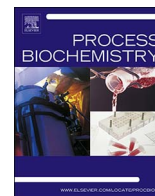




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# Process Biochemistry

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## Foreword: Advances in “Polymer/Fibre Biotechnology”



Biotechnology has been applied as a valuable tool to produce useful bio-based products from non-petrochemical renewable resources. Biologically produced polymers present advantages related with the biodegradability, performance, cheapness of substrate and defined structural variability. Beyond that, enzymes, the most proficient catalysts, continue to offer the most competitive processes compared with the chemical ones. Advances in protein engineering technology and the environmental and economic requirements contribute for the continuous search of acceptable biotechnological solutions for the areas, including, polymer and textile, medical, pharmaceutical, among others. New trends and strategies have been highlighted in recent studies leading to significant advances in enzyme biocatalysis.

The 9<sup>th</sup> International Conference on Fiber and Polymer Biotechnology 2016 (IFPB 2016) was held in September 2016 in Osaka Seikei College, Osaka, Japan. IFPB16 was a joint conference with Textile Biotechnology Symposia 2016 (TBS) and the 40<sup>th</sup> Annual Meeting of Advanced Fiber Materials Research Committee (AFMc). Previous events of IFPB had taken place at: University of Minho, Portugal (INTB 2000); University of Georgia, USA (INTB 2002); Graz University of Technology, Austria (INTB 2004); Korea Institute of Industrial Technology, Korea (INTB 2006); Jiangnan University, Wuxi, China (INTB 2007); University of Gent, Belgium (IPTB 2009), Stazione Sperimentale per la Seta, Milan, Italy (IPTB 2011) and University of Minho, Portugal (IPFB 2014).

The main topics covered by IFTB 2016 comprised industrial enzymes, natural and bio-based polymers and fibers, biofunctionalization of synthetic materials, sustainable processes, smart materials through bio-catalysis and nano/bio-materials and applications. Richard Gross from the Department of Chemistry and Chemical Biology, Rensselaer Polytechnic Institute, USA, was invited as keynote speaker. His remarkable lecture about “Engineered Cutinases for Textile Polymer Surface Modification and Recycling” provided an excellent opportunity to learn more about the potentialities of cutinases as efficient catalysts. Gross highlighted his recent work related with cutinase redesign, its biophysical and biochemical characterization and ability to recycle polyethylene terephthalate (PET) and cellulose acetate (CA). Thomas Rosenau from University of Natural Resources and Life Sciences in Austria and Kanji Kajiwara from the Faculty of Textile Science and Technology of Shinshu University in Japan were the invited speakers and enriched the conference with their new insights about cellulosic derivatives and fibers.

This conference was a worthy occasion to bring together researchers, scientists, engineers, and scholar students to exchange and share their experiences, new ideas, and research results about all aspects of Fiber Processing and Biotechnology, and discuss the practical challenges encountered and the solutions adopted. From the overall papers presented, some of them were selected for this special collection. Among the high number of peer-reviewed articles, sixteen have been accepted by *Process Biochemistry*. This special section is organized by categories divided as: Fermentation & Bioreactor Engineering, Enzyme Engineering & Proteins and Environmental Biotechnology.

Cellobiose dehydrogenase has been emerging as an interesting enzyme for many biomedical applications namely as a strong antimicrobial and antibiofilm agent or as a biocatalyst for the synthesis of many bioactive molecules of medical interest. A summary about the recent advances in the application of Cellobiose dehydrogenase (CDH) as a novel antimicrobial and antibiofilm agent, as biosensor, and for the production of many biomedically important biomolecules is reviewed [1]. The authors report the successful development of systems containing this enzyme which reduced the problems associated with implant- or biomaterial-acquired infections and the continuous removal of reactive species produced in chronic wounds, which are partly to blame for the inability of chronic wounds to heal [1].

