# Microencapsulation of Thyme Essential Oil: A Sustainable Approach for Aroma Finishing of Textiles


## ABSTRACT

**Background:** The current ‘green’ trend is favoring usages of natural ingredients with an increasing interest in the health and wellness benefits of herbal products. Aroma finishing enhances the value of the product by incorporating different fragrance into fabrics. However, longevity of aroma on the textile with passage of time and subsequent launderings is a major concern that can be solved through microencapsulation of core materials protecting them from the surrounding environment. Hence, the present study was conducted to develop microcapsules of thyme essential oil for aroma finishing of textiles.

**Methods:** To achieve the objectives of study, thyme oil was selected as core material and gum acacia and gelatin as wall materials. Complex coacervation technique of microencapsulation was used for development of thyme oil microcapsules. The ratios of core and wall materials, temperature and pH were optimized for standardization of microencapsulation process based on size, distribution and quality of walls of formed microcapsules examined under inverted microscope.

**Result:** The 1.5:4:4 ratio of thyme oil:gum:gelatin at 45$^{o}$C temperature with initial pH 4.5 and final pH 7 were found optimum for development of microcapsules as medium sized microcapsules having good uniformity in size and distribution with sharp and thick walls were obtained at these optimized process parameters. Thus, these optimized variables can be used to prepare thyme oil microcapsules for development of multi-functionalized textiles.



**Key words:** Thyme oil, Gum acacia, Gelatin, Microcapsules, Complex coacervation, Multifuctional textiles




## INTRODUCTION

In present decade, the consumers’ needs, demands and expectations for a healthier and more comfortable life are increasing everyday even when it comes to clothing. After fabricating the mansions of fashion and comfort, textiles are now moving towards high-tech era of performance, which has brought up diversification and expansion of technologies. This realization of technologists has coincided with rapid development in technology and brought about a surge in research and development activities in textiles. The recent interest in multifunctional textiles is mainly directed to the use of specific textiles in medical therapy and prevention of deficiencies. With advent of new eco-technologies, the growing needs of the consumers in the wake of health and hygiene can be fulfilled without compromising the issues related to safety, human health and environment (Sanjay and Malpani, 2013; Thite and Gudiyawar, 2015).

Aroma or fragrance finishing is considered as an emerging area which has tumbledown the textile industry that enhances the value of the product by utilizing the controlled release of different fragrance into fabrics, leading to the production of fragranced fabrics. The fragrance applied by use of essential oil not only provides a pleasant smell but also the beneficial effect of aromatherapy (Kumar *et al.,* 2012). Essential oils are complex mixtures of volatile compounds particularly abundant in aromatic plants, mainly composed of terpenes including monoterpenes and sesquiterpenes. These oils are the concentrated compounds that have long lasting effect onto the substrate and known as the important elements around the world in aromatherapy, natural perfumery, insect repellent and in various fields of natural medicine, psychology, physiology, pharmacology and cosmetology (Dhifi *et al.* 2016).

Thyme oil is one of the essential oils that have multifunctional values and it can be used as an eco-friendly finishing agent to impart functional properties to textiles. Thyme (*Thymus Vulgaris L.*) is an important medicinal plant which belongs to the Lamiaceae family and commonly known as *Ajwain*. The various parts of thyme have medicinal properties and are used for treatment of different ailments. The main phenolic component in thyme is thymol (2-isopropyl-5-methylphenol) which is used in alcohol solutions and in dusting powders for the treatment of ringworm and hookworm infections. The thyme essential oil is extracted from the fresh flowering aerial parts of the plant by steam distillation method. It is white coloured crystalline compound possessing aromatic odour and has strong antiseptic, antioxidant, antibacterial and antifungal properties (Dauqan and Abdullah, 2017).

