CHAPTER I

INTRODUCTION

1.1) MICROENCAPSULATION

The aim of microencapsulation technique is to enclose gases, liquids, or solids within a shell material which can be a wax, resin, or any one of a number of natural or synthetic polymeric materials. The reasons for and applications of microencapsulation are many. In general, microencapsulations are used most advantageously to protect their contents from air, moisture, microorganisms, and other contaminants. In this way, shelf life is increased and spoilage is reduced as well. The contents may be released by breaking, crushing, melting, dissolving, or in some way rupturing the skin. In some case the shell may allow the slow, prolonged release of its contents by diffusion through the wall or by decomposition of the wall in certain environments. Certain other applications may require that the microcapsule contents be retained permanently within the shell walls, which is also possible. Capsule walls resistant to acid can be adapted to dissolve and release their contents in an alkaline environment. Wall materials can be designed to allow the passage of some materials, while selectively blocking out others.

There are many methods to produce microcapsule such as spray-drying emulsion with dissolved film-former as continuous phase, spray-cooling a hot melt emulsion, condensation of a water soluble resin as emulsion continuous phase around liquid core droplets then spray-drying, coacervation of gellable colloid materials as emulsion continuous phase around liquid core droplets. The recent technical development of microencapsulation is interfacial polycondensation. This techniques

produce a thin, high molecular weight polymer film as the capsule shell.

1.1.1) Microencapsulation by Spray-drying

In this method, a stable emulsion is produced in which the continuous phase contains a film-forming material capable of forming the capsule shell which is insoluble in the encapsulated fluid. The discontinuous or dispersed phase of the emulsion constitutes the encapsulated fluid, and comprises other materials suspended or dissolved in a non-volatile liquid. The emulsion is then spray-dried in the hot chamber to evaporate solvent. The resulting product is a dry, free-flowing powder (1).

In the first modification of this process, microcapsules are produced by spray-drying an emulsion with dissolved film-former as the continuous phase. Some film-formers which are useful in this process are casein, zein, carboxy methyl cellulose, methyl cellulose, hydroxy ethyl cellulose, cellulose acetate, petroleum hydrocarbon resins, and other synthetic resins. Inorganic film-formers such as sodium silicate have been employed also, although such film-formers are less desirable because of the difficulty of maintaining a stable emulsion.

The second modification of this process, microcapsules are produced by spraying an emulsion with a hot melt composition as the continuous phase into a cooling chamber. Film-formers useful in this process include natural and synthetic waxes such as mineral, vegetable, animal, and Fischer-Tropsch waxes, sometimes modified synthetic resin materials.

It will be apparent that microcapsules prepared according to these examples permit considerable flexibility in the choice of film-formers and encapsulated fluids.

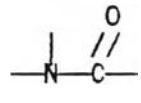
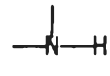
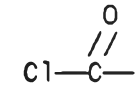
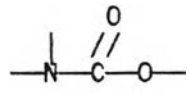
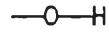
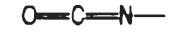
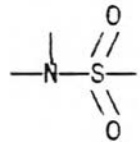
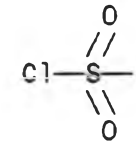
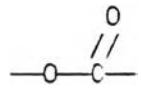
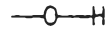
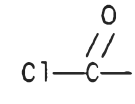
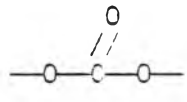
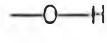
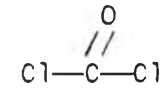
1.1.2) Microencapsulation by Coacervation Process

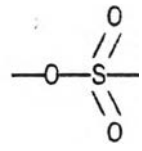
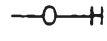
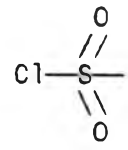
In general, the capsules, which are encapsulated fluid droplets and gellable complex polymeric material solution are formed by causing deposition of complex polymeric material around encapsulated fluid droplets as nuclei by process of causing coacervation by the addition of a coacervating agent into a mixture of different complex polymeric materials solution in which the encapsulated fluid droplets are dispersed. The coacervating agent means that the condition (such as temperature or concentration variation) or addition chemical ingredient (such as acid, base, salt or solvent) which causing the polymeric material separates from the medium to enclose around the fluid droplets. The gellable complex polymeric materials used in the solution must be ionizable and exist in the mixture as opposite electric charges. This may be brought about by selection of the polymeric materials which have anionic, cationic or amphoteric charges after adjusting the pH of the solution mixture. If desired, after the gellation, any of the further steps of hardening the gelled material, such as separating the microcapsules from the remaining liquid, drying it, and sieving it to the desired particle size, may be used. Some gellable polymeric materials which are useful in this process are gelatin, gum arabic, pectins, casein, copolymer of methyl vinyl ether and maleic anhydride, copolymer of ethylene and maleic anhydride, and copolymer of styrene and maleic anhydride (2), (3), (4), (5), (6).

1.1.3) Microencapsulation by Interfacial Polycondensation

This process makes use of a modification of known interfacial polycondensation techniques to produce a thin, high molecular weight polymer film as the capsule shell. Essentially, the process comprises bringing two reactants together at the reaction interface between the emulsion phase where polycondensation occurs

virtually instantaneously to form a thin film insoluble in the parent media of the reactants. Some classes of polymer which can be prepared by this technique and which have been use in encapsulating a variety of materials such as polyamide using diamine reacting with dicarbonyl chloride; polyurethane using bisphenol reacting with diisocyanate; polysulfonamide using diamine reacting with disulfonyl chloride; polyester using bisphenol reacting with dicarbonyl chloride; polycarbonate using bisphenol reacting with phosgene and polysulfonate using bisphenol reacting with disulfonyl chloride. The variety of materials are showed below :-

	Linking Structure in Polymer	Reacting Groups in Intermediates	
Polyamide		 diamine	 dicarbonyl chloride
Polyurethane		 bisphenol	 di-isocyanate
Polysulfonamide		 diamine	 disulfonyl chloride
Polyester		 bisphenol	 dicarbonyl chloride
Polycarbonate		 bisphenol	 phosgene

Polysulfonate   

bisphenol disulfonyl chloride

In this process, the interface for the reaction is provided by emulsifying one reactant for the condensation polymer in a continuous phase containing the second reactant. The substance to be encapsulated will also be contained in the dispersed phase. However, in order to control the formation of the capsules, one reactant for the condensation polymer, together with the substance to be encapsulated, is first emulsified in a continuous phase containing no second reactant. Thereafter, additional continuous phase containing the second reactant is added to the emulsion. The polymer shell will then form at the interface of the dispersed substance and encapsulate the material (7), (8), (9).

