

国家精品课程参考教材

Introduction to Polymer Physics

高分子物理导论

● 华南理工大学高分子物理教学组 编

华南理工大学出版社

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· 广州 ·

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序

华南理工大学高分子材料与工程相关本科专业始于 1952 年华南工学院成立初期的橡胶工学专业,现在高分子材料与工程专业已成为了广东省名牌专业。“高分子物理”课程一直是该专业重要的专业基础课之一,得到了学校一贯的重视和支持,2007 年“高分子物理”被评为国家精品课程。随着我国改革开放和聚合物产业的繁荣,对同时具有专业知识和外语能力的复合型技术人才的需求与日俱增。大学英语课程承担了学生普通英语的教育,而专业英语能力的培养更多的是依靠专业教师和专业课程。因此,有些学校开设了独立的“专业英语”课程。专业基础课程的中、英双语教学,会更有利于提高学生实际应用英语的能力。

当前,我国高分子物理的英文教材主要有两类:一类是国外出版的原版教科书,其价格不是我国的在校大学生都能承受的;另一类是我国出版社出版的国外教科书的影印版,如化学工业出版社出版的 *An Introduction to Polymer Physics* (D. I. Bower 编),种类很少,难以选择。更为重要的是,对于刚开始接触高分子学科的本科生,大量的专业术语成为学习的障碍,而通过母语的教科书能够更准确、更全面地理解高分子物理。因此,最理想的是有与汉语的《高分子物理》教科书相对应的英语《高分子物理》教科书。为了适应这种要求,华南理工大学高分子物理教研组的六位老师,以何曼君等人编写的《高分子物理(修订版)》(复旦大学出版社)的内容为基础,参考国外有关教科书,编写了这本英文版的《高分子物理导论》,供本科生双语教学使用。

这是一个大胆的尝试,中国人编写英文教科书有天生的不利条件,所以他们参考、摘引了许多英文原版教材的内容。这又是一种有益的探索,为我国高分子专业本科生在学习高分子物理基础知识的同时,尽早掌握有关的英语术语与表达,创造有利条件。望他们在探索的道路上不断前进。

中国科学院院士,华南理工大学教授,南京大学教授

程锦时

2010 年 10 月 29 日

PREFACE

The polymer science and engineering course at university includes physical polymer science, polymer blends and composites, and engineering behavior of polymers, organic polymer science, and polymer laboratory for interested seniors and first-year graduate students. There are also broad-based introductory courses at the senior level for students of polymer science and engineering. The students may earn a degree in either polymer science and engineering or materials engineering because courses are normally interdisciplinary and cross-listed. The physical polymer science is usually the basic course that the polymer students would take in university.

This text book is for the undergraduate bilingual course of polymer physics, which is of fundamental to the polymer materials and engineering and desired to be introduced in English to endow the students with the capability of using English in the specific area. Many Chinese students feel difficult to understand text books written in English because the contents are not arranged consistently with a Chinese book for comparison. Therefore, an English text book corresponding to a Chinese one has been looked forward for a long time.

This book is based on the *Polymer Physics* (Revised Edition) published by Fudan University Press. We have referred to lots of books and review journals in this field, such as *Principles of Polymer Chemistry* (P. J. Flory, Cornell University Press), *Introduction to Physical Polymer Science* (L. H. Sperling, 4th Ed., John Wiley & Sons), *An Introduction to Polymer Physics* (D. I. Brower, Press Syndicate of the University of Cambridge), *Principles of Polymerization* (G. Odian, John Wiley & Sons), *Textbook of Polymer Science* (F. W. Billmeyer, Jr., 3rd Ed. John Wiley & Sons), *Contemporary Polymer Chemistry* (H. R. Allcock, F. W. Lampe, J. E. Mark, 3rd Ed., Prentice Hall), *Modern Theory of Polymer Solutions* (H. Yamakawa, Harper & Row), *Viscoelastic Properties of Polymers* (J. D. Ferry, John Wiley & Sons), *Electrical Properties of Polymers* (T. Blythe, D. Bloor, Cambridge University Press), *Progress in Polymer Science* (Elsevier), *Advances in Polymer Science* (Springer), etc. Study questions are accompanied in every chapter to help the students to understand and apply the knowledge.

