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Solubilization of Flavors in Aqueous Media of Triblock
copolymer of PEO-PPO-PEO Pluronic®

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"قل أن صلاتي ونسكي ومحياي ومماتي لله رب العالمين"

Program of Postgraduate Studies in Applied and Industrial Technology

College of Science and Technology

Deanship of Graduate Studies

**“ Solubilization of Flavors in Aqueous Media of Triblock
Copolymer PEO-PPO-PEO Pluronic ®”**

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Copolymer of PEO-PPO-PEO Pluronic®**

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A thesis Submitted in Partial fulfillment of requirements for the degree of master of

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June, 2004

Declaration

I Certify that this thesis submitted for the degree of Master is the result of my own research, except where otherwise acknowledged, and that this thesis (or any part of the same) has not been submitted for a higher degree to any other university or institution .

Signed 

Yasser Taha Hassan Abu Alia

Date : 10/6/2004

**TO MY
PARENTS
AND TO
MY FIANCÉE**

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ABSTRACT

Most flavour materials are strongly hydrophobic in nature, and hence not soluble in water, the formulation efforts have meant a challenge for the professionals. One solution is to solubilize these compounds into aqueous surfactant association structures such as micelle or microemulsions, and liquid crystals. Triblock copolymer poly(ethylene oxide)-poly(propylene oxide)-poly(ethylene oxide) (PEO-PPO-PEO) is a good surfactant were used. Pluronic L62, P104 with general formula $(EO)_6-(PO)_{34}-(EO)_6$, $(EO)_{27}-(PO)_{61}-(EO)_{27}$ respectively are used. The Effect of NaCl as additives were examined in both systems L62/aqueous salt/peppermint oil and L62/aqueous salt/benzaldehyde system in different salt concentration (in 0.5, 1M for benzaldehyde and 0.5 , 1 , 2 M for peppermint oil). The characteristic length (lattice parameter) for lamellar liquid crystal ($L\alpha$) in the P104/water/peppermint oil system was determined at constant copolymer concentration (73wt.%) with different peppermint oil concentration using small angle X-ray scattering (SAXS. Different microscope photographs for different anisotropic regions as lamellar ($L\alpha$), hexagonal (H_1) and reverse hexagonal (H_2) liquid crystal for all previous systems were taken under polarized microscope with digital camera. Ternary phase diagrams were determined for both flavours using a titration technique where L62/water/peppermint oil, L62/water/benzaldehyde, and P104/water/peppermint oil were presented here. L62/water/peppermint oil system shows a better solubility for peppermint oil over benzaldehyde when compared. P104/water/peppermint oil system show a rich one – phase homogeneous region due to its relative hydrophilicity/ hydrophobicity. NaCl as additives shows a pronounced effect on phase behavior for both flavor systems as direction of shifting and effect on solubility.

The trends in the SAXS lattice parameter and the interfacial area per block copolymer molecule help establish the location of the peppermint oil in the lamellar liquid crystal. The calculation show that as peppermint oil introduced to the P104/water binary system, the lattice parameter (d) decreased as a result of increase in the interfacial area.

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CHAPTER I
INTRODUCTION

1.1 Flavors

Flavors are an important category of additives which have been used extensively in many industries. Essential oils are highly concentrated flavors, so they must be diluted before application in order to facilitate handling and to guarantee an equal distribution of flavor among the products. Most essential oils have low solubility in water due to their partial hydrophobic nature. The other alternative is to solubilize them in an organic solvents. These solvents are either expensive or prohibited for religious purposes and for environmental concerns. The most acceptable solution is to emulsify the oil in water ⁽¹⁾.

Oil of *Mentha Piperita* L. (peppermint oil) is, a widely used essential oil ⁽²⁾. It is a colourless, pale yellow liquid with a strong agreeable odour and powerful aromatic taste ⁽²⁾. Its chemical composition has the ratio of menthol (20-54%), menthone (5-45%), menthyl acetate (1-29%) and menthofurans (1-8%). Its composition depends on various factors, such as geographical location, environmental conditions and plant ontogenesis. Peppermint oil has a wide range of applications. It is used in food, cosmetic and pharmaceutical industries ^(1,3). Peppermint oil used as larvicidal against different species of mosquitoes, as a digestive aid for generations, treatment of the irritable bowel, dyspepsia, antibacterial for respiratory tract pathogens ^(1,4-6).

Also the other important flavor is benzaldehyde (bitter almond oil) which has a wide range of applications in industries. More than 150 years ago, benzaldehyde was the first flavor compound identified. Natural benzaldehyde is usually liberated from amygdalin, a cyanogenic glycoside present in fruit kernels, and is used as a key ingredient in cherry and other natural fruit flavors ⁽⁷⁾.

Flavours and fragrance formulation in mostly use alcohol as solvent, but the recent legal restrictions on volatile organic solvents have prompted the industry to change to aqueous solubilized system ⁽⁸⁾. Surfactant that have both hydrophobic and hydrophilic groups in their molecules, can form molecular aggregates such as micelles and microemulsion in aqueous solutions. One of the most important functions of these molecular aggregates is to solubilize oil ⁽⁹⁾. Nagarajan (10), investigated the solubilization of hydrocarbons by aggregates of Pluronic® block copolymer in water. The initial studies of solubilization of fragrance compounds by surfactant association structures were concerned with micellar solubilization in aqueous systems ^(11,12 - 14). Suhaimi et al (15,16), have investigated the solubilization of fragrance compounds into the more complex microemulsion systems of water.