EFFECT OF TEMPERATURE IN THE EMULSIFICATION STEP ON THE YIELD AND QUALITY OF THE ROSE OIL FILLED UREA-FORMALDEHYDE MICROCAPSULES PREPARED BY INTERFACIAL *IN SITU* POLYMERIZATION METHOD

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ABSTRACT

In the present work, the influence of reaction temperature in the emulsification step on the efficiency of the encapsulation process of rose oil by in situ polymerization, using urea - formaldehyde resin, forming the shell of the microcapsules, was studied. The process was carried out at the following temperatures: 40°C, 50°C, 60°C and 70°C. It was established that the effect of temperature during this step is ambiguous, as during the first sub - step (stage A) of the emulsification step, its increase leads to an increase in the efficiency of the encapsulation process, the yield and quality of the obtained microcapsules, as well as a decrease in the size of the obtained microcapsules. This is mainly due to a reduction in the size of the microdroplets obtained during this stage, determining the size of the future microcapsules. In the second stage of the emulsification step, the increase in temperature to certain values (from 40°C to 50°C) also leads to an increase in the efficiency of the process, the yield and the quality of the obtained microcapsules. Increasing the temperature above 50°C (60 and 70°C) leads to desorption of the pre - polymer particles from the surface of the microdroplets, and from there to deterioration of the quality of the capsule shell, which leads to a decrease in the yields and quality of the microcapsules. For these reasons, the author chooses a temperature optimum of 45°C for this stage, changing the other conditions and observing the effect of this change.

Keywords: microencapsulation, in situ polymerization, rose oil, mono methylol urea, pre - polymer, emulsification step, temperature, microdroplet.

INTRODUCTION

In situ polymerization is one of the typical chemical methods for preparing microcapsules. The use of urea and formaldehyde as materials for the construction of the capsule wall makes it possible to obtain a strong capsule shell, providing, on the one hand, long - term stability of the encapsulated substance, and on the other hand, allowing a long - lasting effect of the active substance, released by its gradual passage through the microcapsule wall [1 - 3]. In this method, in the first step a pre - polymer (monomethylolurea) is obtained, which in the emulsification step is adsorbed on the surface of the obtained microdroplets. As a result of the polycondensation between individual molecules of mono methylol urea, the urea - formaldehyde capsule wall is formed, characterized by exceptional strength, flexibility and elasticity. Since the size of the microdroplets obtained in the first sub - step (stage A) of the emulsification step determines the size of the future microcapsules, and the intensity of the pre-polymer adsorption process on the surface of the formed microdroplets in the second sub - step (stage B) determines the other characteristics such as yield, thickness and quality of the capsule shell, as well as content of the encapsulated substance, this led us to study the influence of the conditions during the emulsification step. These conditions are stirring speed,

temperature and time. Here, the focus is on studying the influence of temperature during the emulsification step (stage A and stage B), on the yield and quality of the obtained rose oil microcapsules. For this purpose, the influence of temperature was studied, varying it in the range between 40°C and 70°C.

The temperature (40°C, 50°C, 60°C and 70°C) was varied both during the first sub - step and during the second sub - step of the emulsification step, whereas the stirring speed, the stirring time and the emulsifier concentration were constant. In the first sub - step, when the temperature varied, the stirring speed was 1500 rpm as well as the time was 3.5 h. In the second sub - step, after receiving the milk - like emulsion, where the temperature was changed, the stirring speed was 1500 rpm and the time was 2.5 h.

