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Synthesis, characterization and controlled release studies of ethyl cellulose microcapsules incorporating essential oil using an emulsion solvent evaporation method

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Abstract
A promising technique has been developed to encapsulate clove oil in ethyl cellulose (EC) microcapsules using an emulsion solvent evaporation method. The method consists of emulsifying the oil with variations of core (clove oil) loading (1.5, 3.0 and 4.5) ml into EC shell material crosslinked by epichlorohydrin (ECH). Spherical capsules with a smooth and porous surface were produced and it was noted that by studies of variations of oil loading affects the stability of microcapsules. This is being demonstrated by studies of SEM images and loss on drying method. Release study of synthesized microcapsules was achieved by UV and loss on drying method. Stable and appropriate oil loading formulation was estimated by SEM images and % release of core material.
We prepared three formulations with different loading of core F1 (1.5 ml), F2 (3.0 ml) and F3 (4.5 ml) of which studies revealed that F2 formulation having 3 ml of oil loading for 1 g EC shell was appropriate for microcapsules stability and controlled release application. Furthermore, the stable F2 formulation was studied with variations of ECH loading. A (3.0), B (4.5) and C (6.0) in ml for controlled release study and concluded by U.V absorbance and kinetic studies that formulation B was appropriate for slow release of core. Formulation B was further characterized by FTIR, TGA and particle size analyzer. The controlled releases were monitored by measuring spectral absorption in the UV range and loss on drying method.

Keywords: Controlled release, solvent evaporation, microencapsulation, clove oil, ethyl cellulose (EC).

1. Introduction
Microcapsules are spherical or of irregular shape consisting of tiny particles containing one or more active ingredients (core) surrounded or coated by polymeric shell (synthetic or natural) material which protects or gives sustained release of core materials. In general, the size of polymeric microcapsules ranges from 1 to 100 μm [1-4]. Microencapsulation provides the means of converting liquids to solids, of altering colloidal and surface properties, of providing environmental protection and of controlling the release characteristic or availability of coated or encapsulated materials [5]. Microencapsulation technology is useful for enhancing the controlled release, beneficial effects, handling and formulation stability of drugs, agrochemicals and other biologically active agents [6].
The synthesis of microcapsules can be carried out by microencapsulation method, including physical and chemical techniques. The physical techniques are spray drying, fluidized bed, centrifugation etc. Which yield relatively large and rough surface capsules in comparison with the chemical techniques, i.e., solvent evaporation, *in-situ* polymerization, interfacial polycondensation, coacervation, etc., which yield a small and smooth surface capsules [7-13].
Microencapsulation is one of the most widely used formulation techniques to elaborate controlled release applications in several fields such as textiles[1], cosmetics [4], pharmaceuticals [13-15], essential oils [12, 16, 17], pesticides [18-20], self-healing coatings [21], biomacromolecules [22], adhesives [23], fragrance [24], flavors [25], dyes [26] and also used for taste-masking of some bitter taste of drugs [27].
Nowadays, the use of synthetic chemicals including insecticides, herbicides, nematicides etc. to control insects, herbs, nematodes, respectively, but these impose problems not only on environment but also on human health. Clove and Citronella oil possess good efficacy and are environment friendly repellents which are extracted from *Lauraceae, Myrtaceae, Lamiaceae* and *Asteraceae* plants. These extracts have the potential to provide efficient repellent action

against malarial, filarial and yellow fever vectors and found to be safer for human and environment [28]. Most of these essential oils are highly volatile, light sensitive and sparingly water soluble making them unsuitable for usual applications. These constraints can be addressed by making proper formulation to improve their longevity or shelf life [29].

In recent years, the interest in natural extracts has increased as an alternative for the control of pathogenic microorganisms. Clove oil used in footwear for an anti-germicidal activity to control wound and feet infections caused by dermatophytes [30]. Clove oil is an essential oil extracted from the dried flower buds, leaves and stem of the tree *Eugenia caryophyllata* and *Eugenia aromaticum*, which exhibits good acaricidal activity than benzyl benzoate and for N,N-Diethyl-m-toluamide (DEET) against, *Dermatophagoides farinae* and *Dermatophagoides pteronyssinus* species of mites or acaries [31]. Park and Shine reported that the main compounds of clove bud oils eugenol (86.1%) and α-caryophyllene (11.1%) has anti-termitic activity mainly attributed to the action of eugenol [32]. Eugenol is a major constituent of clove oil possessing wide range pharmacological effects including anti-inflammatory, antioxidant, antifungal, cardiovascular, analgesic, local anesthetic, antimicrobial, antitumor and hair-growing effects [33-34].

Both synthetic and natural polymeric materials are being used for as a wall material. Natural materials such as chitosan, cellulose, starch, gelatin, alginate, and poly (l-lactide) have biodegradability and environment-friendly properties and utilized for the sustained-release system of unstable drugs or specific ingredients in the food [35-37]. Synthetic polymers include phenol, melamine, urea, urethanes and so on. Synthetic materials are toxic to human being as well as to environment; hence it is not recommended for use in food products [38].

The excess release of essential oil causes unpleasant smell; hence control release is expected to overcome the problem. Sustained release is a one mode of controlled release system. It maintains constant concentration of active compounds at the targets [39]. In initial stage, the amount of released oil is large and then slowly the decay becomes constant. The release rate of encapsulated volatile essential oils mainly depends on component materials of microcapsules (core material, emulsifier and coating material) and various environmental factors such as temperature, pH and humidity [35].

