The test materials are shown in Table 1:

Table 1: Test raw material

reagent	specifications	source
gelatin	Chemically pure CP	National medicine group chemical reagent co., ltd
arabic gum	Biochemical reagent BR	National medicine group chemical reagent co., ltd
Mint essence	WY2628	Wuhan yellow crane tower flavors and fragrances co., ltd
absolute ethyl alcohol	Analytical pure AR	Nanchang Lan Xiang Chemical Co., Ltd.
Maltodextrin	Food grade	Shandong xiwang sugar industry co., ltd
anhydrous sodium carbonate	Analytical pure AR	Shanghai Hongguang chemical plant
citric acid	Analytical pure AR	Shanghai Reagent No.1 Factory
glutaric dialdehyde	Biochemical reagent BR	National medicine group chemical reagent co., ltd

3.2 Laboratory apparatus

The experimental instruments are shown in Table 2:

Table 2: Laboratory Instruments

Instrument name	type	source
centrifuge	TGL-10B	Shanghai Anting scientific instrument factory
p H meter	PHS-3C	Shanghai Yidian Scientific Instrument Co., Ltd.
ultraviolet and visible spectrophotometer	UV752	Shanghai Jinghe analytical instrument co., ltd
liquid chromatograph	Syltech 500	Beijing wenfen analytical instrument technology development co., ltd
Heat-collecting constant-temperature heating magnetic stirrer	DF-101S	Gongyi city yuhua instrument co., ltd
Super electronic constant temperature water bath	HWS12	Shanghai yiheng technology co., ltd
Emulsification homogenizer	FA25	laboratory
Electric blowing oven	HD101A	Dongguan Fiona Fang Instrument Co., Ltd.

3.3 Synthesis of mint-flavored microcapsules

Microcapsule technology is a technology of embedding solid or liquid with film-forming materials to form tiny particles. It can use natural or synthetic polymer materials to encapsulate sensitive substances into micron or nanometer particles, which has the functions of protecting core materials from environmental impact, isolating active components, reducing volatility and controlling the release of core materials.

Arabic gum has good emulsifying properties, especially suitable for oil-in-water emulsion system. It can be used as microcapsule film-forming agent to convert raw materials into powder form, thus preventing substances from oxidative deterioration in the external environment. There are a lot of hydroxyl, carboxyl and amino groups in the molecular structure of gelatin, which makes it extremely hydrophilic. It is insoluble in cold water but has good solubility in hot water.

Gelatin can cross-link with glutaraldehyde and other aldehyde cross-linking agents, thus making the microcapsule wall structure compact and not affected by external factors. At the same time, gelatin is widely used as the wall material of microcapsules because of its biocompatibility, biodegradability, film forming, emulsification and gel forming.

In this paper, gelatin and Arabic gum are used as composite wall materials, and mint-flavor microcapsules are synthesized by complex coacervation method. The synthesis principle is that firstly, gelatin and Arabic gum disperse peppermint oil in water medium to form droplets, and then the p H value of the system is adjusted. When the p H value is less than the isoelectric point of gelatin, gelatin exists in the form of polycations, which can generate electrostatic interaction with Arabic gum which exists as polyanion in the system, and the charges attract each other to form positive and negative ion complexes, and then the solubility is reduced to form capsules, and then it is added.

The synthesis of peppermint microcapsules is as follows:

- 1) Gelatin, Arabic gum solution and emulsifier solution with a certain concentration were synthesized respectively.
- 2) Emulsify peppermint oil and emulsifier solution with an emulsifying homogenizer at high speed for 1min, then slowly add gelatin and Arabic gum solution, and emulsify together for

- 3min.
- 3) Under the heating of 50°C water bath, the mixed solution was stirred with a mechanical stirrer, and the pH was adjusted to a certain value with citric acid for about 15 minutes, then the temperature was lowered to below 15°C, then the pH was adjusted to about 10.0 with anhydrous sodium carbonate, and the glutaraldehyde was added to continue the reaction for a period of time.
 - 4) The microcapsule suspension was dried by spray dryer to obtain microcapsule powder.

3.4 Analysis and test of mint-flavored microcapsules

3.4.1 Thermal stability test

The thermal stability of microcapsules was tested by thermogravimetric analyzer. The sample was heated from 50°C to 500°C at the rate of 10°C/min in nitrogen atmosphere.

3.4.2 Determination of transfer amount of mint essence in mainstream smoke

After leaving the cigarette samples for 12 months, according to the industry standards and gas chromatography, the content of mint essence in the cut tobacco, filter rods and mainstream smoke of the cigarette samples was accurately determined.