Microencapsulation is an effective and important tool to prepare oil-based high quality and health-beneficial products in order to improve their chemical, oxidative and thermal stability. The microencapsulation process is used to develop and modify textiles with new improved properties such as polychromic, thermo chromic, fire proofing, fragrance releasing, cosmetic, therapeutic and medical textiles (Wijesirigunawardana and Perera, 2018). Nowadays, more profitable uses of microencapsulation can be found in the textile industry for its capability of being adjusted for different functions. Aroma products with microcapsules could be applied to almost all industrial products, such as papers, plastics, paints, scented stamps, cellular phones, greeting cards as well as textiles, thereby creating scented clothing (Anitha *et al.,* 2011).

## MATERIALS AND METHODS

**Selection of essential oil and wall materials**

Thyme essential oil was selected for the present study on the basis of its aromatic and therapeutic properties, ease of availability and cost effectiveness and provided by Emmbros Overseas Lifestyle Pvt. Ltd., Haryana, India. Gum acacia and gelatin were used as wall materials to encapsulate the core material and purchased from chemical suppliers of Haryana, India. Acetic acid, sodium hydroxide and formalin were also used in the study.

**Selection of microencapsulation technique**

For preparation of microcapsules, many physical and chemical techniques can be used such as solvent evaporation, polymerization, spray drying, pan coating, phase separation-complex coacervation, centrifugal extrusion etc. Out of all these, phase separation- complex coacervation technique was selected for the present study on review basis and the suitability of the process to be carried out in the laboratory of the Department of Textile and Apparel Designing, I.C. College of Home Sciences, CCS Haryana Agricultural University, Hisar.

**Standardization of microencapsulation process**

For preparation of standardized microcapsule gel of thyme essential oil, experiments were carried out with different ranges of variables of microencapsulation process i.e. ratios of oil, gum and gelatin, temperature and pH for optimization on review basis. The recipe standardized by Kumari, 2015 was followed with some modifications for preparation of microcapsules. 16 g of gelatin was weighed and dissolved in 25 ml warm water and then stirred using a high speed stirrer for 10 minutes. 4 g of thyme essential oil was added to the solution at 50°C. 16 g of gum acacia was weighed and dissolved in 25 ml warm water separately. The gum acacia solution was added to the gelatin solution and the temperature of the solution was maintained at 50°C. The pH of the solution was decreased to 4.5 by adding dilute acetic acid. The solution was then stirred at high speed for 20 minutes. The pH of the solution was increased to 8.5 using sodium hydroxide solution to form microcapsule gel. For stabilization, 1 ml of 17 percent alcoholic formalin was added to the formed capsules. The resultant precipitate obtained was observed under inverted microscope to ensure the formation of microcapsules and images were captured after each step of microencapsulation process. The combinations of the ratios of oil, gum and gelatin which produced the desired results were selected for further optimization of other variables. At a time, the ratio or condition of only one variable was varied and other variables were kept constant

**Optimization of essential oil ratio**: To determine the optimum ratio of essential oil, six different ratios of thyme essential oil i.e. 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 were taken while all other variables i.e. gum, gelatin, temperature and pH were kept constant. To ensure the formation and quality of microcapsules, the resultant precipitate obtained was examined under inverted microscope. Microcapsules of medium size with good uniformity in size and distribution having sharp and thick walls were considered as best and appropriate. The ratio at which microcapsules formed of the best quality was selected as optimized ratio of essential oil for further experimental work.

**Optimization of gum ratio:** For determining the optimum ratio of gum, six different ratios of gum i.e. 1, 2, 3, 4, 5 and 6 were taken with optimized ratio of essential oil whereas all other variables were kept constant. Process was carried out to prepare microcapsule gel and ratio of gum was optimized on the basis of microscopic assessment of microcapsules under inverted microscope.

**Optimization of gelatin ratio:** To optimize the ratio of gelatin, six different ratios of gelatin i.e. 1, 2, 3, 4, 5 and 6 were taken with optimized ratio of oil and gum and all other variables were kept constant. Process was carried out to prepare microcapsule gel and ratio of gelatin was optimized on the basis of assessment of microcapsules.