Chapter 1, Structure of polymer chain, by Mr. Ping Ning, covers the structure and conformation of chain molecules.

Chapter 2, Condensed state structure of polymers, by Dr. Feng Xue, describes both of amorphous and crystalline of polymers.

Chapter 3, Properties of polymer solutions, by Dr. Zhen Tong, discusses the interaction

between solvent and polymer, thermodynamics and dynamics, phase behavior.

Chapter 4, Molecular weight and molecular weight distribution of polymers, by Dr. Han Peng, describes the definition and methods for determining the average molecular weight and molecular weight distribution of polymers.

Chapter 5, Molecular chain motion, by Dr. Yurong Yan, covers the bulk state, the glass transition, creep and stress relaxation, and rubber elasticity.

Chapter 6, Mechanical behavior of polymers, by Drs. Yurong Yan and Yonghua Zhang, describes the mechanical behavior and viscoelasticity of polymers.

Chapter 7, Electrical and optical properties of polymers, by Dr. Sa Liu, describes the conductivity, dielectric relaxation and optical properties of polymers.

Publication of this book is financially sponsored by the Foundation for the State Essential Course of the MOE and the Publication Fund of South China University of Technology Press. Authors are very grateful to Mr. Zhaoqiang Wu of the press for his kind help in the editorial work. Authors also express their sincere gratitude for the use of all the references.

Z. TONG

Guangzhou

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Chapter 1

STRUCTURE OF POLYMER CHAIN

1.1 Introduction

1.1.1 Introduction to Polymer Science

Polymer science was born in the great industrial laboratories of the world in the need to make and understand new kinds of plastics, rubbers, adhesives, fibers, and paints. Only much later did polymer science come to academic life. Perhaps because of its origins, polymer science tends to be more interdisciplinary than most, combining chemistry, chemical engineering, materials, and other fields as well.

Chemically, polymers are long-chain molecules of very high molecular weight, often measured in the hundreds of thousands. For this reason, the term “macromolecules” is frequently used when referring to polymeric term that goes back before the chemical structure of the long chains was understood.

The first polymers used were natural products, especially cotton, starch, proteins, and wool. Beginning earlier in the 20th century, synthetic polymers were made. The first polymers of importance, Bakelite and nylon, showed the tremendous possibilities of the new materials. However, the scientists of that day realized that they did not understand many of the relationship between searches that ensued forms the basis of physical polymer science.

This book develops the subject of physical polymer science, describing the interrelationships among polymer structure, morphology, and physical and mechanical behavior. Key aspects include molecular weight and molecular weight distribution, and the organization of the atoms down the polymer chain. Many polymers crystallize, and size, shapes, and organization of the crystallites depend on how the polymer was crystallized. Such effects as annealing are very important, as they have a profound influence on the final state of molecular organization.

Other polymers are amorphous, often because their chains are too irregular to permit regular packing. The onset of chain molecular motion heralds the glass transition and softening of the polymer from the glass (plastic) state to the rubber state. Mechanical behavior includes such basic aspects as modulus, stress relaxation, and elongation to break. Each of these is relatable to the polymer's basic molecular structure and history.

Some of the most significant polymers are listed below with their approximate commercialization dates:

1870	Celluloid (nitrocellulose)
1908	Bakelite (phenolic)
1919	Vinyl acetate
1927	Cellulose acetate
1928	Ureas
1931	Acrylics
1936	Polyvinyl chloride (PVC)
1938	Cellulose acetate butyrate
1938	Polyamides (nylon)
1938	Polystyrene
1939	Melamines
1939	Polyvinylidene chloride
1942	Polyethylene
1942	Polyesters
1943	Silicones
1943	Teflon
1947	Epoxy
1948	Acrylonitrile-butadiene-styrene (ABS)
1948	Polychlorotrifluoroethylene (Kel-F)
1953	Polyurethanes
1957	Polypropylene
1958	Acetals
1959	Polycarbonate
1964	Polyphenylene Oxide
1964	Polyimide
1965	Polysulfone
1965	Polymethyl pentene (TPX)
1969	Polybutylene terephthalate polyesters (PBT)
1973	Polybutylene