It is particularly important to monitor the temperature during the emulsification step, as on the one hand it affects the size of the resulting microdroplets, and on the other hand it influences and controls the adsorption/desorption of the pre - polymer particles onto/from the surface of the newly formed microdroplets. Initial results with the encapsulation of rose oil show that high temperature stirring for about 3 hours is necessary to obtain a milky white emulsion. [4]. After obtaining the milk - like emulsion, the temperature must be lowered to ensure normal conditions for the adsorption of the pre - polymer molecules on the surface of the newly formed microdroplets, and on the other hand, to prevent the desorption process of the adsorbed pre - polymer molecules. Under these conditions, the reaction mixture was kept for another 2 h. As the temperature increases, the size of the capsules decreases, but the initial results show that the influence of temperature on this characteristic is not so great compared to the influence of the speed and time [4]. For example, at a temperature of 40°C the size of the capsules varies between 90 and 70 μm, and at 70°C this size moves between 50-30 μm. This tendency of temperature increasing should also not continue throughout the emulsification step, for the reasons stated above. According to the initial data, there is no significant influence of the parameter on another characteristic such as capsule yield, which, although growing, is not so great [4]. The yield of the encapsulated substance (EE, %) as well as the content of the encapsulated substance (E% core), are not affected by temperature as much as they are by the time and stirring speed [4]. As mentioned, during the emulsification step, the microdroplets are formed around which the pre - polymer particles are adsorbed. Over time, the amount of adsorbed pre - polymer particles increase. Along with the adsorbed particles, however, the number of desorbed ones increases over time. Since adsorption is an equilibrium process, the equilibrium constant of the process is affected by temperature.

An analogy with static membranes could be made, where the Henry's adsorption constant directly depends on temperature according to Eq. (1) [5]:

$$K_H = \int_{-\infty}^{\infty} [\exp(-\beta u) - 1]\, dx \quad (1)$$

$$\beta = 1/k_B.T$$

where k_B is Boltzmann constant, T is the absolute temperature in kelvin degree.

Eq. (1) shows that the value K_H depends only on the potential u and the temperature T. This means that the increase in temperature affects the adsorption process, lowering the Henry's constant, and hence accelerating the desorption process. Thus, the conclusion should be made that to increase the amount of adsorbed pre-polymer particles on the surface of the microdroplets, a decrease in temperature is necessary after its initial increase.

The sorption/desorption phenomenon, in general, is an equilibrium process quantified by the corresponding equilibrium (sorption constant, Ks), representing a ratio between the rate constants of adsorption and desorption of the pre - polymer particles according to Eq. (2) [6 - 8]:

$$K_s = k_1 / k_{-1} \quad (2)$$

The adsorption and desorption processes are characterized by the corresponding rates depending on the concentrations of the adsorbed and desorbed molecules per unit area and representing the product of their rate constants and the molar concentrations of free mono methylol urea found in the solution and that on the surface of the corresponding microdroplet.

$$[MMU] \leftrightarrow [MMU^*] \quad (3)$$

Thus:

$v_1 = k_1 . [MMU]$ (4)

$v_{-1} = k_{-1} . [MMU^*]$ (5)

where [MMU] is the molar concentration of free molecules mono methylol urea, found in solution,

[MMU*] - molar concentration of adsorbed mono methylol urea molecules on the surface of the microdroplets.

When equilibrium is established, the rates of the two processes (adsorption and desorption) equalize:

$v_1 = v_{-1}$ then $k_1 . [MMU] = k_{-1} . [MMU^*]$ (6)

$K_s = k_1 / k_{-1} = [MMU^*]/[MMU]$ (7)

An increase in temperature leads to an increase in the rate of the desorption (v_{-1}) by influencing the rate constant of the process (k_{-1}), whereby it increases while Ks decreases. Accordingly, the equilibrium shifts to the left until new values are reached. At the same time, the increase in temperature in the second sub - step (stage B) of the emulsification step has no effect on the size of the obtained microcapsules. During the stage B of this step, after the formation of microdroplets, the size of which depends on the factors affecting the first sub - step (stage A) of the emulsification step, adsorption of the pre - polymer particles take place.

As mentioned above, to specify the conditions and to give more clarity about the effect of temperature, the microencapsulation of the essential oils was carried out at several different temperatures in each of the stages (stage A and B) of the emulsification step. In this regard, the process temperature was varied in the range of 40 to 70^{o}C during both stages of the emulsification step.

As will be seen from the data presented below, it is concluded that during the first sub - step (stage A) of the emulsification step, the change in temperature affects the yield (%), encapsulation efficiency (EE, %), capsule size and encapsulation factor (EF), as with increasing temperature all characteristics increase, except for capsule size, which decreases. During this sub - step (stage A) of the emulsification step, however, temperature does not affect the other two characteristics, namely resin efficiency (RE, %) and core content (E% core). In the second sub - step of the emulsification step (stage B), the change in temperature affects the yield, encapsulation efficiency (EE, %), resin efficiency (RE, %), E% core and encapsulation factor (EF), all of which increase with decreasing temperature, with except for E% core, which decreases. The change in temperature during this stage of the emulsification step does not affect the size of the capsules.

