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Biaxial stretching of poly(L-lactide) tubes for improvement of mechanical properties

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INTRODUCTION

Poly(L-lactide) (PLLA) is a semi-crystalline bioabsorbable polymer, which has been widely investigated for medical devices despite its inferior stiffness and strength compared with conventional materials such as metal alloys¹. Due to the semi-crystallinity behaviour of the polymer, strain-induced crystallinity is expected to improve such mechanical properties by orientation of the crystals as seen in biaxial films^{2,3}. The objective was to investigate improvement of mechanical properties and the changes in crystallinity, crystal size and orientation in simultaneous biaxial strained PLLA tubes.

EXPERIMENTAL METHODS

PLLA (2003D) was extruded into tubes (OD 3.4mm). The tubes were heated to 74°C and longitudinally strained while applying internal pressure, initiating a tube expansion in the heated zone, resulting in a simultaneous biaxial strain.

The degree of strain in each direction is noted as the ratio between the longitudinal and radial strain (LxR). Mechanical testing was conducted to evaluate material properties and the effect of orientation. Orientation and crystal size was determined by X-ray diffraction (XRD), while total crystallinity (X_c) was determined by differential scanning calorimetry (DSC).

RESULTS AND DISCUSSION

Tensile testing of specimens in the circumferential direction show the typical strain hardening behaviour of



Fig1: Elastic modulus and yield stress for PLLA tubes with different LxR ratio obtained from tensile testing in the circumferential and longitudinal direction.

a semi crystalline polymer, but this is not seen for specimens tested in the longitudinal direction.

The elastic moduli (Fig.1) in the circumferential direction were superior to the modulus in the longitudinal direction at all times, due to the higher degree of strain in the given direction. Also the modulus is improved with degree of total area expansion, whereas the LxR ratio does not determine the

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properties. The yield stress is less affected by the total area expansion (T_{area}).

XRD images in Fig 2 show that crystal orientation is predominant longitudinal when only strained in the longitudinal direction (LxR =1.0x0). The intensity plot in Fig. 2 show that once the tube is expanded, the peak in the 110/200 plane is no longer at perpendicular orientation to the longitudinal direction, but parallel. Despite a larger degree of T_{area} or LxR ratio, the orientation did not change.

 X_c does not increase with the degree of T_{area} or LxR ratio, and the improved stiffness of the tubes is therefore not explained by neither crystal orientation nor an increase in strain-induced crystal formation.

XRD measurement also showed that crystals initially formed during uniaxial strain have a large crystal size circumferentially (183Å) and smaller axially (62Å), but this relation is flipped if radial expansion occurs simultaneously with 65Å radially and 134Å axially. The crystal size does not change significantly in the circumferential direction (65-70Å) with expansion or as function of LxR ratio, whereas the crystal size in the longitudinal direction decrease from 160Å to 137Å.



Fig 2: Intensity plot obtained from XRD of tubes with a different L/R ratio.

CONCLUSION

By biaxial stretching PLLA tubes improvement of stiffness and strength was obtained in both directions. Longitudinal stretching leads to crystal orientation in that given direction. Crystal orientation gained from this strain is reduced by degree of radial strain and altering the crystal alignment longitudinally. X_c does not increase with degree of radial strain and the improved properties are therefore not related to the crystal orientation nor a higher degree of crystallinity, but possibly stretching of the amorphous regions.

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