**Optimization of temperature:** For determination of the optimum temperature for microencapsulation, the process was carried out at different temperatures i.e. 30, 35, 40, 45, 50, 55 and 60$^{o}$C with optimized ratios of oil, gum and gelatin and values of initial and final pH were kept constant. Temperature was optimized on the basis of assessment of microcapsules under inverted microscope.

**Optimization of initial and final pH:** The pH plays important role in microencapsulation as it is considered responsible for phase separation which leads to capsule formation. For optimization of pH, the optimized ratios of essential oil, gum and gelatin at optimized temperature was set to initial pH 4.0, 4.5, 5.0, 5.5, 6.0, 6.5 and 7.0. The microencapsulation process was carried out till gel formation took place and then final pH was maintained at 7.0, 7.5, 8.0, 8.5, 9.0, 9.5 and 10.0 with each initial pH. The initial and final pH was optimized on the basis of microscopic assessment of microcapsules.

## RESULTS AND DISCUSSION

The perusal of Table 1 and Image 1 indicate that thyme oil microcapsules were formed in all ratios of oil, gum and gelatin but only three ratios i.e. 0.5:4:4, 1:4:4 and 1.5:4:4 exhibited good uniformity in size and distribution and thick walls of microcapsules. However, at ratio 1.5:4:4 of oil, gum and gelatin, microcapsules of medium size with good uniformity in size having thick and sharp walls of capsules were formed which were considered as best. Therefore, 1.5 ratio of thyme essential oil was selected for carrying out further optimization. The data shown in Table 2 and visual assessment of microcapsule gel (Image 2) reveal that good microcapsules of thyme essential oil were formed in four ratios of oil, gum:gelatin. However at 1.5:5:4 and 1.5:6:4 ratios of oil:gum:gelatin, large sized and less microcapsules were formed which were not considered appropriate for microencapsulation process. The microcapsules formed with 4 ratio of gum acacia to essential oil was optimized and used for further optimization because best microcapsules were formed at this ratio.

It is evident from data in the Table 3 and microscopic images of microcapsules (Image 3) that the microcapsules formed in the ratio of 1:4:4 were medium sized, having good uniformity in size and distribution and the walls were also sharp and thick as compared to the capsules formed with other ratios of oil, gum and gelatin. Therefore, the ratio 4 of gelatin was used for further development of microcapsules. At higher ratios of gelatin i.e. 1.5:4:5 and 1.5:4:6, large sized microcapsules were formed which was not considered as an appropriate size for microcapsules. Further it was found that when the amount of oil, gum and gelatin was increased, the size of microcapsule became larger but the walls of the capsules started rupturing and lumps were formed due to disproportionate ratio of oil, gum and gelatin. The results of the study are in line with Senem *et al.* (2018) that with the increase in amount of oil and wall material the size of microcapsules and their oil content increased but they tended to agglomerate. It was inferred that after a certain concentration, no capsules were formed. Bhatt and Singh (2018) also used complex coacervation technique to prepare lemongrass microcapsules and found that 1:4:4 ratio of oil:gum:gelatin was the most appropriate to form good quality microcapsules.

It is obvious from Table 5 and visual assessment of microcapsule gel that microcapsules of the thyme essential oil formed at 45$^o$C temperature were medium sized with uniform distribution having thick and sharp walls (Image 4). At higher temperatures, larger sized and thick walled microcapsules were formed which were not considered appropriate. Hence, 45$^o$ C temperature was considered suitable for the development of thyme essential oil microcapsules. Essential oils are volatile in nature and they evaporate very easily, therefore with the increase in temperature, size of the microcapsules was also noticed to be increased but they ruptured immediately. Higher temperatures than 60°C were also tried but no microcapsules were formed on those temperatures. El-Molla *et al.* (2015) also emphasized that the best temperature for microcapsule formation was between 40-50$^o$C. The results are also in corroboration with the study of Kumari *et al.* (2017), Srivastava and Verma (2017) and Sannapapamma *et al.* (2018).