EXPERIMENTAL

Methods and materials

Urea as a technical product (with technical grade), sodium hydroxide, citric acid as well as formalin as a 37 % formaldehyde solution were purchased from Valerus. Rose oil was purchased from licensed Bulgarian producers. Glutaraldehyde and Sodium dodecyl sulfate (SDS) were purchased from Sigma Aldrich.

A professional benchtop pH meter: BANTE Instruments, Model 920 - UK with a combined pH electrode with BNC connection was used to control the pH of the reaction mixture. An electromagnetic stirrer with heating, DIAB, Model MS7 - H550 - S, 1030 W, 0 - 1500 rpm, + 30 - 550°C, was used. A homogenizer for solid and liquid media, Velp Scientifica, model OV5, 1000 - 22000 rpm, was used to regulate the stirring speed. Weight analyses were performed on precision analytical balances with internal calibration - "KERN" models ABJ 120 - 4NM and ABJ 220 - 4NM.

For image analysis, a CARL ZEISS JENA light microscope, model 30 - G0020a with magnifications of 12.5 x, 25 x, 40 x and 100 x, as well as a Nikon reflective optical metallographic microscope included in the CSEM Scratch tester equipment (Switzerland) and digitized with a 14 - megapixel camera were used. With these two microscopes, the microcapsules were initially determined in terms of their shape, morphology and approximate size. A laser diffraction apparatus brand MICROTRACK MRB model SYNC, with a working range of 0.01μm - 4mm was used for determination of the microcapsule size as well as their size distribution.

FT-IR analyses of rose oil microcapsules were carried out on PerkinElmer Spectrum™ 3 FT - IR apparatus (21 CFR Part 11 Compatible) operating at the wavelength range between 7800 cm^{-1} - 225 cm^{-1}. After their freeze drying using KBr pellets or NaCl crystals, the spectra of the prepared microcapsules were obtained.

Preparation of microcapsules

Pre-polymer synthesis step

In a three - necked round - bottomed flask with a volume of 1000 mL, equipped with a thermometer, a reflux condenser and a mechanical or electromagnetic stirrer, 120 g of urea (Mm = 60.06 g mol^{-1}; 2 mol) are placed. Then, with vigorous stirring, 240 mL of 37 % formalin (88.8 g formaldehyde, Mm = 30.03 $g.mol^{-1}$; 2.96 mol) are added to them, and the pH of the solution is adjusted to pH 8 - 8.3 by slowly adding dropwise 10 % NaOH [9 - 11]. The reaction mixture is heated in a water bath at a temperature of 70°C for about 1 hour, after which the flask is removed and tempered. After tempering of the flask, the reaction mixture is diluted with distilled water to 500 mL to obtain a pre - polymer solution of precisely defined concentration. Due to the decrease in the pH value of the mixture during the reaction and the creation of conditions for the formation of undesirable side by-products, the pH must be maintained in the range of 8 - 8.3 [9 - 11]. This is done by adding a dilute solution of sodium hydroxide dropwise. For this purpose, instead of sodium hydroxide solution, various salts can be used, such as ammonium chloride, ammonium carbonate, sodium acetate, sodium citrate, TRIS. HCl, as well as weak bases such as TRIS - base, melamine, urotropine, triethanolamine, etc [12 - 14].

Emulsification step

In an Erlenmeyer flask, 3 % sodium dodecyl sulfate (SDS) was added to 100 mL of pre-polymer solution at a constant stirring speed of 1500 rpm [4, 15]. The resulting pre - polymer and surfactant solution was transferred to a 500 mL round-bottom flask equipped with a thermometer, a reflux condenser, and a mechanical or electromagnetic stirrer/homogenizer. Then, 5 mL of essential oil was added to the pre-polymer and surfactant (SDS) solution, varying in the temperature (40°C, 50°C, 60°C, 70°C). The duration of the emulsification step from its beginning to obtaining a milky white emulsion (stage A) was 3.5 h and then to the end of the emulsification step (stage B) it was 2.5 h.