1.2) SOME IMPORTANT INFORMATION ON MICROENCAPSULATION

Several important methods are presently available for microencapsulation as follows :-

Green (10) described the encapsulation of hydrophilic oil by gellation and gelatin-gum arabic complexes utilizing a "coacervation" process. The process is limited to water-soluble hydrophilic colloids and cannot be used for encapsulation of water or water-soluble substances. Moreover, materials which are sensitive to the acidic pH of the process are inoperable.

The microencapsulation of water-insoluble material filled within a water-insoluble non-thermoplastic synthetic resin of the aminoplast type comprising reaction products of urea and formaldehyde which are capable of polymerization from a water-soluble prepolymer state under

acid conditions in aqueous media to form substantially water-insoluble polymers as a microcapsule shell is described.

Brynko has disclosed the encapsulation by addition polymerization process whereby at least one selected monomer is dissolved in an oil in which the solid polymer material is insoluble. The solution is then dispersed as droplets in a polar liquid where polymerization occurs due to the influence of a catalyst which can be introduced in any of the phases. The polymer material deposits at the interface of the oil droplets and polar liquids forming an individual solid wall around each oil droplet resulting in discrete, pressure rupturable, substantially spherical, oil containing capsules. Additionally, this process also excludes the encapsulation of water-soluble materials.

Vandegaer (9) described in one U.S. Patent that encapsulation was developed by interfacial polycondensation between intermediates contained in two immiscible liquids. Droplets of one of the two immiscible liquids containing one of the intermediates, are successively formed and released, by an appropriate injection. These occur within a body of the other liquids and are forced to travel along a predeterminated path while content of the second intermediate is maintained in the second liquid over, at least, part of such a path. The intermediates thereby react at the interface of the droplets with the other liquid to produce the solidified polymeric skin.

Macaulay (1) enumerated a wide range of substances encapsulated by a variety of film formers. The suspension-evaporation method described can be used for encapsulation with polymers which are soluble in suitable solvents. For high molecular weight polymers, however, available solvents are very limited and solution viscosities are high, substantially limiting the process as a practical matter.

1.3) STRUCTURES OF MICROCAPSULE

Microcapsules have a wide variety of structures and some of these are shown in Figure 1. Figure 1-a illustrates a capsule where A is the capsulated material and B is a shell of high molecular weight polymer produced by interfacial polycondensation. Figure 1-b depicts a capsule where A is the encapsulated material and B is the shell composed of a high molecular weight polymer produced by interfacial polycondensation and C is a second shell applied by a spray-drying process or similar technique, or by coating the dry particulate material. In Figure 1-c, A is the encapsulated material, B is the interfacial polycondensation shell and E represents a second phase which may be similar to or different from A. D represents a second condensation polymer shell which may be the same as or different from B. In Figure 1-d, G represents the capsules of Figures a, b, or c, F is a dispersion medium for capsules G, and H represents an interfacial polymer shell. The structures of any microcapsule can be investigated by electron microscopy (7).

1.4) DETERMINATION OF MICROCAPSULE CHARACTERISTICS

1.4.1) Microencapsulation Efficiency

The efficiency of encapsulation is measured by determining the amount of unencapsulated fluid. The unencapsulated fluid is extracted from the capsule slurry by mixing it with an inert extracting solvent. The quantity of unencapsulated fluid in the extracting medium is then measured by gas chromatograph. The total fluid in the system is determined by extracting the capsule slurry with a reactive extracting solvent which will remove the fluid through the capsule walls, again followed by chromatographic analysis. The ratio of unencapsulated fluid to total fluid can be used to determine the