The microcapsules formed at different ranges of initial and final pH were analyzed to determine the optimum pH values. The data in Table 5 and microscopic analysis of the microcapsule gel (Image 5) display that the microcapsules of thyme essential oil formed at initial pH 4.5 and final pH 7.0 were medium sized, had good uniformity in size and distribution and the walls of capsules were also sharp and thick as compared to capsules formed at other ranges of pH. Therefore, these pH values were chosen as optimum initial and final pH for preparation of microcapsules of thyme oil. Dima *et al*. (2016) reported that adjusting initial pH between 4.0 to 5.0 is a very important step in phase separation for microcapsules formation. Findings of Siow and Ong (2013) also support the results as they reported that highest oil encapsulation yield and loading efficiency occurred at initial pH 4.5 and final pH was kept 8.5 for application on cotton fabric. Pakzad and Alemzadeh (2018) reported that best microcapsules of peppermint oil were formed at initial pH 4.0 in complex coacervation technique.

## CONCLUSION

The 1.5:4:4 ratio of essential oil, gum and gelatin at temperature 45$^{o}$C with initial pH 4.5 and final pH 7 were optimized as the medium sized microcapsules having good uniformity in size and distribution with sharp and thick walls were obtained at these optimized ratios and conditions. The best microcapsules were obtained when the ratio of thyme essential oil was lower as compared to wall materials. High temperature of microencapsulation process produced large sized microcapsules which due to thin capsule wall ruptured immediately. Complex coacervation technique of microencapsulation for preparation of thyme essential oil microcapsules is very efficient to acquire long term sustained finish. The thyme essential oil have many reported therapeutic properties therefore microencapsulated thyme essential oil treated fabrics can be used for apparel, home and healthcare textiles.



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**Table 1:** Optimization of thyme essential oil ratio in microcapsule gel

| Ratio of oil: gum: gelatin | Size of microcapsules | Uniformity in size and distribution | Wall of Microcapsules | Ranks |
|---|---|---|---|---|
| 0.5:4:4 | Very small + Small | Average | Very thick | III |
| 1.0:4:4 | Small +Medium | Good | Thick | II |
| **1.5**:4:4 | **Medium** | **Good** | **Sharp + Thick** | **I** |
| 2.0:4:4 | Large | Poor | Very thick | IV |
| 2.5:4:4 | Very large | Poor | Very thick + Ruptured | V |
| 3.0:4:4 | Very large | Poor | Very thick + Ruptured | VI |

**Table 2:** Optimization of gum acacia ratio in microcapsule gel

| Ratio of oil: gum: gelatin | Size of microcapsules | Uniformity in size and distribution | Wall of Microcapsules | Ranks |
|---|---|---|---|---|
| 1.5:1:4 | Very small + Small | Average | Thick | IV |
| 1.5:2:4 | Small | Average | Very thick | III |
| 1.5:3:4 | Small +Medium | Good | Thick | II |
| 1.5:**4**:4 | **Medium** | **Good** | **Sharp + Thick** | **I** |
| 1.5:5:4 | Very large | Poor | Very thick | V |
| 1.5:6:4 | Large + Very large | Very poor | Thick + Ruptured | VI |