Microencapsulation (polymerization) step

The stirring speed of the reaction mixture was reduced to 750 rpm for about 20 min., after which a solution of citric acid was added to the emulsion (to achieve pH 3), at a temperature of 45°C [11, 13, 14]. The reaction mixture was stirred for 3 h under the same conditions, after which the resulting microcapsules were filtered, washed with distilled water and dried at room temperature or in a dryer at a temperature of 55 - 60°C for 6 h. To harden the microcapsules, after the expiration of the time, the solution was cooled to room temperature (or previously obtained capsules were placed in water), with constant stirring, 2 mL of a 37 % alcoholic solution of formalin was added, and the stirring was continued for another 15 min until their solidification. Then, the obtained microcapsules were filtered, washed with distilled water and dried under the conditions mentioned above.

Product analysis

Weight analysis

The encapsulation efficiency (EE %), microcapsule yield (%) and core content (E % core) were calculated using the presented equations from previous works [16]. Using equations (1) and (2) from another article, the encapsulation factor and the resin efficiency (%) are calculated [17].

Particle size analysis

150 measurements were made to determine the mean microcapsule diameter, capsule size distribution and standard deviation. The mean diameter of the microcapsules was calculated as the arithmetic mean of the particle size measured with a laser diffraction apparatus.

FT-IR spectroscopic analysis

The spectra of the prepared microcapsules are obtained after freeze drying of the whole microcapsules and the capsule shell is analysed using the FT-IR spectrometer. The infrared spectra are at 3500 cm^{-1} and 3300 cm^{-1}, 2750 cm^{-1} and 2600 cm^{-1}, 1600 cm^{-1} and 1480 cm^{-1}, 1140 cm^{-1} and 1050 cm^{-1}, corresponding to C-H, N-H, C-N and C=O vibrations. N-H of the amine is at 3350 cm^{-1} and 3230 cm^{-1} [18, 19]. The indicated absorption maxima of the spectrum are of the urea - formaldehyde polymer, which makes up the capsule wall.

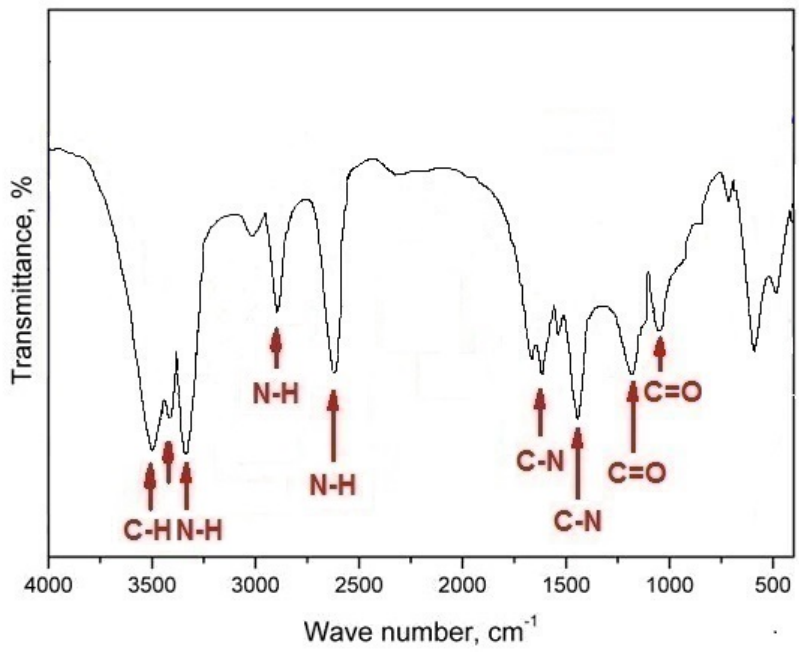

Fig. 1. FT-IR spectrum of poly(urea - formaldehyde) shell of the microcapsules filled with rose oil.

Table 1. Effect of temperature in the first sub - step (stage A) of the emulsification step on the characteristics of the obtained rose oil microcapsules.

