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Methodological Basis of Integrated Improvement of the Course "Chemistry of Polymers"

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ANNOTATION: Certain well-defined trends are discernible in the chemistry of polymers: (1) an improvement in mechanical properties and processing, especially with respect to the development of stiffer and tougher materials; (2) an increase in physical and chemical inertness, for example good ageing characteristics, stability against light, reduction of flammability and resistance against chemicals; and (3) the widening of the temperature range in which the material may be used, for example, polymers which may be used at high temperatures. New products are being introduced, and processes are being improved to obtain better and more desirable properties from a wide range of existing materials. The broad correlation of molecules, structure and properties which determines the end-use of the material is discussed. Particular attention is paid to heat-resistant and heat-stable polymers and fiber-reinforced plastics.

KEYWORDS: Particular attention, final mechanical and thermophysical, glass transition temperature, melt and solution viscosity

I. INTRODUCTION

The molecular weight of a polymer is one of the most important parameters because it controls, to a large extent, the final mechanical and thermophysical properties such as glass transition temperature, melt and solution viscosity and mechanical strength, to name only a few properties. For many applications, a high molecular weight is desired. However, polymers with high molecular weight can only be obtained by step-growth when three critical requirements are met: First, the two dysfunctional monomers (A-A & B-B) have to blend at perfect stoichiometric balance. In other words, the ratio of the concentrations of the reactive groups, $r = [A] / [B]$, has to be equal or very close to 1.0. In the case of a single A-B type functional monomer such as an amino acid or hydroxyl acid, this balance is automatically provided. Second, monomer impurities that can react and terminate step-growth have to be kept at an absolute minimum because any trace amount of a nonfunctional reactive compound will cap the growing polymer chains and thus reduce the molecular weight. However, stringent purification of the reactants is often difficult and adds cost to the final product. Third, the step-growth polymerization has to be driven close to completion, i.e. to a very high yield without any side reactions.

The molecular weight of a step-growth polymerization can be easily calculated if we assume that each functional group has the same probability of reacting with other groups regardless of the size of the oligomer and type of functionality. This assumption is called *Flory's equal reactivity principle*. According to this principle, the probability that a given reactive group has reacted at time t is equal to the fraction of monomers p that have undergone condensation which is called the extent of the reaction.¹ Then the average *degree of polymerization (DP)* is equal to the total number of bifunctional units initially added N_0 divided by the remaining number of molecules N :

$$DP = N_0 / N = N_0 / N_0(1 - p) = 1 / (1 - p)$$

Consider a reaction that can be driven to 90% completion which is a relative high yield and not easy to achieve in a bulk polymerization. Then the average degree of polymerization DP is $1 / (1 - 0.9) = 10$. However, a decamer is to



short to have useful mechanical properties. In general, the molecular weight has to be about two times the entanglement weight which equals about 15 to 100 repeat units, depending of the structure of the molecules.² For example, to obtain a $DP = 50$, polymerization has to be carried out to 98% completion, $DP = 1 / (1-0.98) = 50$. Therefore, a relative high conversion is required to attain a high degree of polymerization.

As has been pointed out by Flory and many other scientists, the true average degree of polymerization is often lower than the theoretical value. The main reasons for this discrepancy are threefold: a) some monomer may evaporate during polymerization, b) some monomer may undergo side reactions, and c) (trace amounts) of monofunctional reactive compounds could be present. This will lead to a stoichiometric imbalance of the number of reactive groups A and B and a depression of the average molecular weight.

The stoichiometric imbalance is defined by the ratio $r = N_{A,0} / N_{B,0}$, which cannot exceed unity by definition. Then the total number of reactive units is

$$(N_{A,0} + N_{B,0}) / 2 = N_{A,0} (1 + 1/r) / 2$$

where $N_{A,0}$ and $N_{B,0}$ are the total number of A and B groups initially present. The number of unreacted A and B groups after the reaction has reached extent p is

$$N_{A,0} = (1 - p) N_{A,0}, N_{B,0} = (1 - p r) N_{A,0} / r$$

and the total number of chain ends

$$(1 - p) N_{A,0} + (1 - p r) N_{A,0} / r = (1 - 2p + 1/r) N_{A,0}$$

which must equal twice the total number of molecules since each molecule has two terminal functional groups.