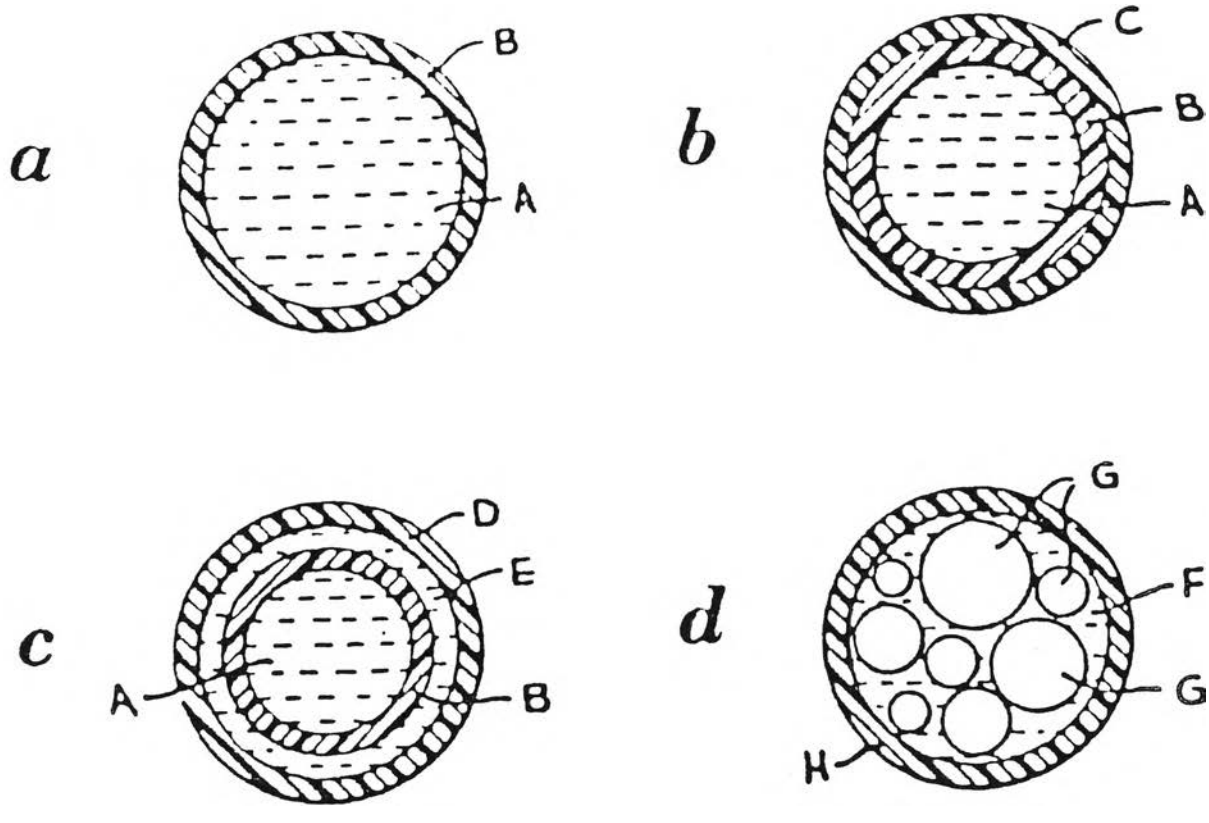


Figure 1 Illustration of Microcapsule Structures.

encapsulation efficiency. The polycondensation process can be controlled quite readily at an encapsulation efficiency of at least 98%. To demonstrate the impermeability of the capsule walls, the experiments have shown that encapsulated volatile solvent such as toluene and xylene can be contained without measurable loss at temperature close to their boiling points for periods in excess of 24 hr (11).

1.4.2) Average Size Determination by Turbidimetry

This method owes its popularity to the fact that it can be carried out with any ordinary photometer (12). The theoretical basis is Lambert's law :-

$$I = I_o \cdot e^{-\tau \cdot x} \qquad \text{..... Eq.(a)}$$

which says that light of the primary intensity I_o is more strongly subdued the longer there is the way x through a scattering medium. I is the light intensity on passage of the medium, τ is referred to as the turbidity coefficient.

The value of this turbidity coefficient depends on the weight concentration c and the radius rT of the scattering particle as well as on the relative refractive index NT/N_o of particles and medium and the wavelength Φ of the light. This means on the other hand that the particle size can be calculated from the turbidity coefficient if all other quantities are given. To this end, a reduced quantity τ/c which is referred to as the specific turbidity is used. Again the true particle radii are obtained only if the individual particles contribute to the overall turbidity uninfluenced by each other. This is guaranteed only in the case of an infinite dilution. Thereof, the specific turbidity must be measured at several concentrations and extrapolate to the value of τ/c where $c \to 0$.

The relationship between the specific turbidity at infinite dilution and the particle radius can now be defined as follows :-

$$\left(\frac{\tau}{c}\right)_{c \to 0} = \frac{4\pi N_o}{\Phi \cdot \sigma T} \cdot f\left(\frac{NT}{N_o}, N_o, rT, \Phi\right) \quad \ldots\ldots \text{Eq.(b)}$$

σT is the density of the polymer pellets. The dependencies of the specific turbidity, relative refractive index, the particle radius and the quantities N_o and Φ are a very complex problem. In practice they are calculated with the aid of tables which relate the quantities to each other. The rT values are weight average.

Since the intensity of a light beam which has passed a medium is the complementary quantity to the intensity of the scattered light, the reservation to be made are the same as in light scattering method. In particular, the relationships between τ and rT vary considerably in different wavelength regions. If $rT < \Phi$, the turbidity increases by the third power of the particle radius. Where the particle dimensions approach Φ, the turbidity merely increases linearly with the radius. If the particles are larger than that, τ even takes the course of 1/rT. It thus happens that the turbidity as a function of the particle size passes through a maximum and declines thereafter. The higher the refractive index of the particles and shorter the wavelength of the light, the smaller is the particle size at which this maximum is reached. Above this maximum the relationship between turbidity and particle radius becomes rather arbitrary and therefore it is, for a given length of light waves and a given polymer NT, the upper limit at which particle size determination via turbidimetry still makes sense, this limit lies between rT = 3,000 and 4,000 Å.

In particular, a photometer is taken and compared the

light intensity I of a beam which has passed a dilute dispersion of a given concentration with the intensity I_o of this same beam after passage of a medium of pure water. This measurement is carried out at different concentrations of the dispersion. Having obtained the refractive indices and the particle density σT, everything else is left to the computer. The details of computer program are showed in Appendix I. Finally, the simplify equation for calculating the average particle size can now be defined as follows :-

$$d = 10^{(0.386A + 3.384)} \quad \text{..... Eq.(c)}$$

Where

$$A = \frac{\sum_{N=1}^{5} X Y - N \bar{X} \bar{Y}}{\sum_{N=1}^{5} X^2 - N \bar{X}^2}$$

X = log Φ (Φ = 400, 450, 500, 550, 600)

Y = log Γ (Γ = 100 - % Transmittance)

$\bar{X}$ = Average of log Φ

$\bar{Y}$ = Average of log Γ

N = Number of measurements

d = Particle diameter (μm)

1.4.3) Size Distribution Determination by Ultracentrifuge

The principle combines the sedimentation and photometric methods, so analysis is based on Stokes' law and the proportional relationship between absorbance and particle concentration (12).