№	Temperature, °C	Yield, %	EE, %	$E\%_{core}$	RE,%	EF	Size, µm
1	40	33.1	49.8	41.6	56.4	0.53	70 - 50
2	50	39.1	56.8	35.2	54.2	0.73	50 - 30
3	60	49.2	63.6	42.2	55.7	1.04	30 - 25
4	70	63.5	82.1	38.4	56.6	1.85	25 - 15

RESULTS AND DISCUSSION

Effect of the temperature in the first sub - step (stage A) of the emulsification step on the characteristics of the obtained microcapsules

Further research shows that temperature has a huge impact on the process, as it affects every step of microencapsulation, especially the emulsification step and the polymerization step. As mentioned above, the author divides the emulsification step into two sub - steps (stages: A and B): A. Formation of microdroplets; B. Pre - polymer molecular adsorption on the microdroplet surface.

Increasing the temperature during the microdroplet formation stage leads to a decrease in their size, which subsequently affects the size of the resulting microcapsules. For example, when encapsulating rose oil, if the temperature at the stage of microdroplet formation is 40°C, the size of the obtained microcapsules varies from 70 - 50 µm, while when the reaction mixture is heated to 70°C, at this stage the microdroplets acquire a size between 25 - 15 µm, which also affects the final diameter of the microcapsules (Table 1, Fig. 3).

Similarly, the increase in temperature has a favourable effect on other characteristics of the obtained microcapsules, such as capsule yield (%), which increases from 33.1 % at 40°C to 63.5 % at 70°C (Table 1, Fig. 2). The encapsulation efficiency (yield of the encapsulated substance, %) increased from 49.8 % (40°C) to 82.1 % (70°C).

Regarding the content of the encapsulated substance (E% core), with increasing temperature it

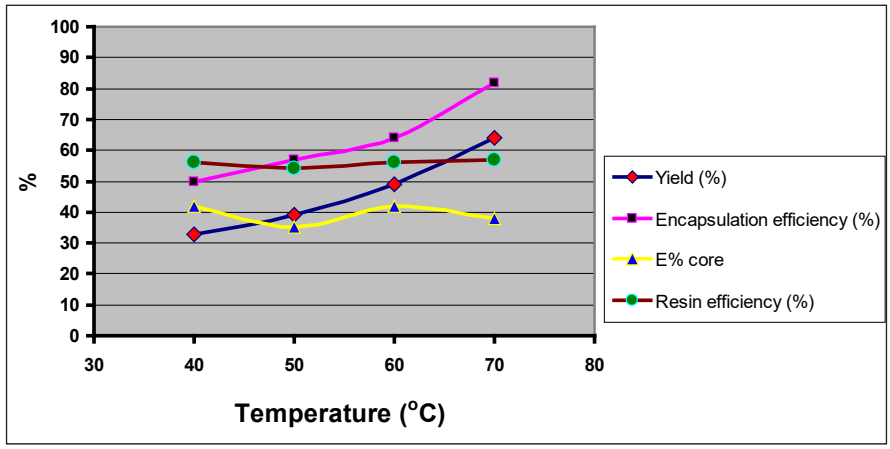

Fig. 2. Effect of temperature in the first sub - step (stage A) of the emulsification step on the characteristics of the obtained rose oil microcapsules.