Ethyl cellulose is a non-biodegradable and biocompatible polymer, which is one of the extensively studied encapsulating materials for the controlled release. The literature survey revealed that use of Ethyl Cellulose (EC) shell for encapsulation of cores such as drugs 5-flurouracil [13], aceclofenac [14], diclofenac sodium [15], oils such as jojoba [16], rosemary [17], pesticides such as 2,4-D(2,4-Dichlorophenoxy) acetic acid [18], Alachlor and Metolachlor [19], Atrazine and Metribuzin [20] but has not yet been explored for encapsulation of clove oil as a core. On the contrary, clove oil has been used as a core which encapsulates by polymeric shell walls gelatin with sodium carboxy methyl guar gum by complex coacervation method [40].

2. Experimental

2.1 Materials

Ethyl cellulose (EC) was purchased from Aldrich, Austria (viscosity 22cP 5% in toluene/ethanol 80:20 (lit.), extent of labeling: 48% ethoxyl) made of USA. Clove oil was purchased from Diamond Biotech, Indore (M.P.), Epichlorohydrin was procured from Lobal Chemie, India, Ethyl acetate was purchased from Merck and Sodium dodecyl sulfate was obtained from Sigma-Aldrich (Germany)which was used as an anionic emulsifier. All other chemicals used for analysis were of analytical grade.

2.2 Synthesis of Microcapsules:

Clove oil microcapsules were prepared by an emulsion solvent evaporation technique, employing ethyl acetate as solvent and ethyl cellulose as a polymer with a variation of oil loading.

Organic Phase: Ethyl cellulose (1 g) was dissolved in ethyl acetate (15 ml), under continuous stirring at 5000 rpm. (1.5, 3.0 and 4.5) ml of clove oil was added to this solution. The stirring was maintained for 2 min.

Aqueous Phase: Sodium dodecyl sulfate (500 mg) was dissolved in water (50 ml). 5 ml of ethyl acetate was added to this aqueous solution under a continuous stirring (1500 rpm). Next, ***Organic phase*** was added slowly into ***Aqueous phase*** under continuous stirring (5000 rpm) for 3 min. The microcapsules emulsion thus formed was transferred into 250 ml 3 neck round bottom flask and adjusted for pH=10 by adding appropriate amount of 2 M NaOH solution. Subsequently, the product got cross linked by slow addition of (3.0, 4.5, 6.0) ml of ECH. The temperature of the system was then raised to 45-50°C and stirring was continued for another 2–3 h to complete the crosslinking reaction and the solvent was evaporated. The microcapsules formed (Fig 2) were filtered using Whatman No 41 filter paper washed 2-3 times with water and finally dried at room temperature.

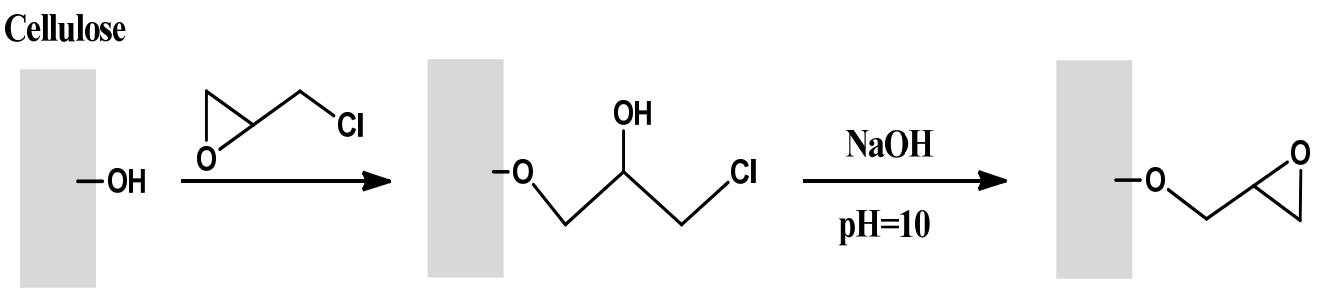

Fig 1: Possible Mechanism of EC cross-linking with ECH.