1. 1. 2 From Small Molecules to Big Molecules

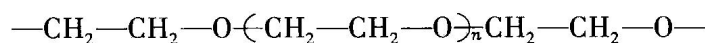
Polymers are macromolecules built up by the linking together of a large number of much smaller molecules. The small molecules which combine with each other to form polymer molecules are termed *monomers* and the reactions by which they combine are termed *polymerizations*. There may be hundreds, thousands, tens of thousands or more monomer molecules linked in a polymer chain. When one speaks of polymers, one is concerned with materials whose molecular weights may reach into the millions. Most of the polymers however, that one usually encounters either in laboratory or in practice will usually fall into the 5,000 ~ 200,000 molecular weight range.

All macromolecules, both organic and inorganic, consist of at least one chain of atoms boned together and running through the whole molecules. This chain forms so to speak, the backbone of the macromolecules. It can contain carbon-carbon bonds, carbon-oxygen bonds, carbon-nitrogen bonds, or non-

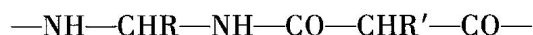
carbon-carbon bonds for examples :



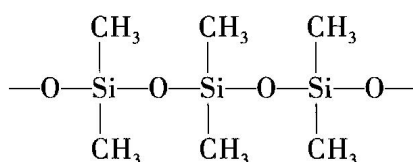
polyethylene



polyethylene oxide



polyamide (nylon)



poly(dimethylsiloxane)

As a simple example, consider the normal polyethylene, it has the general structure :



Where the number of $\text{—CH}_2\text{—}$ groups, n , is allowed to increase up to several thousand. The progression of their state and properties is shown in Table 1. 1.

Table 1. 1 States and Properties Alkane to Polyethylene Series

Number of Carbons In Chain	State and Properties of Materials	Use, Dependent on Chain Length
1 ~ 4	Simple gas	Bottle gas for cooking
5 ~ 11	Simple liquid	Gasoline
9 ~ 16	Medium-viscosity liquid	Kerosene
16 ~ 25	High-viscosity liquid	Oil and grease
25 ~ 50	Crystalline solid	Wax
50 ~ 1000	Half crystalline solid	Adhesive and coating
1000 ~ 5000	Tough plastic solid	Containers
$3 \times 10^5 \sim 6 \times 10^5$	Fiber	Fishnet

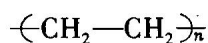
At room temperature, the first four members of the series are gases. Pentane boils at 36.1°C and is a low-viscosity liquid. As the molecular weight of the series increases, the viscosity of the members increases. Although commercial gasolines contain many branched-chain materials and aromatics as well as straight-chain alkanes, the viscosity of gasoline is markedly lower than that of kerosene, motor oil and

grease because of its lower average chain length.

These later materials are usually mixtures of several molecular species, although they are easily separable and identifiable. This point is important because most polymers are also "mixtures", that is, they have a molecular weight distribution. In high polymers, however, it becomes difficult to separate each other of the molecular species, and people talk molecular weight averages.

Compositions of normal alkanes averaging more than about 20 ~ 50 carbon atoms are crystalline at room temperature. These are simple solids known as wax. It must be emphasized that at up to 50 atoms the materials are far from being polymeric in the ordinary sense of the term.

The polymeric alkanes contain 1000 ~ 3000 carbon atoms and are known as polyethylene. Polyethylene has the chemical structure:



which originates from the structure of the monomer ethylene, $\text{CH}_2=\text{CH}_2$. In some places the structure is written:



or polyethylene. (then, $n' = 2n$.) The relationship of the latter structure to the alkane series is clearer. While true alkanes have $\text{CH}_3\text{—}$ as end groups, most polyethylene have initiator residues.

Even at a chain length of thousands of carbons, the melting point of polyethylene is still slightly molecular-weight-dependent, but most linear polyethylenes have melting temperature, T_M , near 140 °C. The approach to the theoretical asymptote of about 145 °C at infinite molecular weight is illustrated schematically in Figure 1.1.

The greatest differences between polyethylene and wax lie in their mechanical behavior, however. While wax is a brittle solid, polyethylene is a tough plastic.

