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## Dielectric Relaxation of Oxide Polymers in Dilute Solution\*

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Received August 29, 1983

**ABSTRACT:** Dielectric relaxation measurements were made on dilute solutions of poly(propylene oxides) in benzene over a wide frequency range from 10 MHz to 10 GHz using time-domain reflectometry. Two relaxation processes were found. The lower frequency process, depending strongly on the molecular weight, is explained completely by the normal mode theory of Rouse and Zimm. Another process, independent of molecular weight, is caused by an elementary process in the backbone motion. The magnitude of the relaxation time is explained satisfactorily by Kramers' rate constant theory at the low-friction limit, if the elementary process is assumed to be a rotational transition of a chain bond. The relaxation process observed in poly(ethylene oxides) in benzene is also explained by Kramers' theory. The effect of chemical structure of oxide polymers on the higher frequency relaxation process is discussed.

### 1. Introduction

Three kinds of dielectric relaxation processes have been found in solutions of flexible polar polymers. If a polymer has dipole components parallel to its chain contour, a rotational diffusion of the polymer molecule gives rise to a relaxation process depending on molecular weight in a relatively low frequency region.<sup>1</sup> The relaxation time is explained quite satisfactorily by the normal mode theory of Rouse and Zimm.<sup>2,3</sup> The theory predicts relaxation time as

$$\tau_{\text{rot}} = A_1 M[\eta]\eta_0/RT \quad (1)$$

where  $M$  is the molecular weight,  $[\eta]$  the intrinsic viscosity,  $\eta_0$  the solvent viscosity,  $R$  the gas constant, and  $T$  the absolute temperature. The factor  $A_1$  takes a value of 1.21 for the free-draining case and 0.85 for the nondraining case. Poly( $\epsilon$ -caprolactone) (PCL) and poly(olefin sulfones) in solution exhibit this relaxation process.<sup>4-6</sup>

Generally, polar polymers have dipole components perpendicular to the chain contour and show a relaxation process independent of molecular weight which is observed in a relatively high frequency region.<sup>7-9</sup> Chain motions associated with conformational transitions in the chain backbone give rise to this relaxation. Recent study of methyl methacrylate-methyl acrylate copolymers in dilute solution has indicated that the elementary process in the chain motion is essentially a bond rotation which occurs independently between isomeric transition states.<sup>10</sup>

Poly(*p*-chlorostyrene) is a typical polymer having a dipole component of this type. The relaxation peak was found at about 30 MHz in solution.<sup>7-9</sup> The relaxation time is expressed experimentally by<sup>7</sup>

$$\tau_{\text{back}} = A_2 \eta_0 \exp(\Delta H_A/RT) \quad (2)$$

This expression is explained quite satisfactorily by Kramers' rate constant theory at the high-friction limit.<sup>11,12</sup> The value of  $\Delta H_A$  represents the potential barrier height for bond rotation. If correlated motions are assumed to exist among several neighboring bonds, the magnitude of the factor  $A_2$  is explained by Kramers' theory.<sup>7</sup>

If the relaxation peak is found at a frequency lower than 100 MHz, the same argument can hold well for other polar polymers such as vinyl polymers<sup>7,8</sup> and poly(alkyl methacrylates)<sup>13</sup> in dilute solution.

Recently, a relaxation caused by internal rotations of side groups has been found in poly(methyl vinyl ketone) in benzene.<sup>14</sup> Rotation of acetyl side groups gives a relaxation peak at 1 GHz. Relaxation of this kind in the solid state is observed in a number of polymers. This result indicates that relaxation caused by side-group rotation exists even in a dilute polymer solution.

If a polymer contains oxygen atoms in the chain backbone, as in the polyethers, a dielectric relaxation peak occurs at a very high frequency. Poly(ethylene oxide) (PEO) exhibits a peak at about 10 GHz in benzene,<sup>15</sup> and PCL has one at 2 GHz in dioxane.<sup>4,16</sup> Poly(styrene oxide) (PSO) shows a peak at 450 MHz in benzene, which is about 5 times higher than that of polystyrene.<sup>14</sup>

Most likely, the backbone motions in these polymers also cause the relaxation observed. Therefore, it is thought to be quite important in interpreting chain dynamics to examine whether Kramers' rate constant at the high-friction limit can or cannot be applied to the relaxation times of these polymers as well as of vinyl polymers and to determine why the relaxation time is very small in comparison with other polar polymers.

In this work poly(propylene oxides) with different molecular weights were used as the polymer having oxygen atoms in the chain backbone, and dielectric relaxation

\* This paper is dedicated, with great pleasure and appreciation, to Professor Walter H. Stockmayer on the occasion of his 70th birthday celebration.

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