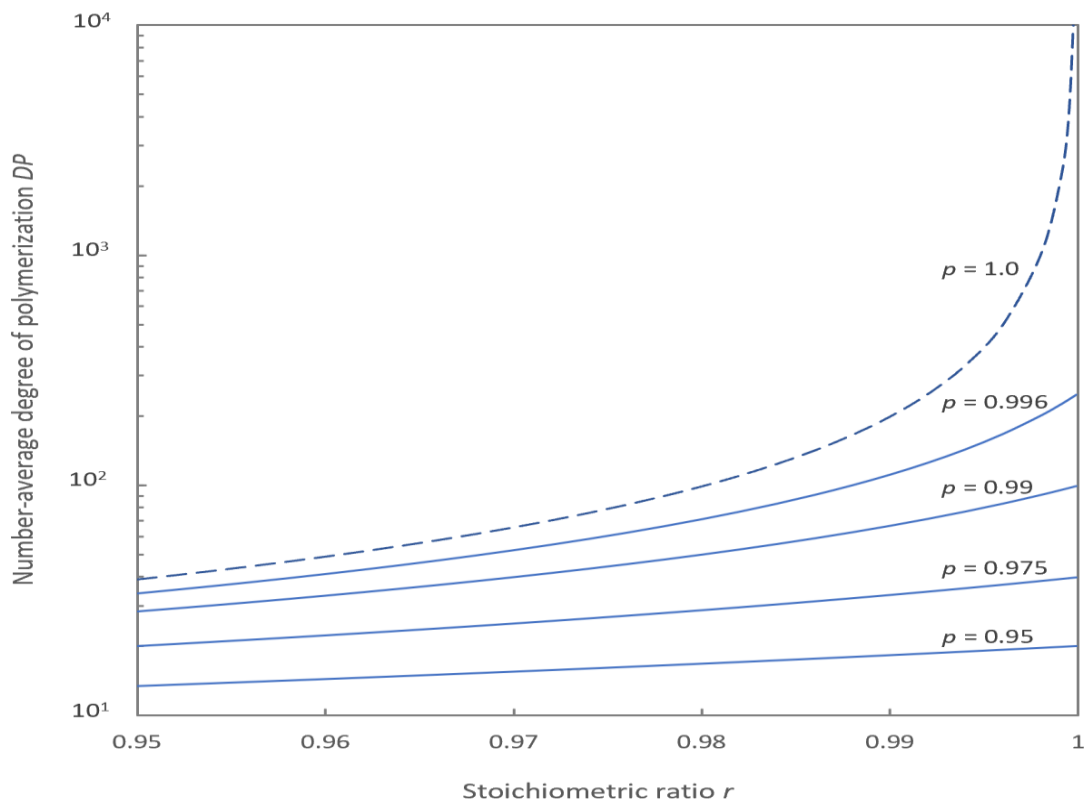
The number average degree of polymerization DP equals the number of repeat units divided by the number of chains:^{4,5}

$$DP = (1 + r) / (1 + r - 2pr)$$

At 100 % conversion ($p = 1$), this expression reduces to

$$DP = (1 + r) / (1 - r)$$

NUMBER-AVERAGE DEGREE OF POLYMERIZATION AS A FUNCTION OF THE STOICHIOMETRIC RATIO



The Figure above shows the effect of stoichiometric imbalance and conversion on the average degree of polymerization. The DP drops with increasing stoichiometric imbalance and decreasing conversion. For example, an



excess of 1% of B-B monomers reduces the degree to polymerization to about 100 units. Thus, a small loss of reactant through evaporation of A-A or B-B monomers or through side reactions can substantially limit the molecular weight of the final product. If, in addition, the polymerization is carried out to 99% completion instead of 100%, the degree of polymerization will reduce to 50 units. Thus, to achieve a high DP, the reaction has to be driven close to completion.

DEPRESSION OF MOLECULAR WEIGHT BY MONOFUNCTIONAL REACTANTS

The addition of a monofunctional reactant or impurity to an equimolar mixture of A-A and B-B monomers will depress the molecular weight through end-capping. As Flory has shown, the same equations can be applied to this situation if r is redefined as follows:⁵

$$r = N_{A,0} / (N_{B,0} + 2N_{B,1}) = N_{A,0} / (N_{A,0} + 2N_{B,1})$$

where $N_{B,1}$ is the initial number of monofunctional B monomers. The factor 2 arises here because a B molecule has the same quantitative effect in limiting the molecular weight as one excess B-B monomer. With this definition, the plots above describe the depression of the molecular weight by monofunctional monomers as well.

Branched polymers can be formed when some of the monomers have three or more functional groups per molecule. However, the addition of trifunctional monomers can also generate cross-linked polymers (polymer gel). In other words, a step polymerization involving trifunctional units can lead to the formation of insoluble cross-linked structures (gel fraction) embedded in linear and branched polymers (sol fraction) or to an infinite insoluble polymer network that is, to one gigantic molecule. The point in the polymerization at which the polymer starts forming crosslinked structures (insoluble gel) is called the *gel point*. According to Flory, an infinite network becomes possible when at least one of the functional groups of the $f-1$ branches has reacted with a functional group of another branch. The probability α_c of this occurring is 1 in $f-1$ or,⁵⁻⁷

$$\alpha_c = 1 / (f - 1)$$

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