The relationship between terminal velocity and particle size under gravity is expressed by Stokes' equation in the following manner :-

$$V = \frac{H}{T} = \frac{g\,(\delta_p - \delta_1)}{18\,\Omega} \cdot d_x^2 \quad \text{..... Eq.(1)}$$

Where

d_x = Particle diameter (cm)

V = Terminal velocity of the particle with size dx (cm/sec)

H = Sedimentation distance

T = Time required for H (cm) sedimentation (sec)

g = Gravitational acceleration (cm/sec^2)

δ_p = Particle density (g/cm^3)

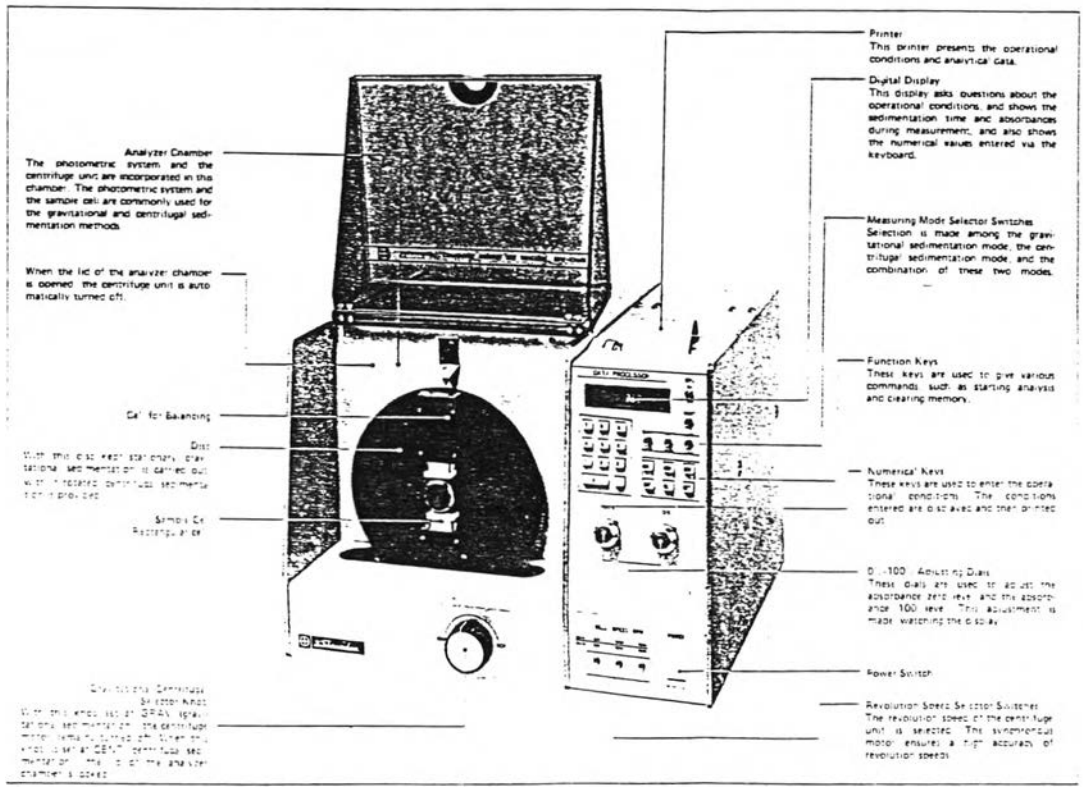
δ_1 = Medium density (g/cm^3)

Ω = Medium viscosity (g/cm·sec)

Since g, δ_p, δ_1 and Ω are constants that depend upon the sample. The details of principle are showed in Appendix II. Thus, the particle size distribution can be automatically calculated by the microcomputer which links to the analyzer as shown in Figure 2.

1.4.4) Particles Image by Microscopy Methods

Optical and electron microscopy methods are the most popular for determination of the average particle size and particle size distribution. The photography is the method which offers much more information than any indirect methods of determination. The complicated conclusion must be drawn and given the enlargement factor must be included as well. The photographed particles are shown in the micrograph and any required information can be obtained exactly by measuring and counting the size and the number of particles. But the optical microscopy cannot make the very small particles visible as such. Because the wavelength of visible light is longer than the diameter of particles. There is a way-out to determine the very small particles by using electron microscopy method. The wavelength is reduced to employ radiation to such an extent that it is much smaller than the object to



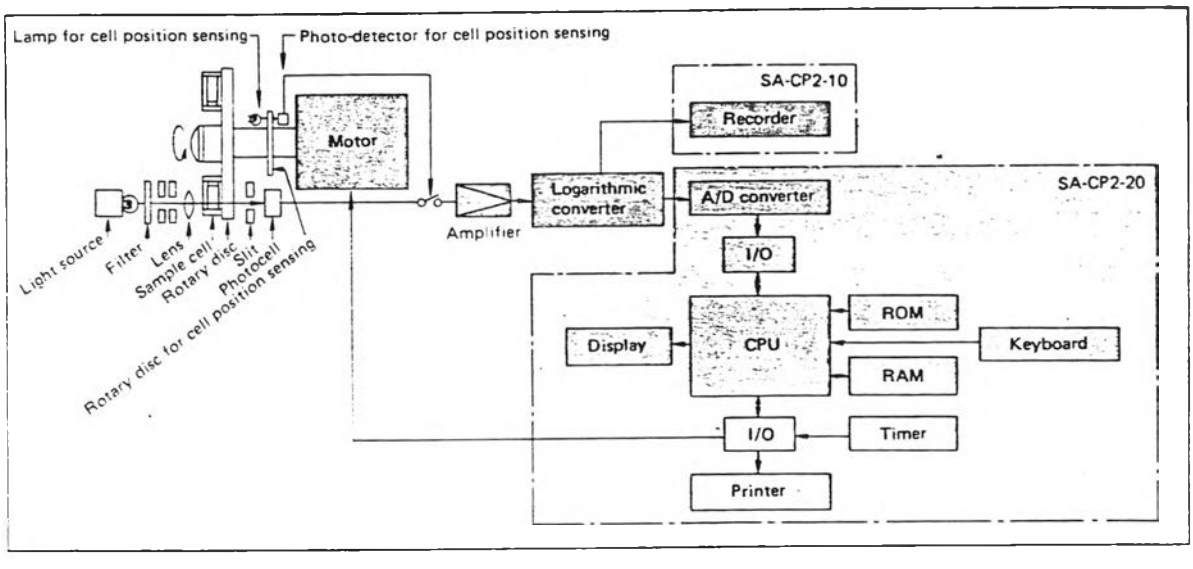


Figure 2 Structural Diagram of Particle Size Analyzer with Computer.

be imaged. The usable wavelength range of the visible light has its lower limit at about 0.4 micrometer, therefore the very small particles cannot be seen in the optical microscope. Short-wave electron radiation, however, increases the magnification of the optical microscope about two decimal powers and this suffices to show even the small dimension of the particles. In its order form transmission electron microscope is very such like the optical microscope. The difference lies in the lenses, which are electrostatic or magnetic fields in the electron microscope. The resolving power of such instruments lies at < 10 angstrom. Of a different structure is so-called scanning electron microscope which derives its functions from video technique. The object to be imaged is scanned in rapid succession by a beam of primary electrons. Recorded, however, is the intensity of the secondary electrons emitted by the object, which are synchronously focused on a screen. The scanning electron microscope is thus an emission electron microscope. At about 100 angstrom its resolving power lies between that of the optical microscope and the transmission electron microscope.