Polyethylene is a tough plastic solid because its chains are long enough to connect individual stems together within a lamellar crystallite by chain folding (see Figure 1.2). The chains also wander between lamellae, connecting several of them together. These effects add strong covalent bond connections both within the lamellae and between them. On the other hand, only weak Van Der Waal forces hold the chains together in wax.

In addition, a certain portion of polyethylene is amorphous. The chains in this portion are rubber, imparting flexibility to the entire material. Wax is 100% crystalline, by difference.

The long chain length allows for entanglement (see Figure 1.3). The entanglement helps hold the whole material together under stress. In melt state, chain entanglement causes the viscosity to be raised very significantly also.

The long chains shown in Figure 1.3 also illustrate the coiling of polymer chains in the amorphous state, the conformation of amorphous chain in space being random coils; that directions of the chain por-

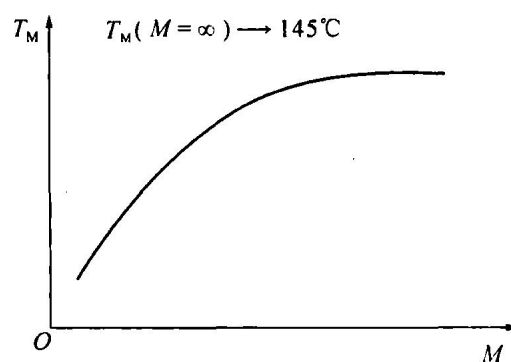


Figure 1.1 The molecular weight-melting temperature relationship for the alkane series.

tions are statistically determined.

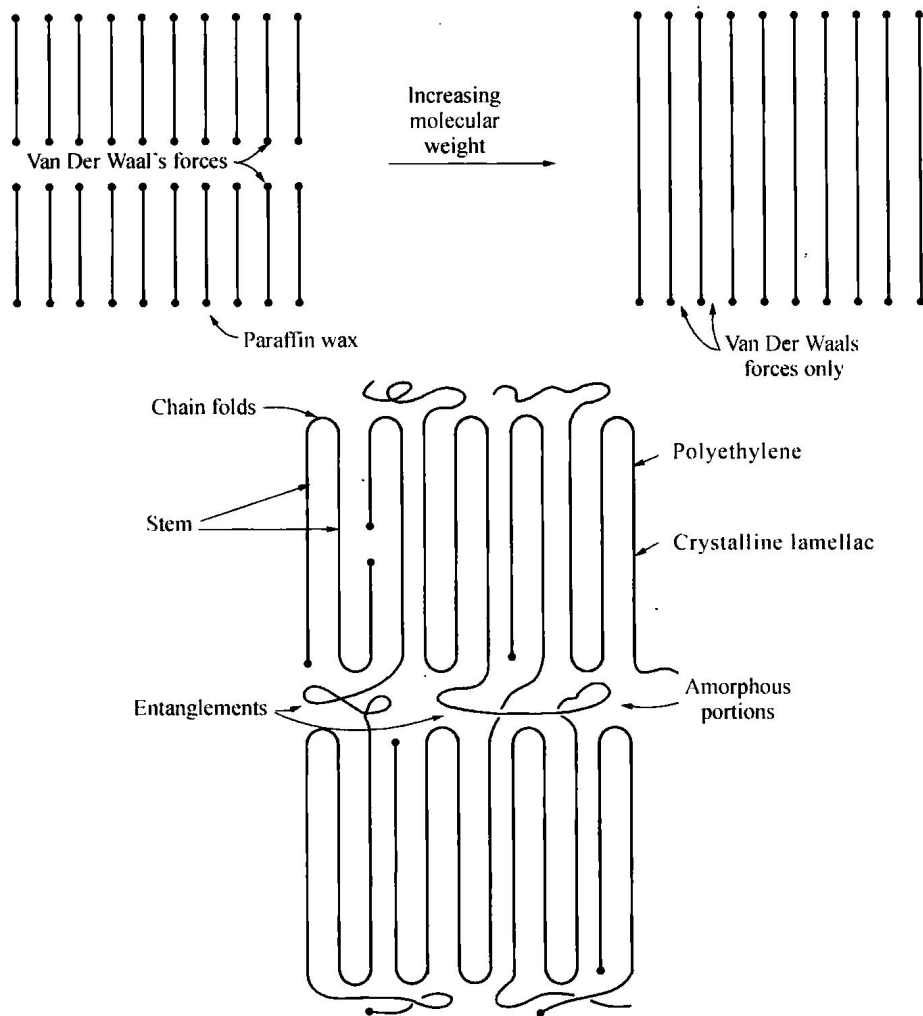


Figure 1.2 Comparison of wax and polyethylene structure and morphology.