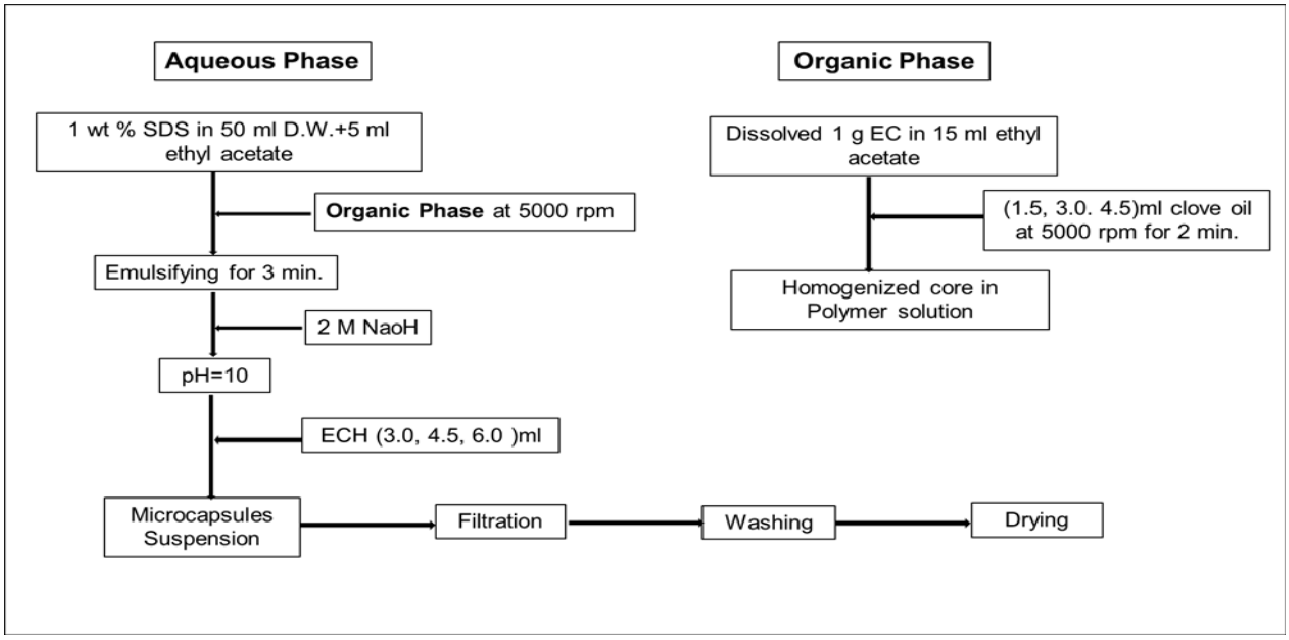

Fig 2. Flow chart for the preparation of the EC microcapsules containing clove oil.

2.3 Characterization

***2.3.1. Fourier transform infrared (FTIR) spectroscopy*:**
The FTIR spectral scanning were carried out at ambient temperature on Perkin Elmer (spectrum one) spectrophotometer using nujol mull technique.

***2.3.2. Field Emission Scanning Electron Microscope (FESEM)*:**
Prepared microcapsules were observed for their morphological study under scanning electron microscopy using thin layer of samples coated with gold on a FESEM (FESEM, HITACHI High Technologies Corporation, Japan, Model No. S-4800-II) with an applied voltage 5000 Volt and the emission current is 9400nA.

***2.3.3. Thermo gravimetric analysis* (*TGA*):**
Thermal decomposition or stability for the prepared microcapsules and core (clove oil) were studied by the thermo gravimetric analysis (Model - TGA 4000, PerkinElmer, USA) in the range of 37–700 °C temperature at heating rate of 20 °C /min in presence of nitrogen gas as an inert atmosphere.

2.3.4. Particle size analyzer:
Particle size of prepared microcapsules was determined by using laser particle size analyzer (Mastersizer 3000, Malvern, UK).

2.3.5. *UV-VIS Spectrophotometer:*
Core release study was carried out using double beam UV-VIS Spectrophotometer (Chemito, Spectrascan UV 2700).

3. Results and Discussion:

3.1 The effect of variations in oil loading:
The data of observed effect of variations in the concentration of clove oil on oil load and % release rate are shown in Table 1 and Figure 3. The examination of results reveal that with an increase in the loading of the clove oil used in the microencapsulation process, % release gets decreased at 4.5 ml (F3) loading due to ruptured of capsules, so not encapsulated high loading, 12% release was observed because of some oil retained internally or externally part of shell material. A possible reason for the decrease % release may be due to increase in oil loss during encapsulation of the oil resulting from the high oil loading used and the amount of shell not have been sufficient for encapsulation of all oil droplets [12].

Table 1: Effect of variation of oil loading on the behavior of release of microcapsules

	F1		F2		F3	
Time (hrs)	Core released (g)	Released (%)	Core released (g)	Released (%)	Corer released (g)	Released (%)
1	0.005	4.16	0.010	8.33	0.008	6.66
2	0.013	10.83	0.023	19.16	0.012	0.012
3	0.016	13.33	0.030	24.16	0.014	11.66
4	0.017	14.16	0.031	25.83	0.0142	11.83
5	0.0173	14.41	0.032	26.66	0.0142	11.83
6	0.0174	14.50	0.0325	27.08	0.0142	11.83
7	0.0175	14.58	0.0328	27.33	0.0142	11.83
8	0.0175	14.58	0.0328	27.33	0.0142	11.83

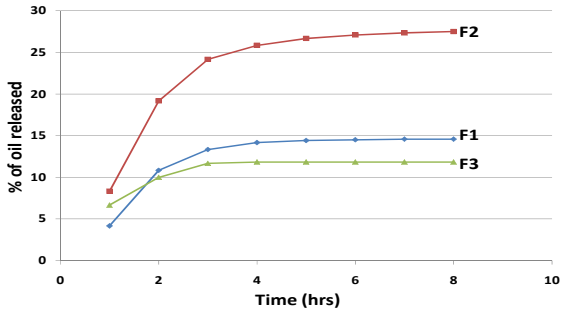

Fig 3: The effects of variation of oil loading on the release rates (**F1**) EC 1 g, ECH 3.0 ml, Clove oil 1.5 ml; (**F2**) EC 1 g, ECH 3.0 ml, Clove oil 3.0 ml; (**F3**) EC 1 g, ECH 3.0 ml, Clove oil 4.5 ml.

The increase in the oil release of F2(27%) than F1(16%)may be the result of a decrease in the wall thickness of the microcapsules caused by the amount of the EC used as a wall material which was being kept constant while the oil concentration was increased(F2) or decreased (F1) are shown in Table 1 and Figure 3.

3.2 The effect of variations in cross linker concentration:

The results related to variations in ECH concentration are given in Table 2 and % releases with respect to time is shown in Figure 4. It is clear that as the amount of cross linker increases, the amount of core released decreases. An investigation of the values obtained reveals that, as the ECH concentration increased, the oil content remains same. In addition, it is observed that A having releases all 27% oil within a 8 Hrs but an increase in the ECH concentration (B and C) lead to a constant % release and time extended up to 10 Hrs to achieve all 27% release (Table 2). Maji *et al.* have pointed out that the increase in the oil holding capacity of the microcapsules may be due to the reaction of the wall substance to the cross linker [12].