Both electron microscopes serve to produce the images of polymer particles. The problem is, however, to contrast the particles with their environment and to ensure that the high-energy radiation causes the least possible change in them. Therefore, specimen preparation is the most important aspect of electron microcopy. It is essential to make sure that the image of the spatial distribution of the polymer particles in a dispersion be as true to nature as possible. The polymer particles are soft and deformable under the highly energetic electron beam. "Electron Beam Hardening" is the most widely used method. To this end, the specimen is exposed at very low temperatures to high energy electron beam, which creates many reactive sites at the macromolecules within the polymer particles to be cross-linking. However, the polymer particles are slightly shrinks as a result of

cross-linkage, its diameter now being somewhat smaller than in the original state. To make quite sure, there is the method of oblique sputtering, in which a carbon or metal film is deposited on the specimen under vacuum for a given angle. The sputtering angle being known, the dimensions of the imaged polymer particles can be accurately determined from the typical shadow. If the specimen is not examined with the transmission electron microscope but with the scanning electron microscope, the specimen surface must be created to be a good emitter of secondary electron. In this case the deposition of a conductive layer, carbon or gold film, is particularly important (13).

1.5) THE APPLICATIONS OF MICROENCAPSULATION

The art of encapsulation has been the object of considerable attention in recent years in view of the increased need to maintain a reactive material in an inert stage until such time as it is called upon to perform a given function. Thus, inertness is provided by interposing, by encapsulation, a non-reactive barrier or shell between the reactive material and its immediate surrounding. Removal of the barrier or shell by any suitable means "activates" the encapsulated substance. In this manner, the handling properties of solids can be conferred on liquids and gases. Adhesive can be made non-adhesive until adherence is required and reactive chemicals can be maintained inactive until such time as their reactivity is released. Flammable substance can be rendered non-flammable while being transported or during storage, but readily released unchanged when desired. More specifically, the technique of encapsulation is employed effectively in the manufacture of manifold paper where a thin layer of microscopic capsules containing a suitable marking substance are bonded to a pliable sheet. The marking substance is maintained inactive until the capsules are ruptured by pressure of a stylus or other means. Encapsulated substances are useful in the preparation of drugs where a delayed effect is desired. Thus, the active

ingredient can be coated with a suitable protective layer and when the drug is taken internally, it will not immediately affect the patient but remains inactive until the polymer layer is dissolved. As is apparent, depending on the encapsulated substance, the influence of the drug can be delayed from a matter of minutes up to several hours. The encapsulation technique can also be used in the preparation of pressure sensitive adhesives for application to placards, envelopes, etc., where it is necessary that the surface remain non-adhesive until adherence is desired. By employing encapsulating substances, it is not necessary to wet the surface but adherency is obtained by merely applying pressure (14), (15).

1.6) THE GENERAL INFORMATION OF MICROENCAPSULATION BY INTERFACIAL POLYCONDENSATION

Due to the variety of techniques available for microencapsulation, in this thesis, the interfacial polycondensation method was selected for synthesis of polymeric shell in which a conventional emulsion process was also involved. This thesis relates to a process of encapsulation and to novel encapsulated substances. More particularly, the thesis embraces a substance having a shell of insoluble, infusible, high molecular weight condensation polymer. The encapsulation process comprises division and dispersion of the substance to be encapsulated as a discontinuous phase, within a continuous fluid phase. Each phase must contain an intermediate or an intermediate must be added, which will react with the intermediate in the other phase to form a continuous high molecular weight condensation polymer film at the interface of the two phases. The dispersed substance is enclosed within the polymer film.

1.6.1) Microencapsulation by Interfacial Polycondensation

The preferred and mechanically most simple method of providing the interface for encapsulation is to disperse or emulsify one reactant for the condensation polymer in a continuous phase containing the second reactant. The substance to be encapsulated will also be contained in the dispersed phase. However, in order to more accurately control the formation of the capsules, it can be convenient to emulsify or disperse one reactant for the condensation polymer, together with the substance to be encapsulated in a continuous phase and thereafter add additional continuous phase containing the second reactant to the dispersion. The polycondensation polymer shell will form at the interface of the dispersed substance and encapsulate the material. The dispersion and emulsion can be stabilized by addition of surface active agents or protective colloids to the continuous phase (8).

1.6.2) The Advantages of Microencapsulation by Interfacial Polycondensation Method

The spray-drying and coacervation encapsulation processes described earlier utilized film-forming polymeric materials soluble in a range of solvents. For high molecular weight polymers, the solvents available are very limited and solution viscosities are high, thereby restricting their use in the process. The interfacial polycondensation technique can produce a capsule shell consisting of high molecular weight polymer insoluble in organic solvents. A particular advantage of interfacial polycondensation process is that it provides a method of encapsulating water or water soluble substances. To control the formation of the capsules, one reactant for the condensation polymer, together with the substance to be encapsulated, is first dissolved in water phase and emulsified in continuous oil phase containing emulsifier. Thereafter, the second reactant which dissolves

in the continuous oil phase is added to the emulsion phase. The polymer shell will then form at the surface of the dispersed water phase and encapsulate the material. The spray-drying and coacervation processes tend to give cluster of discrete capsules illustrated in Figure 3. The interfacial polycondensation technique, to a certain extent, gives discrete capsules shown in Figure 3, due to the encapsulation occurring at the individual dispersed droplets. The interfacial polycondensation technique produces a microcapsule having high uniformity in thickness of polymeric shell which is insoluble in the continuous phase and infusible during processing at high temperature. Because the interfacial polymerization occurs at each encapsulated fluid droplets, so that the efficiency of encapsulation is nearly 100%. Therefore, the polymeric shell formation should occur instantaneously after the second reactant has been added (7).

