Enzymatic degradation of semi-dilute polymer solutions: coupling between enzyme mobility and activity

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Biodegradable polymers (synthetic or natural) can be degraded by the action of living organisms. In most cases, biodegradation occurs through enzyme-catalysed depolymerisation, where enzymes hydrolyse bonds along the polymer chains. Lignocellulosic biomass (LC) is one of the most abundant renewable polymers that, when degraded, represents a tremendous source of interesting products. However, LC is difficult to degrade because plant cell wall polysaccharides are structurally and chemically complex molecules, forming a heterogeneous network of varying density and porosity.

During industrial degradation, enzymes are confronted with LC substrates (e.g. wheat straws) that are insoluble and partially hydrated. They penetrate and propagate in the material, while at the same time changing its properties through hydrolysis. Although fascinating, little is known about the coupling between enzyme activity and mobility in the context of dense polymeric systems. In extremely tight polymer networks, the coupling is obvious: enzymes cannot just enter the polymer mesh and consequently move by 'eroding' the polymer surface [1]. In the more general case, enzymes can enter the network, but their activity is affected by the restricted diffusion in the polymer mesh [2]. Nevertheless, studies only consider the impact of the concentration of the polymer, neglecting its structural and dynamical evolution with hydrolysis time [3]; two characteristics that must have an effect on enzyme propagation in the material.

Here we explore the coupling between enzyme mobility and activity by using a model polymer, arabinoxylan, extracted from wheat LC and a fluorescently labelled enzyme. We prepare semi-dilute polymer solutions that are concentrated at different mesh sizes. Then, the radial diffusion of the enzymes is observed using fluorescent microscopy. We present the results obtained with catalytically active and inactive enzymes. To complement these results, we present ex-situ experiments aiming at characterizing the polymer during the degradation (viscometry, SEC-MALS).

Keywords: Enzymatic mobility and activity, semi-dilute polymer solution, fluorescence microscopy, degradation of lignocellulosic biomass

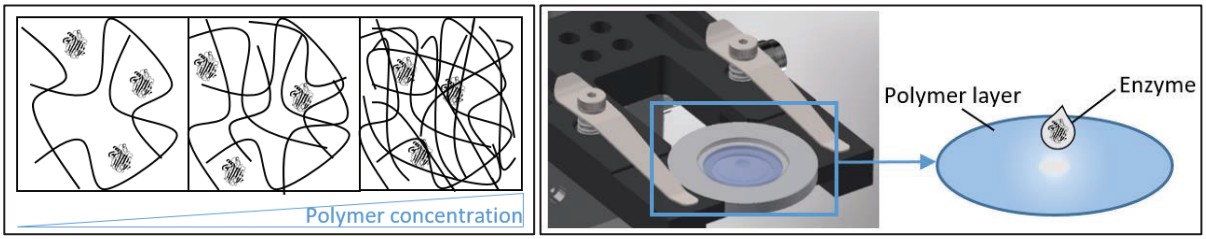

***Figure 1**. Left-hand side: The evolution of the mesh size and confinement with increasing polymer concentration. Right-hand side: The circular microscopy chamber containing the polymer solution. The enzyme is deposited in the middle and the chamber is sealed. The fluorescent front induced by the radial diffusion of the enzymes is then followed over time.*

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