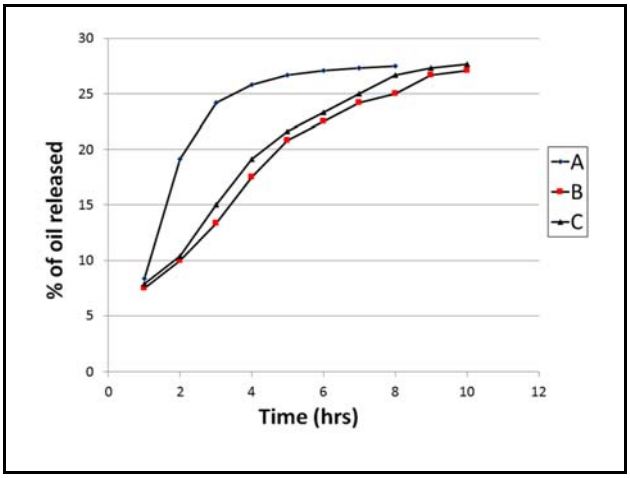

Fig 4: The effects of variation of ECH on the release rates (**A**) EC 1 g, ECH 3.0 ml, Clove oil 3.0 ml; (**B**) EC 1 g, ECH 4.5 ml, Clove oil 3.0 ml; (**C**) EC 1 g, ECH 6.0 ml, Clove oil 3.0 ml.

Table 2: Effect of variation of ECH on the behavior of release of microcapsules

	A		B		C	
Time (hrs)	**Core released (g)**	**Released (%)**	**Core released (g)**	**Released (%)**	**Corer released (g)**	**Released (%)**
1	0.010	8.33	0.009	7.5	0.0095	7.91
2	0.023	19.16	0.012	10.00	0.0125	10.41
3	0.030	24.16	0.016	13.33	0.018	15.00
4	0.031	25.83	0.021	17.50	0.023	19.16
5	0.032	26.66	0.025	20.83	0.026	21.66
6	0.0325	27.08	0.027	22.50	0.028	23.33
7	0.0328	27.33	0.029	24.16	0.030	25.00
8	0.0328	27.33	0.030	25.00	0.032	26.66
9	--	--	0.032	26.66	0.0328	27.33
10	--	--	0.0325	27.08	0.0332	27.66

It is observed that the release of the Clove oil from the microcapsules shows sustainably as the ECH concentration is increasedfrom3.0 to 4.5 ml this is not noticeable for to changes in concentration between 4.5-6.0 ml (Figure 4). We have thus established that for 1 g EC shell material 3.0 ml oil loading with a 4.5 ml ECH as a crosslinking agent is suitable for stable and sustainable release of clove oil.

3.3 FTIR Analysis

FTIR spectra's of the extracted core material (2A), standard clove oil (2B), ethyl cellulose microcapsules (2C) and extracted shell (2D) are shown in Figure 5. The observed absorption band at 1462 cm^{-1} corresponds to the C=C aromatic ring stretching frequencies. The band at 3450 cm^{-1} represent the characteristics of the O-H stretching present in core moiety respectively, while the absorption band (2C) noted at 1273 cm^{-1} in the ethyl cellulose is due to the presence of C–O stretching. The band at 1748 cm^{-1} corresponds to the C=O stretching frequency of eugenyl acetate (present in clove oil about 10%) and 1601cm^{-1}correspondsto the C=C stretching.

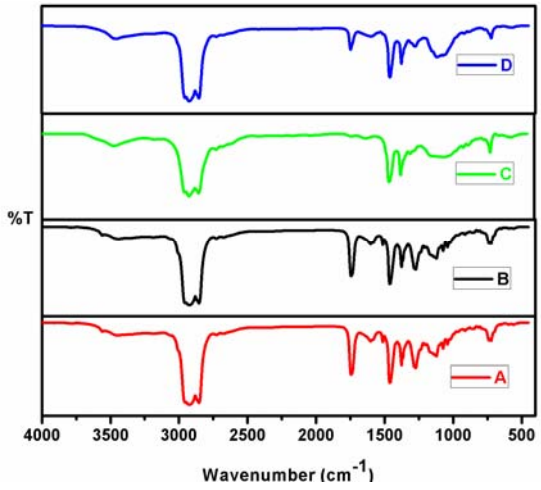

Fig 5: FT-IR spectra of (A) Extracted clove oil, (B) Standard clove oil, (C) EC microcapsules and (D) Extracted shell

Also, for the extracted shell and core molecule all frequencies and all functional groups are present in EC microcapsules. The same frequencies for different functional groups present in core and in EC microcapsules confirmed the formation of EC shell and core moiety present in the microcapsules.

3.4 Surface Morphology of Microcapsules:

Surface morphology of synthesized microcapsules was studied by an optical microscopy in laboratory followed by scanning electron microscopy study as summarized below.

3.4.1 Optical Microscope

The surface morphology of EC microcapsules containing different loading of clove oil, prepared under the high speed agitation was studied using an optical microscope (Labomed STC - ML) at 40X resolution for primary conformation of shell formation (Figure 6). Optical pictures clearly show formation of spherical microcapsules. We note from Fig 6(F3) that there is an agglomeration of microcapsules because of the excessive amounts of surface oil which promoted significant agglomeration causing reduction in encapsulation efficiency. However, the results of Fig 6 (F2) shows that appropriate loading of oil reduces the agglomeration and formation of uniform spherical capsules.