1.6.3) Dispersion and Emulsification Process

The dispersion or emulsion can be produced by the standard suspension and emulsification techniques known in the art. Thus, dispersions or emulsions can be prepared by agitation, preferably in the presence of one or more emulsifying agents. The efficiency of the emulsification depends among other factors, on the type and degree of agitation and the manner in which the emulsifying agent is introduced. The primary function of the agitation is to break up both phases of the emulsion so that the one which will become the dispersed phase is able to form small globules. The emulsifier is employed to lower the interfacial tension since the lower the interfacial tension the lower the amount of mechanical energy needed to break up the phases. Where the interfacial tension of a system is extremely low, spontaneous emulsification may result. The preparation of the emulsions can be facilitated by various types of colloid mills and homogenizers engineered to obtain maximum shear action of the fluid and enhance the

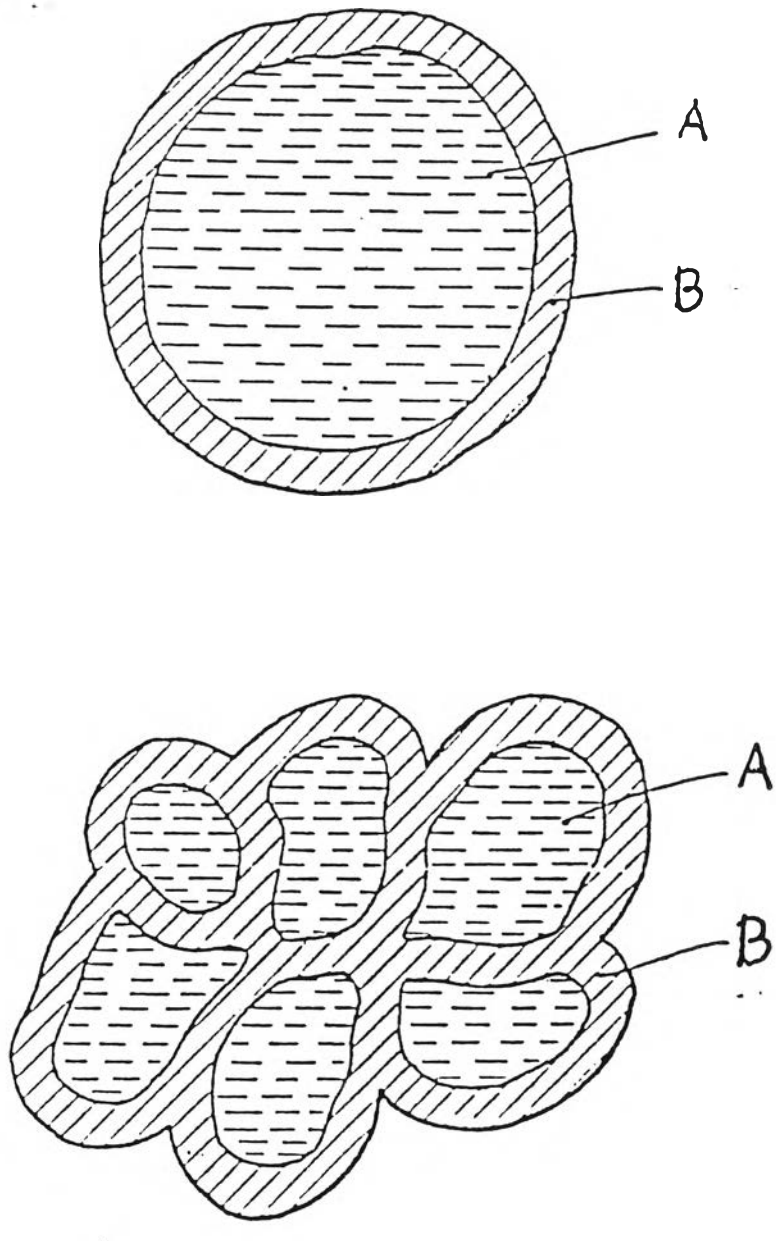


Figure 3 Structure of Cluster and Discrete Microcapsule.

formation of fine uniform globules (16).

1.6.4) Emulsifying Agent

Emulsifying agents which are operable in preparing the emulsions include surfactants as well as protective colloids. Surfactants are the long chain polar and non-polar compounds which have the hydrophilic and hydrophobic part in the same molecule. They are classified as anionic such as sodium lauryl sulfate, cationic such as stearyl amine, nonionic such as ethylene oxide derivatives of nonylphenol, and amphoteric such as cetyl betaine according to the charge of the effective part of the molecule. Protective colloids are the high molecular weight hydrophilic molecule which are classified as natural products such as gums, starches, proteins, etc., and synthetic products such as poly(vinyl alcohol), poly(sodium methacrylate), etc. Emulsifying agent is known to be readily adsorbed at the interface to prevent agglomeration of the non-continuous phase (16).

1.6.5) Polycondensation between Diacid Chloride and Diamine

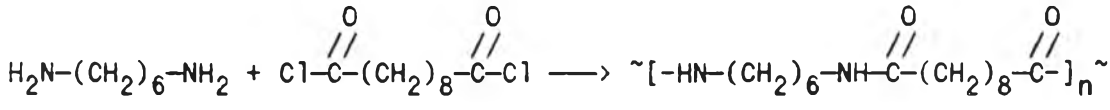
A number of highly reactive compounds, interaction of which results in amide linkages, can be used as reactants for interfacial synthesis of polyamide (17), (18), (19). Acylation reaction of diamines with acylhalides is most widespread and useful. This reaction is practically irreversible and can be described by a second-order rate expression. It follows the mechanism of nucleophilic substitution. The reaction is rapid, is catalyzed strongly by water and by a number of other additives, and is multistage :-

$$H_2N\text{–}R_1\text{–}NH_2 + Cl\text{–}\overset{\overset{O}{\parallel}}{C}\text{–}R_2\text{–}\overset{\overset{O}{\parallel}}{C}\text{–}Cl \xrightarrow{k1} [\sim R_1\text{–}NH_2 \cdots \overset{\overset{O}{\parallel}}{C}\text{–}R_2\sim]\overset{\ominus}{Cl} \xrightarrow{k2}$$

$$\sim R_1\text{–}NH\text{–}\overset{\overset{O}{\parallel}}{C}\text{–}R_2\sim + HCl$$