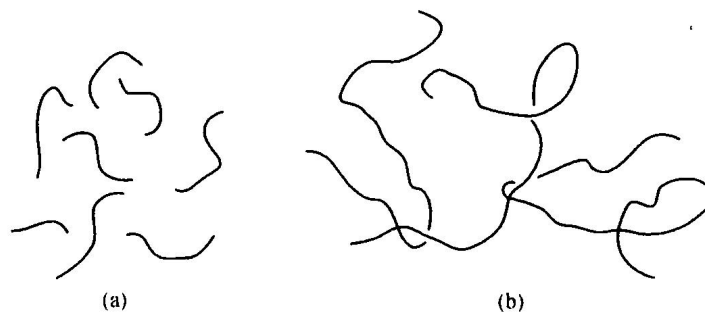


Figure 1.3 Entanglement of polymer chain.

1.1.3 Future Topics for Polymer Science

(1) According to the types of polymers, scientists improve old polymer products to provide some no-

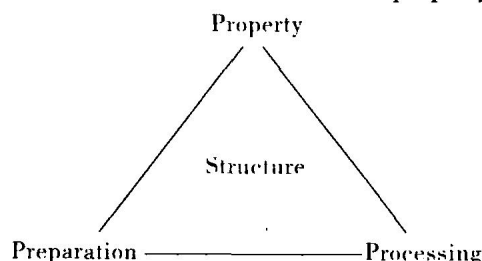
vel. properties. The study on polymer blending and co-polymerization has become an important field.

(2) According to polymer industry, it is necessary to develop new equipment, new technology to raise catalyst efficiency and to lower production cost.

(3) According to the properties of polymer material, it is necessary to study the polymers which possess high performances and can be used in special environments. For example, it is necessary to develop high-temperature resistance for space; low-temperature resistance, high modulus materials, polymer semi-conductivity and super-conductivity polymer materials and to raise weather-resistance property of normal polymer materials.

(4) According to biological science, scientists are developing medical polymers and biology polymers, such as biology film and structural materials for medical purposes.

(5) Based on the relationship of synthesis—structure—property, it is necessary to carry out the “macromolecular design” of polymer materials for different purposes.



1.1.4 Major Characteristics of Polymer Structure

The structures of macromolecules are very complex. It has the following characteristics compared with small molecules:

(1) A polymer is simply a very large molecule formed by joining many small molecules known as monomer. (It has the molecular weight about $10^3 \sim 10^5$). The repeating units are the same as the small molecules which can polymerized into uniform polymer (when repeating units are same) or polymerized into copolymer (when the repeating units are different). The products can be linear molecules, branch molecules and crosslinking network molecules.

(2) The backbone of macromolecules can rotate, bend and can be flexible. The shape of backbone can be changed because of the movement of the segments.

(3) The process of polymerization is random, therefore some molecules grow much large than others. Polymer is a mixture of different size molecules. Molecular size is expressed as average molecular weight.

(4) The interaction of macromolecules influences on the condensed state structure of polymer significantly. After crosslinking between molecules, the mechanical properties of polymer will be changed significantly. The crosslinked polymers do not melt on heating and do not dissolve in any solution.

(5) The integrate structure of polymer can be classified into crystalline state and non-crystalline state structure. It is needed to point out that the crystallinity of polymer is less than that of small molecule.

(6) The mixture structures of polymer have a significant influence on properties of polymer materials.

1.1.5 Contents of Polymer Structure

Based on different study units, polymer structure can be classified into two parties: The first party is the structure of polymer chain. It is to study the arrangement of atoms or group in a polymer chain, that is, the structure of internal macromolecule. The second party is the condensed structure of polymer. It is to study the structure of between macromolecules and macromolecules.