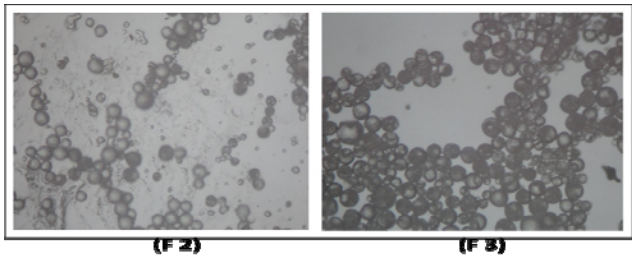

Fig 6: Optical microscopic images (F2and F3) of the EC microcapsules containing clove oil.

3.4.2 Scanning Electron Microscope (SEM)

Scanning electron microscope imaging was used to determine the microcapsules shape and morphology. The SEM photomicrographs of clove oil with variation in loaded microcapsules (Fig 7) show spherical microparticles with a smooth and porous surface. It is well known that the microcapsule size is an important parameter that affects the stability and the release of the active ingredient from microparticles [16]. Therefore the smooth surface morphology of the microcapsules has proved to be useful with regard to the protection and sustained release of interior core material.

The SEM picture in Fig 7 (F1) clearly shows very small particles which were found to adhere to large particles and irregular shape was observed for low loading of core. In Fig 7 (F3) capsules were ruptured and opening of microcapsules was observed for high loading of core. This may happen because of volatile nature of the core material that not allowed high loading into EC shell material. Furthermore, the particles appear to be in the form of agglomerated microcapsules. Such a behavior was also observed by Tan *et al.* in the microencapsulation of marine oil with alginate/starch blends, where microspheres with excessive amounts of oil loading promoted significant agglomeration reducing microencapsulation efficiency [41]. Fig 7 (F2) clearly shows stable and uniform size microcapsules with not an agglomeration or rupture.

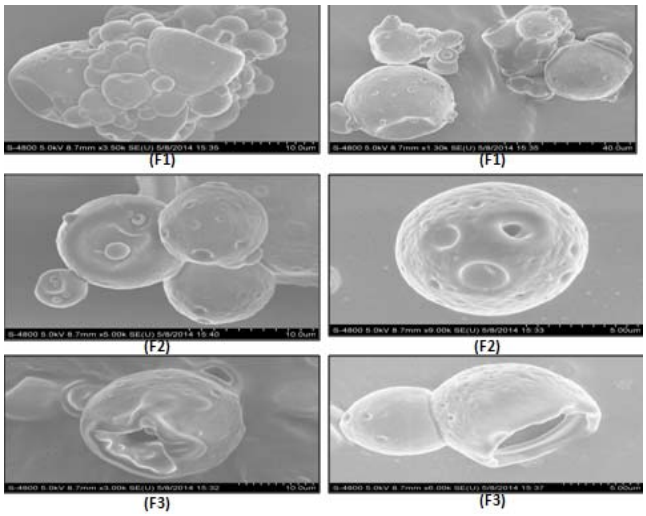

Fig 7: The effects of variation of oil loading on the SEM images (F1) EC 1 g, ECH 3.0 ml, Clove oil 1.5 ml; (F2) EC 1 g, ECH 3.0 ml, Clove oil 3.0 ml; (F3) EC 1 g, ECH 3.0 ml, Clove oil 4.5 ml.

SEM results reveal that prepared microcapsules with different loading of oil exhibit different images and stability. We decided that F2 formulation with 3 ml oil loading into 1 g EC shell material is possible and stable. Formulation F1 and F3 are not suitable for low loading and high loading of oil. Using these results (shown as SEM images) formulation F2 was adopted for variations of ECH concentration in the formulations A, B and C for their release studies.

3.5 Thermo gravimetric Analysis

The thermal properties and stability of extracted ethyl cellulose shell and core (clove oil) were investigated using thermo gravimetric analysis (TGA). The thermo grams of EC shell and clove oil are shown in Figure 8. The TGA curve (a) shows that the onset weight loss of core gets started at 80-90ºC and then continues up to 190 ºC in a single stage but TGA curve (b) of EC shell undergoes the high weight loss (90%) between 145 °C and 390 °C. The TGA curve shows a shoulder in the 180 °C to 300 °C region and an intense peak centered at 360 °C, revealing involvement of two stages of the thermal degradation reaction of ethyl cellulose shell material.

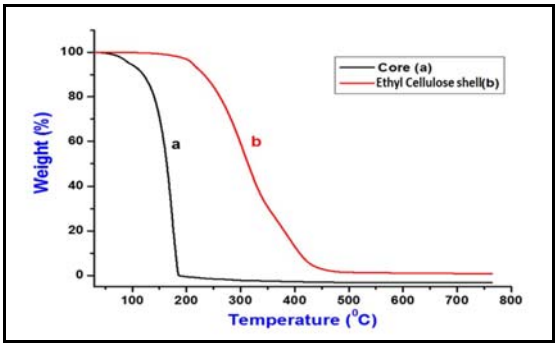

Fig 8: TGA curve of (a) core and (b) extracted shell of EC.

Perusal of one results indicate that the EC shell material degrades slowly, meaning that the shell have higher thermal stability than core. Therefore core clove oil could be better preserved in the EC shell material for microencapsulation purpose. It should also be helpful to reduce the degradation of core material for its longer and efficient utilization.

3.6 Particle Size Analysis

The particle size distribution is an important parameter for the development of formulating microcapsules. The small size of microcapsules is preferred so as to have a large contact area which adheres fast to the surface of target. In addition, the microcapsules should have a long stability in releasing the active material.