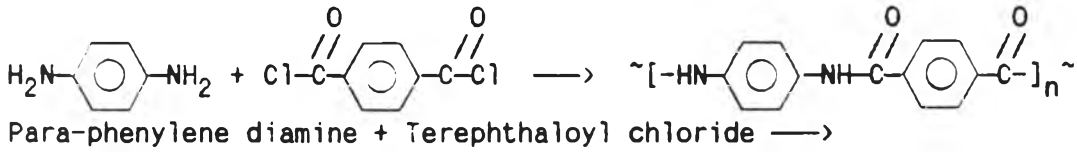
In the first reaction stage, the formation of an intermediate complex is more rapid than its decomposition. Hydrogen chloride, formed in the reaction, is capable of reacting with nonacylated amino groups in the intermediate, resulting in the formation of nonactive salts of amines under the condition of the synthesis which can be avoided by the additional of an acid-receiver such as sodium hydroxide.

By the appropriate selection of chemicals for this thesis, polyamide can be synthesized by the following reactions :-



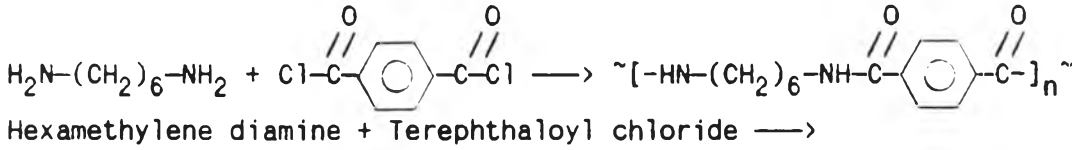
Hexamethylene diamine + Sebacoyl chloride ——>

Poly(hexamethylene sebacamide)



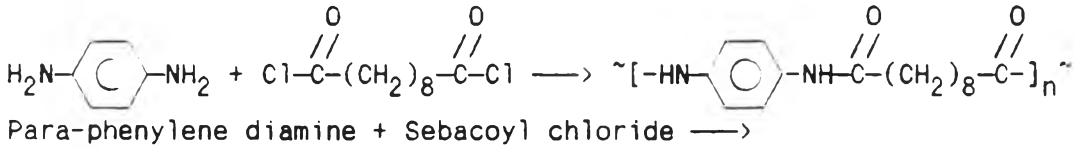
Para-phenylene diamine + Terephthaloyl chloride ——>

Poly(para-phenylene terephthalamide)



Hexamethylene diamine + Terephthaloyl chloride ——>

Poly(hexamethylene terephthalamide)



Para-phenylene diamine + Sebacoyl chloride ——>

Poly(para-phenylene sebacamide)

1.6.6) Homogenizer

In the emulsification process, the high-shear type

emulsifying mixer is an important machine to break up oil phase into tiny oil droplets. T.K. Homo Mixer is a revolutionary mixer operated by the concept entirely different from that of ordinary mixers. With their original design and mixing principle, they are capable of emulsifying, homogenizing, dispersing, dissolving and reaction-accelerating in liquid-liquid, liquid-powder or liquid-solid processes quickly and easily.

Its mixing section consists of a high-speed turbine, a stator and disc-type deflection baffle as shown in Figure 4. The revolution of the turbine creates pressure difference between the top and the bottom of the turbine. This pressure sends up the processing material to the higher part of the vessel through the gap between the turbine and the stator, during which the processing material receives severe shearing, impact and turbulence. Finally the processing material bumps against the deflection baffle and flows down to the receiver the same processes repeatedly until it is finely emulsified, homogenized, dispersed, dissolved or milled.

On all of T.K. Homo Mixer models, the major components such as mixing section, stator rod, and motor incorporate basically the same design. They are mixer of vertical type as shown in Figure 5. The mixing section consists of a turbine which rotates at high speed, and a stator held to stator rods (stationary member), both being made of high-grade stainless steel. The turbine is connected to the shaft which is directly coupled to motor with a flexible coupling. It rotates along the inside periphery of the stator at several hundred to several thousand revolutions per minute, while maintaining a precise and even gap. Processing material is sheared in this gap, receiving emulsifying, homogenizing, dispersing, dissolving and grinding actions. Three stator rods, made of high-grade stainless steel, support the mixing section accurately and firmly to motor base. These rods are fitted with a disc-

