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Water in Renewable Polymers: Nonequilibrium Thermodynamics[1]

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The design of polymers derived from sustainable resources (renewable polymers) as replacements to nonrenewable plastics for various applications will require an accurate assessment and fundamental understanding of the dynamics water sorption in glassy polymers. In this work, water sorption and diffusion in a number of glassy polymers (including the renewable polymer poly(lactide)) were measured using gravimetric and spectroscopic techniques. Non-Fickian diffusion was observed in all polymers studied, which was indicated by rapid, initial water uptake (driven by a concentration gradient), followed by continuous, gradual uptake of water at later experimental times (driven by slow polymer relaxation). Additionally, water sorption in these glassy polymers was predicted using two nonequilibrium thermodynamic models, where excellent agreement between the model prediction and experimental data was achieved for both models. Furthermore, contrasting physical pictures of water clustering were obtained between the Zimm-Lundberg theory and direct measurements.

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