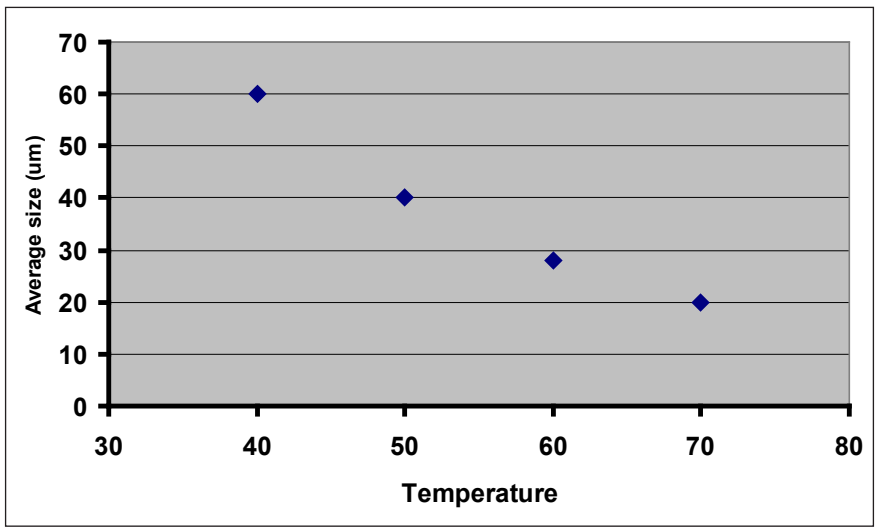

Fig. 3. Effect of temperature in the first sub - step (stage A) of the emulsification step on the size of the obtained microcapsules of rose oil, represented by the average value of the diameter, in µm.

slightly decreases (from 41.6 % at 40°C to 38.4 % at 70°C) (Table 1, Fig. 2), but the temperature at this sub - step does not directly effect of this characteristic, as this sub - step of the emulsification step is influenced by another factor (stirring speed), which was discussed previously. When encapsulating the rose essential oil, the E% core hardly changes, i.e. remains relatively constant.

Other conditions: stirring speed 1500 rpm; time 3.5 h; surfactant concentration (SDS) 3 %.

Effect of the temperature in the second sub - step (stage B) of the emulsification step on the characteristics of the obtained microcapsules

The temperature in the second sub - step (stage B) of the emulsification step has a key influence on one of the characteristics of the obtained microcapsules, which is the content of the encapsulated substance (E% core). The reduction in the percentage of encapsulated substance gives an indication of the quality of the capsule shell, and particularly its density. Since E%

core is a ratio between the mass of the encapsulated substance to the mass of the capsule, this means that the decrease in the value of this ratio gives information about the quality of the resulting shell, i.e. changing these parameters it becomes denser.

Good information in this regard gives us another characteristic, based on the ratio between the weight of resin in product (insoluble shell, microcapsule wall) and the initial weight of resin in solution in solution multiplied by 100, called resin efficiency [17, 20]. This characteristic is inversely proportional to E% core, as the increase in the value of the content of the encapsulated substance (E% core) leads to a decrease of the resin efficiency and vice versa.

The temperature increases in the second sub - step (stage B) of the emulsification step leads to an increase in the content of the encapsulated substance (E% core) (Table 2, Fig. 4), respectively, to a decrease in the wall quality by the characteristic resin efficiency (Table 2, Fig. 4). This is due to the fact that an increase in temperature leads to an increase in the rate of desorption of pre - polymer particles from the surface of the formed microdroplets, respectively to thinning and a decrease in density, as well as in general to the quality of the polymer shell obtained after polymerization of the reduced number of pre - polymer mono methylol urea particles.

As mentioned above, increasing temperature leads to an increase in the desorption rate (v_{-1}) by affecting the rate constant of the process (k_{-1}), whereby it increases while Ks decreases. Accordingly, the equilibrium shifts to the left until new values are reached. At the same time, increasing the temperature in the second sub - step (stage B) of the emulsification step has no effect on the size of the resulting microcapsules (Table 2, Fig. 5). During stage B of this step, after the formation of microdroplets, adsorption of the pre - polymer particles take place.

Therefore, temperature does not affect the size of the microdroplets at this stage and the size of the finally obtained microcapsules.

The rise in temperature has a negative effect on

Table 2. Effect of temperature in the second sub - step (stage B) of the emulsification step on the characteristics of the obtained rose oil microcapsules.