3.4.3 Determination of emulsion stability

Put the microcapsule emulsion into a calibrated test tube with a stopper, the speed of the centrifuge is 2 000 r/min, the temperature is 20°C, and the centrifugation time is 10 min. Observe the height of the emulsified layer and the free layer after centrifugation, and calculate the stability ES of the emulsion.

$$ES = \frac{(EH - FH)}{EH} \quad (1)$$

EH is the total height of emulsion and FH is the height of free layer.

3.4.4 Sensory quality test of cigarettes

Microcapsules with known oil content were added to the contact between cigarette filter and cut tobacco according to a set amount, and then samples were rolled according to the standard of (0.82±0.01) g of total weight of each cigarette. After being balanced in a constant temperature and humidity box with a temperature of (21±3)°C and a relative humidity of (62 ±5)% for 24 h, the samples were evaluated by a five-person sensory evaluation team.

4. Results and discussion

4.1 Thermogravimetric analysis of microcapsules

Fig. 1 is a thermogram of microcapsules synthesized with emulsifier concentration of 0.5%, crosslinking agent dosage of 5.5mL, stirring speed of 700rpm, wall material concentration of 1.5%, core-wall ratio of 1:3, and re-coacervation pH value of 3.5.

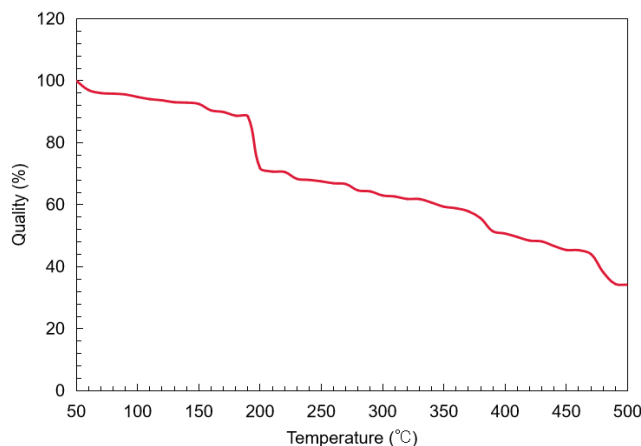


Figure 1 Thermogravimetric curve of microcapsules

The mass loss occurs when the temperature is about 100°C, which is mainly caused by the low boiling point components in peppermint oil and the residual water in the sample itself. The relatively gentle mass loss in the temperature range of 100°C ~ 200°C is mainly caused by the continuous evaporation and release of high boiling point peppermint oil components in microcapsules.

After 200°C, there is a large gradient of mass loss, which is caused by the decomposition of microcapsule wall materials. It can be proved that the wall material can protect peppermint oil, make peppermint oil resistant to high temperature, delay its violent volatilization or decomposition, and achieve the effect of embedding.

4.2 Analysis of transfer quantity of mint essence in mainstream cigarette smoke

Figure 2 lists the changes of mint essence content in cigarettes (filter+cut tobacco) and mint essence transfer amount in mainstream smoke after storage.

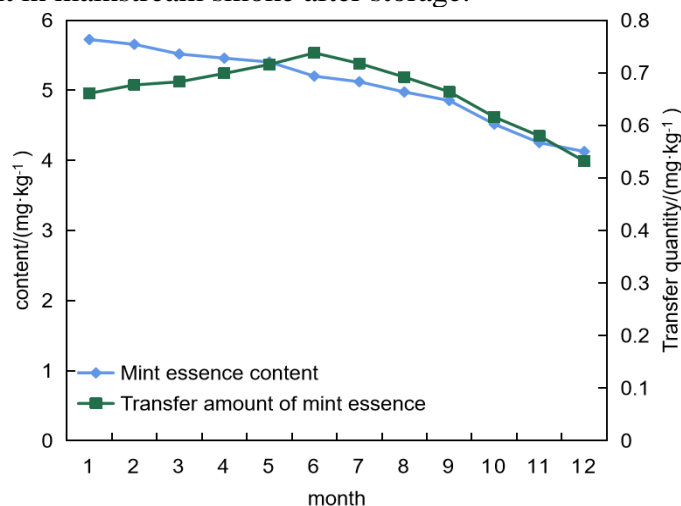


Figure 2 Transfer amount of mint essence in mainstream cigarette smoke

It can be seen that with the extension of cigarette storage time, the content of mint essence in cigarettes (filter+cut tobacco) showed a downward trend, and the decline was more obvious after the fifth month. The content of mint essence in mainstream smoke did not change much in the first 9 months, but the transfer amount was the highest in the fifth month. Therefore, it is feasible to characterize the coolness of cigarettes by the content of mint essence in mainstream smoke. It can be seen that the mint additive synthesized in this experiment has strong durability and can meet the needs of cigarette circulation and storage.