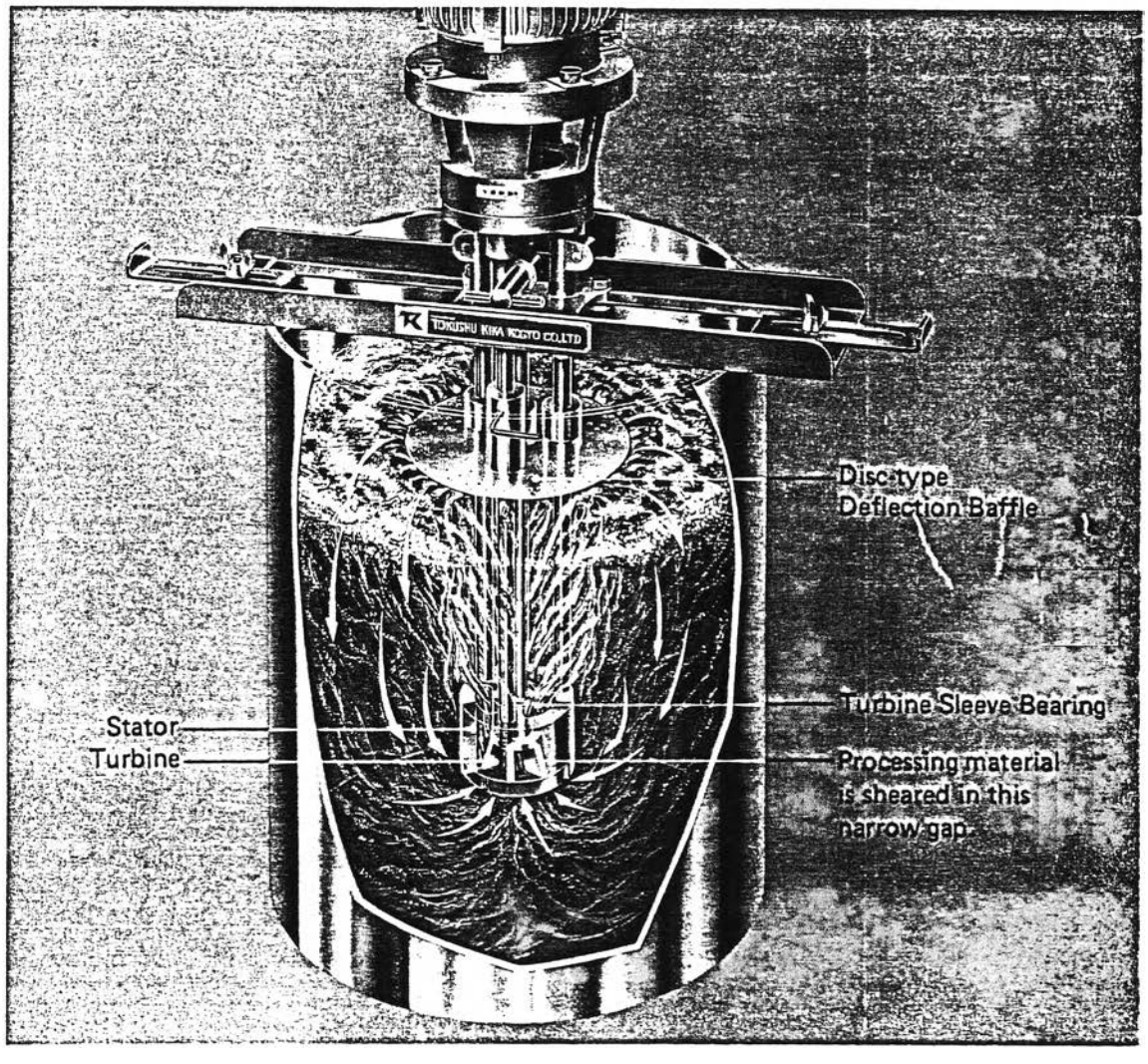
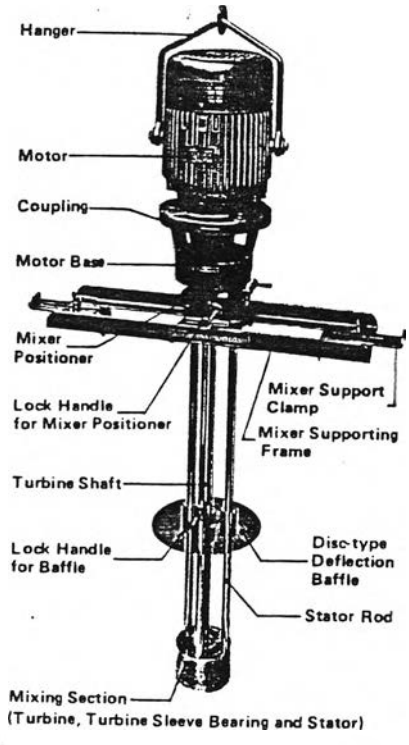
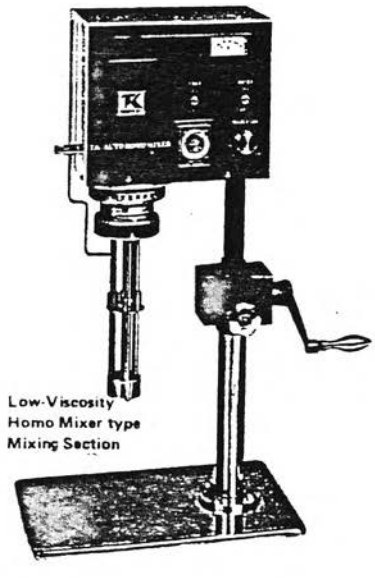


Figure 4 Original Design and Mixing of T.K. Homo Mixer.





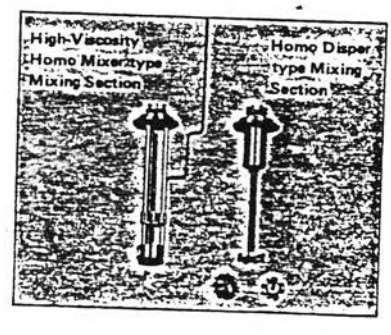


Figure 5 Production Scale (above) and Laboratory Scale (below) T.K. Homo Mixer with Major Components.

type deflection baffle which can be set in a desired position depending on volume of the liquid handled. This baffle facilitates a convection. The mixing section is accurately connected to the motor by means of the motor base. Bearings are built inside this motor base to support the high-speed turbine shaft which is directly coupled to the motor by means of a flexible coupling (20), (21).

1.7) OBJECTIVE AND SCOPE OF RESEARCH

Upon the great number of literature survey on syntheses of microcapsules, for industrial applications, by any existing techniques, it was found that, the state-of-the-art and the technique of interfacial polycondensation via emulsification step offered the most advantages as described in detail in the previous sections. Although this advancement is of current interest and the technique finds applications in various fields, the detailed information is still undisclosed. It is the objective of this research to study indepth the influential parameters governing particle size distribution and encapsulation efficiency of the microcapsules from which all chemical and physical properties, as well as shelf life and servicing ability are under control. This fundamental research should enable us to acquire the insight details of microcapsule syntheses which can be regarded as another model for the state-of-the-art as well.

Simple diacid chlorides and diamines were used as models for the syntheses of polyamide microcapsules under the following parameters :-

1) Types and concentrations of diacid chlorides and diamines.
2) Types of encapsulated materials.
3) Types and concentration of emulsifying agent.
4) Speeds of the homogenizer propeller.
5) Duration of homogenizing.

6) Stability of emulsification.

7) Encapsulation efficiency.

Under intensive investigations of the dependency of the parameters, the desirable polyamide microcapsules are to be obtained such as single microcapsule with controllable releasing properties, etc.

Furthermore details of the experimental work are presented in Chapter II.