**Table 3:** Optimization of gelatin ratio in microcapsule gel

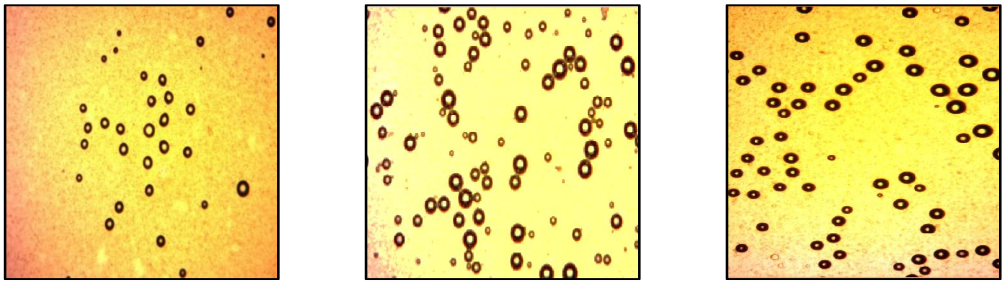
| Ratio of oil: gum: gelatin | Size of microcapsules | Uniformity in size and distribution | Wall of Microcapsules | Ranks |
|---|---|---|---|---|
| 1.5:4:1 | Large | Average | Very thick | IV |
| 1.5:4:2 | Small + Medium | Average | Thick | III |
| 1.5:4:3 | Small + Medium | Good | Thick | II |
| 1.5:4:**4** | **Medium** | **Good** | **Sharp + Thick** | **I** |
| 1.5:4:5 | Large | Average | Thick + Ruptured | V |
| 1.5:4:6 | Very large | Poor | Very thick + Ruptured | VI |

**Table 4:** Optimization of temperature for microencapsulation process

| Temperature (°C) | Size of microcapsules | Uniformity in size and distribution | Wall of Microcapsules | Ranks |
|---|---|---|---|---|
| 30 | Very small | Poor | Thick | VI |
| 35 | Small | Average | Very thick | V |
| 40 | Small + Medium | Good | Thick | II |
| **45** | **Medium** | **Good** | **Sharp + Thick** | **I** |
| 50 | Large | Average | Very thick | III |
| 55 | Large | Average | Very thick | IV |
| 60 | Very Large | Poor | Thick + Ruptured | VII |

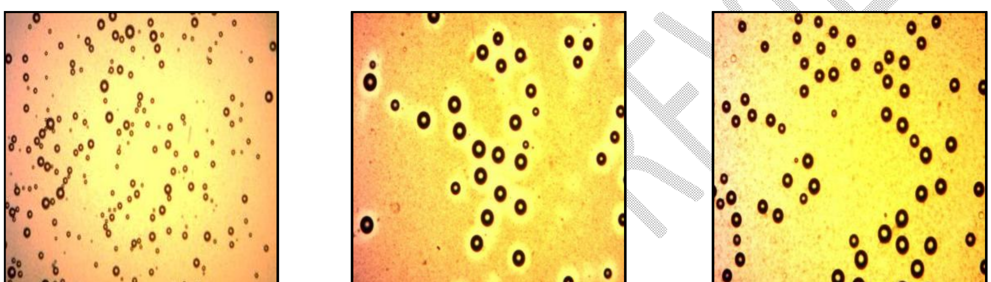
**Table 5:** Optimization of pH for microencapsulation process

| pH initial/final | Size of microcapsules | Uniformity in size and distribution | Wall of Microcapsules | Ranks |
|---|---|---|---|---|
| 4.0/7.0 | Very small | Poor | Very thick | IX |
| 4.0/7.5 | Small | Average | Thick | VI |
| 4.0/8.0 | Medium + Large | Good | Sharp + Thick | II |
| 4.0/8.5 | Very large | Poor | Very thick + Ruptured | XI |
| **4.5/7.0** | **Medium** | **Good** | **Sharp + Thick** | **I** |
| 4.5/7.5 | Medium | Average | Thick | V |
| 4.5/8.0 | Very small + Small | Average | Thick | VII |
| 4.5/8.5 | Large | Poor | Thick + Ruptured | X |
| 5.0/7.0 | Medium | Average | Thick | IV |
| 5.0/7.5 | Medium + Small | Good | Sharp + Thick | III |
| 5.0/8.0 | Medium + Large | Average | Thick + Ruptured | VIII |
| 5.0/8.5 | Very large | Poor | Ruptured | XII |
| 5.5/7.0 | Medium+ Large | Average | Very thick | XIII |
| 5.5/7.5 | Large | Average | Thick + Ruptured | XIV |
| 5.5/8.0 | Very large | Poor | Ruptured | XV |
| 5.5/8.5 | Large + very large | Very poor | Very thick + Ruptured | XVI |



