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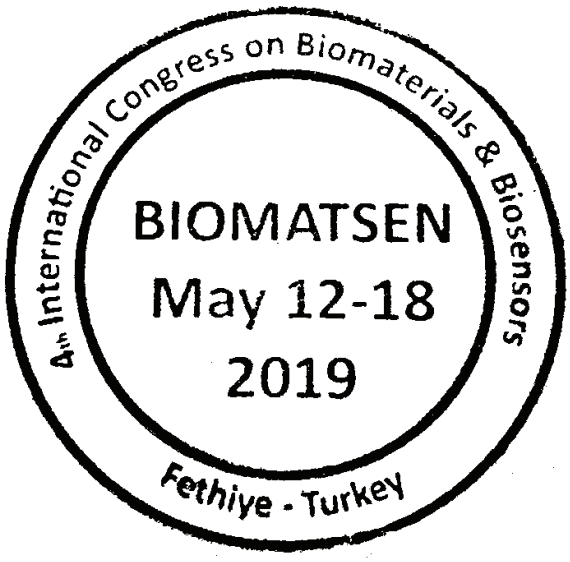

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Editorial Board

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Functionalization of Aliphatic Polyester Fibers Formed by Electrospinning - The Effect of Crystallinity on Aminolysis

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Abstract: The major drawback of aliphatic polyesters in tissue engineering applications is related to their poor hydrophilicity and the lack of reactive functional groups limiting interactions with cells. Various methods of surface modification followed by immobilization on the surface of bioactive molecules have been developed so far. One of the extensively studied methods of polyester surface modification is aminolysis which is characterized by attacking on the backbone ester bonds by diamine molecules at the interface between the diamine solution and polyester material, endowing the polyester surface with amino and hydroxyl groups which are the basis for subsequent conjugation of bioactive molecules. It is known that aminolysis is selective with respect to the state of supermolecular organization, being faster in amorphous than in crystalline phase. The objective of our work is to analyse the effect of crystallinity on aminolysis using various aliphatic polyesters. Three different aliphatic polyesters - poly(□-caprolactone) (PCL), poly(L-lactic acid) (PLLA), and copolymer PLA-PCL (70/30) (PLCL) in a form of nanofibers and films were investigated. Nanofibers were formed by solution electrospinning using hexafluoroisopropanol as a solvent while films were formed via solution casting method. Aminolysis was carried out using 6% w/v ethylene diamine in isopropanol solution at 30°C, at 5 and 15 minutes. Effectiveness of aminolysis was evaluated using ninhydrin test. Complementary information was obtained from ATR-FTIR and mechanical tensile tests. Crystallinity was determined from wide-angle X-ray scattering experiments (WAXS). The results of ninhydrin tests show that for PLLA and PLCL fibers the progress of aminolysis increases with time while for PCL fibers the aminolysis does not occur. Contrary to the fibers, for all investigated polymers in the form of film, including PCL, the ninhydrin test results indicates effective aminolysis. The results of ninhydrin tests are supported by FTIR results and by the stress and strain at break indicating molecular degradation for PLLA and PLCL which accompanies aminolysis. From WAXS analysis it is evident that crystallinity for PCL is higher than for PLCL, being 0,66 and 0,53, respectively, while PLA is practically amorphous (crystallinity 0,03). Similar relation between crystallinity was observed for films. It should be aware that the measured WAXS crystallinity is a bulk crystallinity and it is known from the conventional high speed spinning that the crystallinity at the fiber surface can be much higher than in the core due to faster solvent evaporation resulting in a radial gradient of polymer concentration [1]. There is no data on radial gradient structure in nanofibers so far, but there is no reason to expect that such gradient doesn't exist on smaller scale in electrospinning. Considering the effect of higher crystallinity at the surface of electrospun fibers, we

explain the problem with aminolysis of PCL fibers by a highly crystalline barrier formed at the surface, which slows down the diffusion of diamines and reduces the exposure of ester bonds. In the case of films, the structure gradient perpendicular to the surface is lower because of slower rate of solvent evaporation than for electrospun fibers.

Keywords: Nanofibers, polyesters, surface functionalization, aminolysis, structure

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