№	Temperature, °C	Yield, %	EE, %	$E\%_{core}$	RE, %	EF	Size, µm
1	40	62.6	77.8	38.1	57.3	1.83	25 - 15
2	50	59.1	62.8	42.3	57.7	1.78	30 - 20
3	60	49.2	45.6	68.9	44.1	1.05	25 - 20
4	70	37.8	29.1	79.9	35.4	0.68	30 - 15

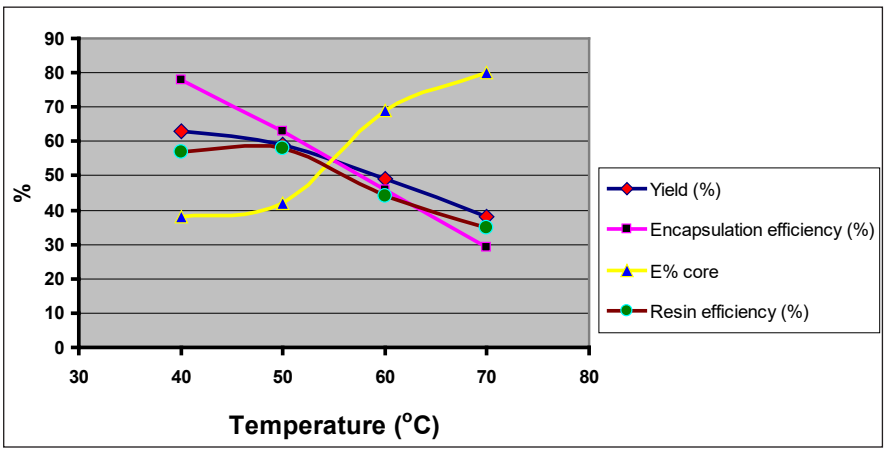

Fig. 4. Effect of temperature in the second sub - step (stage B) of the emulsification step on the characteristics of the obtained rose oil microcapsules.

the remaining two characteristics: capsule yield (%) and encapsulation efficiency (%), which decrease (Table 2, Fig. 4). Therefore, based on the results presented below, the most suitable temperature for the second sub - step (stage B) of the emulsification step ranges between 40°C and 50°C, i.e. it should be lower in order to prevent the undesired process of desorption of mono methylol urea molecules from the surface of the microdroplets obtained during the first sub - step (stage A) of the emulsification step. For this reason, the consideration and study of the influence of the other factors was carried out under the conditions maintaining a temperature of 45°C for the second sub - step (stage B) of the emulsification step.

Due to the fact that in parallel with the formation of the microcapsules, a side process of formation of microparticles made of the corresponding polymer also takes place, the influence of the factors on another parameter characterizing the efficiency of the microencapsulation process, namely the ratio between the weight of the capsules and the weight of the microparticles, is considered, which is directly related to the microcapsule yield, called the encapsulation factor [17].

The formation of polymer microparticles is probably, because during the microencapsulation process at a higher temperature, a large part of the pre - polymer particles desorbed from the surface of the microdroplets during the second sub - step (stage B) of the emulsification (II) step and during the polymerization (III) step of formation of the microcapsule shell. The lighter microcapsules stay on the surface due to their lower density due to being filled with essential oil, while the denser microparticles fall to the bottom.

Since that at a lower temperature agglomeration of the microdroplets is induced, although this is partially compensated by the high speed of the process during the emulsification step, a slightly higher temperature is required during the second sub - step (stage B) of the emulsification step, for example 50°C or between 40°C and 50°C (45°C) which is sufficient to not cause agglomeration.

Other conditions: stirring speed - 1500 rpm; stirring time - 2.5 h; surfactant concentration (SDS) - 3 %.

As seen from the results presented above, the temperature during the emulsification step has a great influence on the efficiency of the microencapsulation process of rose oil by *in situ* polymerization, as well as the quality of the obtained microcapsules. However, this influence is multidirectional and must be considered.

In the first stage of the emulsification step (stage A), the increase in temperature from 40°C to 70°C leads to an improvement in the efficiency of the process and the quality of the obtained microcapsules, represented by the corresponding values almost of all characteristics. However, the increase in temperature does not reflect the quality of the capsule wall, represented by the