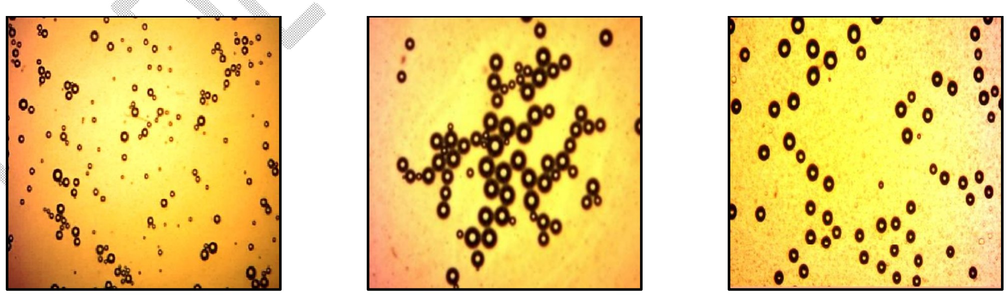
0.5:4:4 (Rank-III) 1:4:4 (Rank-II) 1.5:4:4 (Rank-I)

**Image 1:** Inverted microscopic images of microcapsules at different ratios of thyme essential oil



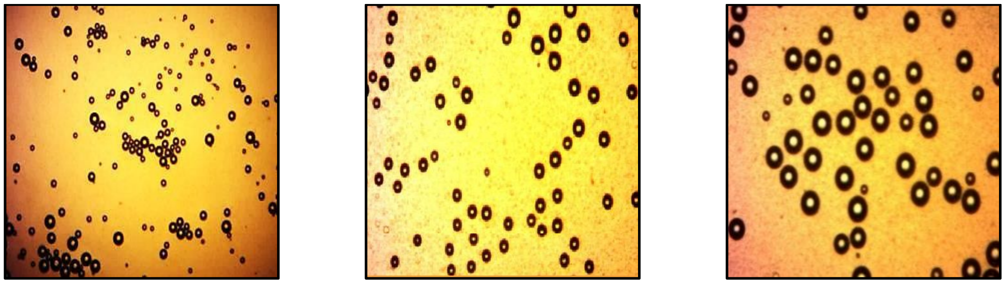
1.5:2:4 (Rank-III) 1.5:3:4 (Rank-II) 1.5:4:4 (Rank-I)

**Image 2:** Inverted microscopic images of microcapsules at different ratios of gum acacia



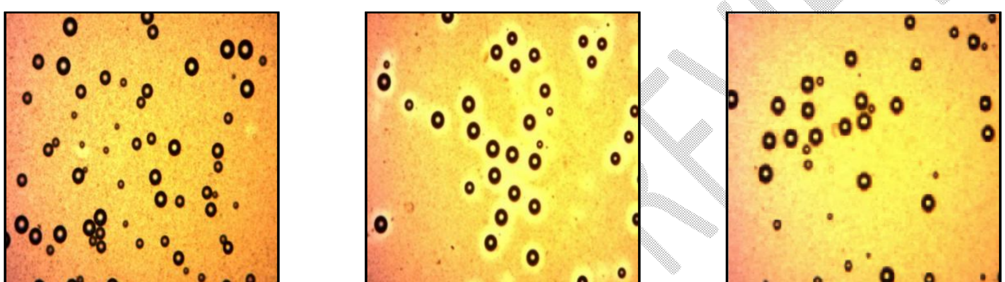
1.5:4:2 (Rank-III) 1.5:4:3 (Rank-II) 1.5:4:4 (Rank-I)

**Image 3:** Inverted microscopic images of microcapsules at different ratios of gelatin



40°C (Rank-II) 45°C (Rank-I) 50°C (Rank-III)

**Image 4:** Inverted microscopic images of microcapsules at different temperatures



4.0/8.0 (Rank-II) 4.5/7.0 (Rank-I) 5.0/7.5 (Rank-III)

**Image 5:** Inverted microscopic images of microcapsules at different initial and final pH