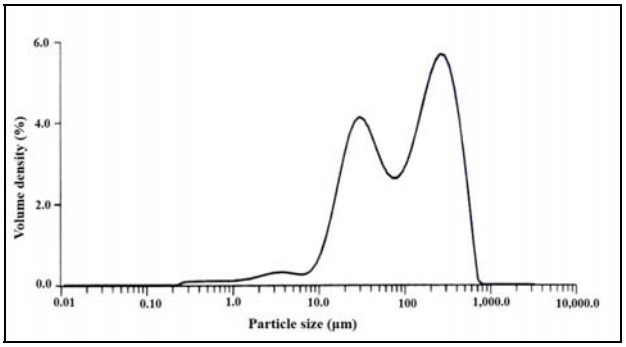

Fig 9: Particle size distribution of the EC microcapsules containing clove oil

"The particle size distribution histogram of the synthesized EC microcapsules containing of clove oil as a core" is shown in Fig 9. The microcapsules size is estimated to be in the range of 10–520 µm and having the mean particle size distribution of 110 µm (1% sodium dodecyl sulfate was used as a surfactant in the formulation to facilitate particle size reduction).

4. Controlled release study

The % release rate of core material was calculated by loss on drying following the method reported earlier [2] and making measurements of UV absorbance as a function of time.

4.1 For Oil loading

Controlled release behavior or characteristic property of EC microcapsules containing different loading clove oil formulations (F1, F2 and F3) were investigated with loss on the drying method. The data are collected in Table **1** and exhibited in Figure 3. Formulation F2 shows sustainable release of about 27% in 8 hrs. Which is better than F1 and F3. Order/Kinetics of core release of sustainable F2 formulation was studied using the data of optical absorbance measurements at 282 nm obtained from UV Spectral results as shown in Fig 10a and Fig 11a.

For this study 0.12 g of microcapsules were transferred to a dry Gooch crucible, 10mL of xylene was added and stirred gently at room temperature and allowed to extract the core material for about 1h.After 1 h, the eluent was collected and the absorbance was recorded with a UV spectrophotometer. Similarly 8 such samples were collected up till 8 Hrs. The data are given in Fig 10a.

4.1.1 *Loss on the drying method*

The remaining microcapsules' residue in the Gooch crucible was kept in an oven for drying at 50 °C (+-5) and then the dried sample was weighed. This process was repeated with an interval of 1 h until all core material was extracted (in about 8 h), to determine the loss of weight after drying for a quantitative release study. The experimental data of the controlled release study and its graphical presentation are given in Table 1 and Figure 3, respectively.

Formulation F2 having a 3 ml oil loading exhibits about 27% release in 8 hrs. But formulation F3 having a 4.5 ml of loading causes about 12% of release. This is supported on the basis of SEM images of F3 which shows opening and rupture of capsules may probably be due to the volatile nature of constituents in the core. 12% release was noted as some content of oil remain inside the capsules or absorbed into the shell. Formulation F1 exhibits about 16% release up to the 8 hrs. However, for this no uniform spherical capsules were seen.

4.2 For ECH loading (cross linker)

Similar experiment was performed for ECH loading.

As we noted earlier that 3 ml oil loading is suitable for 1 g. EC shell for, SEM images and % release data. The ECH concentration in formulation was varied and the results revealed that 4.5 ml (B) of ECH in formulation help to retard the release of clove oil encapsulated in EC shell for about 10 hrs. This statement is based on UV spectral results shown in Fig 10a, 10b and 10crespectively.Order of releases is evaluated by kinetic study as shown in Fig 11a, 11b and 11c respectively.

3.0 ml (A) loading retarded oil just up to 8 hrs. Not remarkable effect was observed on 6.0 ml loading(C) which also retarded release up to 10 hrs. The results collected in Table 2, Figure 4 and Figure 12 respectively show similar trend for 4.5 ml (B) loading.

Weight loss on drying study

The total amount of released core material from microcapsules was determined by extraction method, as described above in Table 1 and Table 2. The amount of core material released was calculated with the following equation [2].

$$\%\text{Core release} = \frac{Wa\text{-}Wb}{Wa} \times 100$$

Where

Wa, weight of microcapsules before extraction and

Wb, weight of microcapsules after extraction

4.3 UV Absorbance

The above collected eluents from microcapsules formulation A, B and C were subjected to analysis of absorbance obtained to confirm the release of clove oil. Clove oil contains about 87% of eugenol and 8.1% eugenyl acetate. UV spectrum of eugenol absorption exhibits absorbance peak at 282 nm and 226 nm for eugenyl acetate in clove oil which are in agreement with literature data [42]. Initially λ_{max} was optimized and was found to be 282. All spectral results are depicted in Fig.10a, 10b and 10c respectively.

Formulation B and C results (Fig 10b and Fig 10c respectively) indicate that A is having large difference in absorbance for 1 to 2 hour and all oil gets release up to 8 hrs., (Fig 10a) However, samples B and C show continuous release up to 10 hrs., i.e. retard oil release for 2 h and constantly release when added 4.5 or 6.0 ml of ECH as a cross linking agent.

We have also studied that the effect of ECH concentration on the release of eugenol in Formulation A, B and C at 282 nm. The absorbance-time profile is shown in Fig 12. It is found that amount of ECH help to cause sustainably release of eugenol (the pharmacologically main component present in the Clove oil).