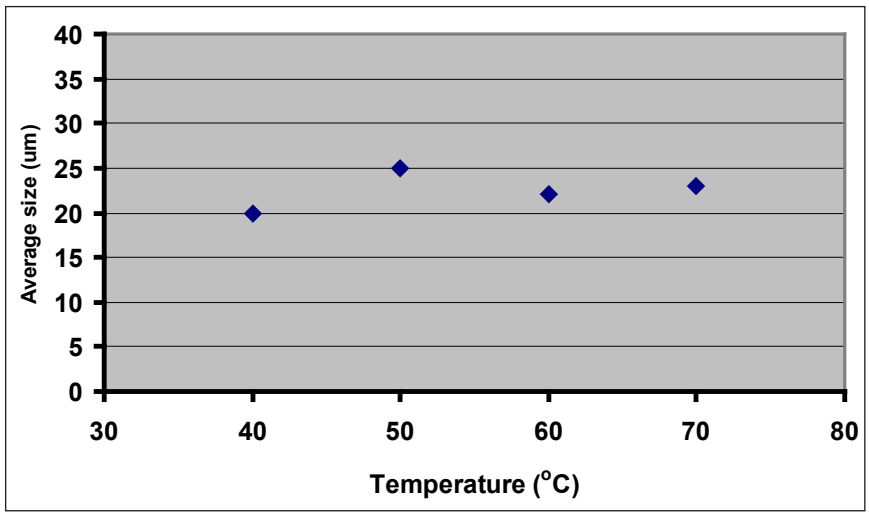

Fig. 5. Effect of temperature in the second sub - step (stage B) of the emulsification step on the size of the obtained microcapsules of rose oil, represented by the average value of the diameter, in μm.

content of the encapsulated substance (E% core) and the resin efficiency (RE, %), which remain constant.

As the temperature increases during the second stage of the emulsification step (Stage B), however, the content of the encapsulated substance (E% core) and the resin efficiency (RE, %) change. In the second stage of the emulsification step, the increase in temperature to certain values (from 40°C to 50°C) also leads to an increase in the efficiency of the process, the yield and the quality of the obtained microcapsules, which is mainly due to the favourable temperature sufficient to maintain the size of the microdroplets obtained in the first stage, while preventing their agglomeration, as well as for the adsorption of the prepolymer particles on the surface of the microdroplets, which favourably affects the quality of the capsule wall, and hence the yield and quality of the obtained microcapsules. However, a further increase in temperature did not lead to good results, probably because high temperatures of the order of 60°C and 70°C cause desorption of the pre-polymer molecules from the surface of the microdroplets. This also reflects the characteristics such as yield (%), encapsulation efficiency (EE, %), resin efficiency (RE, %) and encapsulation factor (EF), which decrease and the core content (E% core) increases. Lowest values of E% core are observed at a temperature between 40°C and 50°C, and with increasing temperature (60°C and 70°C), the content of the encapsulated substance grows, whereas at a temperature between 40°C and 50°C yield (%), encapsulation efficiency (EE, %), resin efficiency (RE, %) and encapsulation factor (EF) have highest values and at a temperature of 70°C these characteristics acquire lowest values. On the other hand, the size of the microcapsules remains constant, which means that increasing the temperature above 60°C does not affect this characteristic, and values of the order of 40°C and 50°C are quite sufficient to protect the microdroplets from their agglomeration and gluing.

The second step (emulsification step) is probably the most important because it is then that microdroplets are formed, on whose surface the pre - polymer is adsorbed. In the first part of the step, a higher temperature is needed to obtain finer microdroplets. The second part of the step requires a lower temperature to prevent desorption of the pre - polymer particles from the surface of the microdroplets. Therefore, it is of particular importance that this step depends on time, stirring speed and temperature. The effect of time was studied in the previous work [17]. A study on the effect of stirring speed in the emulsification step is forthcoming, which will be discussed in another article.

Of course, there are more precise studies to be done regarding the influence of temperature, both during the two sub - steps (stages A and B) of the emulsification step and during the polymerization (microencapsulation) step.

CONCLUSIONS

The research performed and presented in this work examines the influence of temperature during the most important step of microencapsulation by *in situ* polymerization i.e. emulsification step. The results show that the influence of this factor is undeniable, but divergent during the two sub - steps (stage A and stage B) of the emulsification step. Thus, by studying the influence of temperature, along with stirring speed and time [17], discussed in other works, the author hopes that this will contribute to optimizing the conditions to improve the efficiency of the process and obtain better quality microcapsules.

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Authors' contributions: *All experimental results, including pre-polymer synthesis, microcapsule preparation, weight analysis, microscopic analysis, particle size analysis, and IR analysis, were performed by the author.*

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