4.3 Effect of mass ratio of mint essence to sustained-release material on microcapsule effect

Under the same technological conditions, the ratio of gelatin to anhydrous sodium carbonate is 28:70, and the mass ratios of mint essence to sustained-release materials are 1:3, 1:5, 1:7, 1:9, 1:11, 1:13, 1:15, and other conditions are unchanged. The test results are shown in Figure 3.

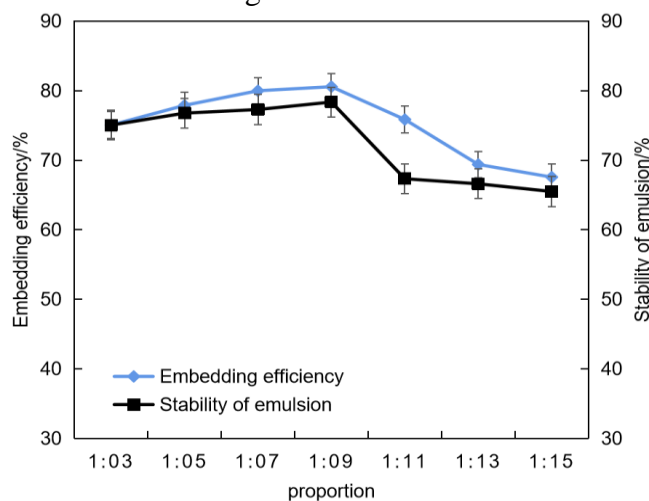


Figure 3 Effect of mass ratio of mint essence to sustained-release material on encapsulation efficiency and emulsion stability of microcapsules

The core material of this experiment is mint essence, and the mass ratio of mint essence to slow-release material has a great influence on the formation process of emulsion, which has a great influence on the embedding efficiency of mint essence microcapsules synthesized by spray drying. When the mass ratio of mint essence to two slow-release materials is 1:9, the embedding efficiency of microcapsules is the highest.

4.4 Test and analysis of cigarette sensory quality

Mint-flavored microcapsules were added to cigarettes, and the smoking evaluation results are shown in Table 3. The oil content of peppermint microcapsules used for flavoring was determined to be 42.71%.

Table 3: Sensory quality test of cigarettes

additive amount(mg·kg ⁻¹)	Essential oil content(mg·kg ⁻¹)	Test description
0	0	The aroma is average and irritating
5	2.915	The increase of aroma is not obvious, and the irritation is weakened
10	5.212	When the amount of aroma increases, the coordination increases and the irritation decreases obviously
15	5.647	The aroma increased obviously, the coordination decreased, and the mint aroma was obvious
20	6.469	The aroma of mint is obvious, which can't be coordinated with the smell of tobacco, and the aftertaste is not comfortable and the stimulation is weakened.

From the analysis in Table 3, it can be seen that when the amount of mint-flavor microcapsules is 10 ~ 15 mg/ cigarette, the aroma of cigarettes increases obviously, the irritation decreases, and the aroma of mint essential oil can also be coordinated with the cigarette aroma.

5. Conclusion

In this study, gelatin and Arabic gum were used as composite wall materials, and mint-flavor microcapsules were synthesized by complex coacervation method. Through experiments, the application of mint-flavor microcapsules in cigarettes was initially realized. The test results show that the wall material can protect peppermint oil, make peppermint oil resistant to high temperature, delay its violent volatilization or decomposition, and achieve the effect of embedding. The mint additive synthesized in this experiment has strong durability and can meet the needs of cigarette circulation and storage. When the mass ratio of mint essence to two slow-release materials is 1:9, the embedding efficiency of microcapsules is the highest. When the dosage of mint-flavor microcapsules is 10 ~ 15 mg/ cigarette, the aroma of cigarettes increases obviously, the irritation decreases, and the aroma of mint essential oil can also be coordinated with the cigarette aroma. The results show that the addition of mint-flavor microcapsules can increase the aroma of cigarettes, reduce irritation and improve aftertaste, and it is a good cigarette flavor. The synthesis process of peppermint microcapsules is simple, which can effectively reduce the thermal volatilization loss of essential oil and has good application value.

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