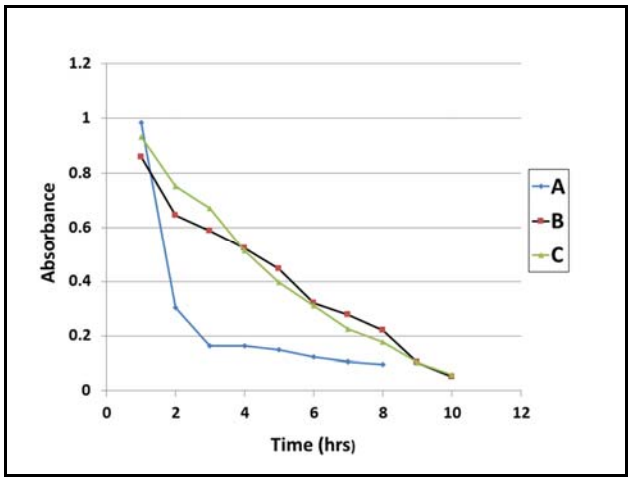

Fig 12: Absorbance of Eugenol at 282 nm

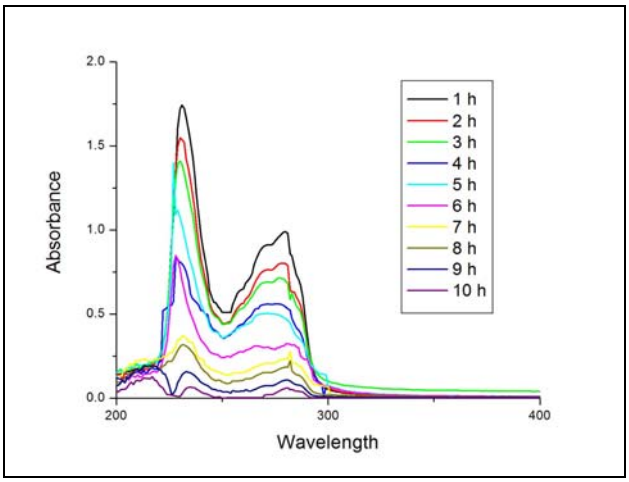

Fig 10b: UV Curves of eluents from Formulation B

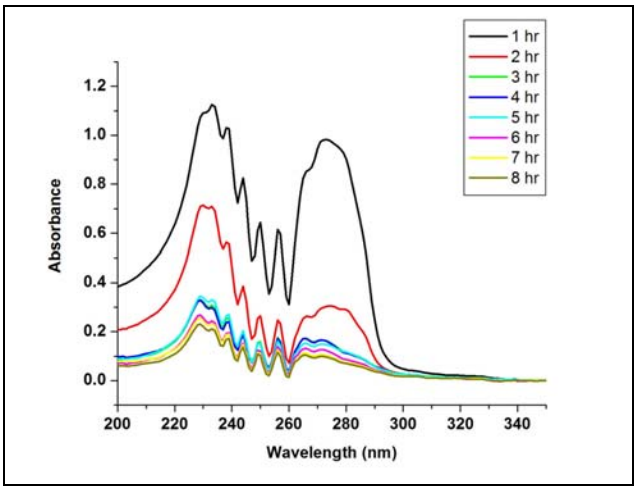

Fig 10a: UV Curves of eluents from Formulation A

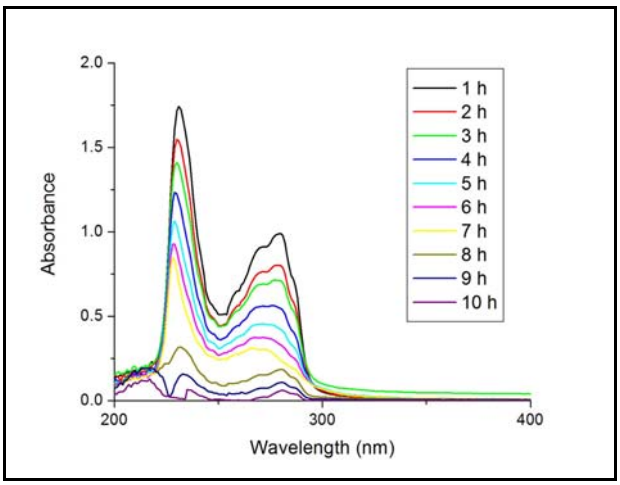

Fig 10c: UV Curves of eluents from Formulation C

In initial stage, the amount of released oil is large which slowly becomes constant as shown in Fig 10a. This means that the decay is exponential and follows first order kinetics.

4.3.1 Kinetics of release study

By studying the plot of the recorded UV-visible absorption profile for release of eugenol at 282 nm present in the core material in xylene, we have analyzed the kinetics of the core release. "We assumed that the release follows a first order kinetics and made a graph of ($\log A_\infty - A_0 / A_\infty - A_t$) verses time[Fig 11a, 11b and 11c**]** Where A_∞, A_t and A_0 refers to absorption values (optical density / extinction coefficient) at infinity, time (t) and initially. "All the data calculations are collected in Table-3. The equation used for calculation of first order velocity constant K is given below.

$$K = \frac{2.303}{t} \log \frac{A_\infty - A_0}{A_\infty - A_t}$$

Where t is the time in hours.

The linear plots of formulation B and C have fairly higher regressions of 0.9758 and 0.9007, respectively, than Formulation A(0.7049), thus indicating excellent suitability of Formulation B model on the kinetic release data for studying the release kinetics of core clove oil.

Table 3: Mathematical values for kinetic study.

	A			B			C	
Time(hrs)	**Abs.**	$\log \frac{A_\infty - A_0}{A_\infty - A_t}$	**Time(hrs)**	**Abs.**	$\log \frac{A_\infty - A_0}{A_\infty - A_t}$	**Time(hrs)**	**Abs.**	$\log \frac{A_\infty - A_0}{A_\infty - A_t}$
1	0.983=A∞	--	1	0.8600=A∞	--	1	0.934=A∞	--
2	0.303	0.1149	2	0.645	0.5760	2	0.753	0.6848
3	0.163	0.0335	3	0.588	0.4739	3	0.671	0.5225
4	0.162	0.0330	4	0.524	0.3821	4	0.514	0.3192
5	0.148	0.0257	5	0.447	0.2925	5	0.397	0.2150
6	0.123	0.0129	6	0.321	0.1768	6	0.310	0.1473
7	0.105	0.0039	7	0.277	0.1427	7	0.224	0.1025
8	0.097=Ao	--	8	0.221	0.1029	8	0.178	0.0552
			9	0.104	0.0299	9	0.104	0.0234
			10	0.050=Ao	--	10	0.058=Ao	--

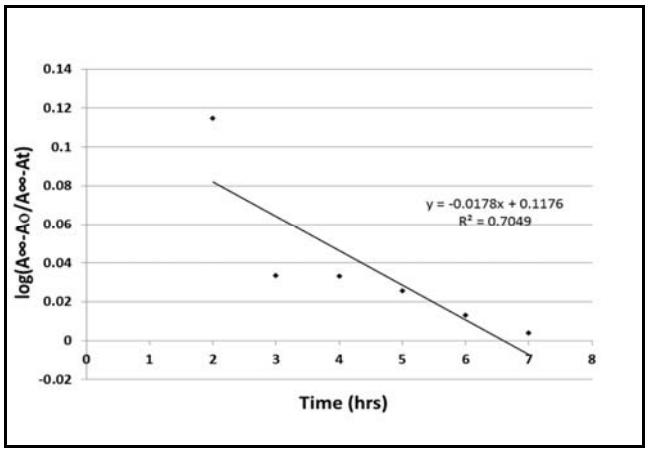

Fig 11a: Kinetics of release study for Formulation **A**

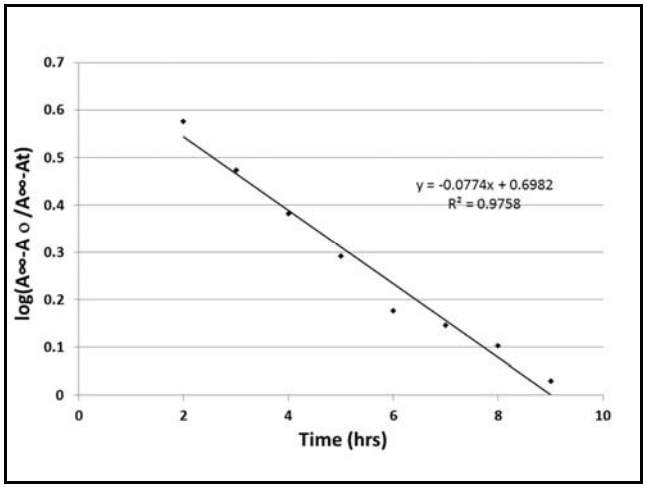

Fig 11b: Kinetics of release study for Formulation **B**

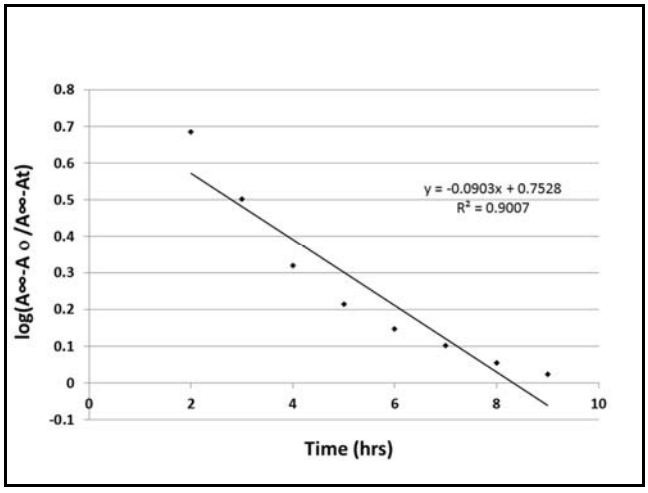

Fig 11c: Kinetics of release study for Formulation **C**

5. Conclusion

The purpose of the present work was to prepare controlled release microcapsule formulations (F1, F2 and F3) with variations in loading of clove oil and (A, B and C) with variations in loading of ECH as a crosslinking agent using ethyl cellulose as a shell material, by applying the solvent evaporation technique. The physical characteristics of stable formulation were carried out by Scanning electron microscopy (SEM), Particle size analysis, Thermogravimetric analysis (TGA) and infrared spectroscopy.

In present work we prepared EC microcapsules. The chemical composition of synthesized microcapsules and core clove oil was confirmed that core moiety present in microcapsules by FTIR spectroscopy. Particle size distribution of synthesized microcapsules were in the range of 10-520 µm and mean particle size was 110 µm. SEM images capsules are uniform and spherical in shape. The release study of clove oil follows first order/Kinetics meaning that encapsulated material stability is dependent on shell wall of EC.

The release behavior of clove oil through the ethyl cellulose (EC) wall was studied using a weight loss method by extracting core material and measuring UV absorbance. The decreasing absorbance with time confirmed the slow release of the core material and about 27% release was observed for F2 in 8hrs. It is observed that a variation of ECH concentration in formulation affects the retardant capacity of clove oil. The extracted EC shell from microcapsules was also found to have a good thermal stability than the core. The present investigation thus, confirmed that clove oil as an essential oil could be microencapsulated with 1 g of EC polymer, 3.0 ml of oil loading and 4.5 ml of ECH for better protection against volatility and efficiently controlled release application.

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