Articles contemplating the enzymatic-assisted functionalization of fibers/polymers are well documented in this special collection. Facing the current problems related with the increase of plastic wastes, Gamerith *et al.* investigated the ability of cutinase from *Thermobifida cellulositytica* (The Cut1) to hydrolyse poly (ethylene terephthalate) (PET) moieties in different polymer blends without prior separation [2]. The tendency to use polymer blends for plastics production increases the need of enzymes for selective depolymerisation. The authors report that when incubated with PET blended with polyethylene (PE) or polyamide (PA) from packaging and bottles without prior separation, The Cut1 selectively hydrolyzed the PET moieties releasing terephthalic acid (TPA) and mono(2-hydroxyethyl) terephthalate (MHET). Polymer blends were hydrolyzed in an up to 9 times higher extent compared to higher crystalline pure PET.

The concerns about PET depolymerisation and recycling are also highlighted by Carniel *et al.* [3]. In their study, a screening of 10 commercial enzymes (Lipases from *porcine pancreas* (PPL), *C. rugosa* (CrL), *M. miehei* (MmL), *P. cepacia* (PcL), *P. fluorescens* (Pfl), *R. miehei* (RmL), *R. niveus* (RnL) and *T. lanuginosus* (TlL), Liquid preparations of *C. antarctica* lipase B (Lipozyme©CALB) and *Humicola insolens* cutinase) using bis-(hydroxyethyl) terephthalate (BHET) as model substrate was performed. Their achievements revealed *Candida antarctica* lipase B (CALB) and *Humicola insolens* cutinase (HiC) as potential biocatalysts for PET depolymerization [3].

Surface modifications of cotton with long hydrophobic molecules are expected to improve the textile hand perception of consumers and the crease recovery, being an added-value at the moment of purchase. Despite its hydrophilicity, cotton cellulose has superb merits as a substrate for the

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production of hydrophobic materials. The hydrophobic modification of cotton cellulose has involved surface modification using hydrophobic compounds, such as fluorocarbons, silicones and hydrocarbons. However, these technologies are constricted by their sustainability and environmental concerns and cost. An ecologically greener alternative to current practice is presented by Zhao and collaborators [4], who carried out a lipase-catalyzed solvent-free synthesis of poly (ethylene glutarate) monomers and oligomers to coat the surface of cotton fabrics for the first time.

Synthetic fibres play an important part in textile industry as they are used in numerous applications, where cotton fibers cannot meet up due to their properties. They present several advantages such as strength, and elasticity, less wrinkles, lightness and fast drying, making them highly suitable for use in sportswear, however, their high hydrophobicity causes a problem that seeks attention, since it hinders the process of dyeing, permeability and sweat evaporation. Therefore, the increase of hydrophilicity of synthetic fibers is crucial in order to improve certain properties and expand further applications. The enzymatic surface modification of polyamide (PA) 6.6 fabric was studied with the use of the commercial protease Alcalase 2.4 L at optimal conditions by Kanelli et al. [5]. The enzymatic process increased the hydrophilicity with 2.7-fold increased water absorbency and 1.24-fold enhanced color strength of PA textiles, while maintaining the thermal and mechanical properties of the bulk synthetic material.

In order to improve the potential value of starch and widen its utility, native starch has been modified using physical, chemical, and other methods. In this special section the preparation of starch-g-poly(butyl acrylate) (starch-g-PBA) copolymers using horseradish peroxidase (HRP), in the presence of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and acetylacetone (Acac) is described by Wang et al. [6]. The grafting of PBA onto the molecules of starch, using HRP catalyst, was confirmed by FT-IR, <sup>1</sup>H NMR, and elemental analysis. Moreover, the FT-IR results demonstrated that gelatinization and ethanol favoured graft copolymerization of starch with BA. Meanwhile, the viscosity of starch paste increased, and the grafted starch paste showed more elasticity after graft modification. This environmentally friendly process provides an attractive alternative for the preparation of modified starch for industrial applications [6].

The surface of cotton substrates served as support for the deposition of nanoparticles containing vitamin E, leading to the development of new cosmetotextiles by the group of Cavaco-Paulo [7]. In this study, cotton fabrics were coated with protein-based nanoparticles containing vitamin E ( $\alpha$ -tocopherol) by the pad-cure method. Samples coated with nanoparticles containing the highest amount of encapsulated vitamin E (20% of the oil phase) showed the highest antioxidant activity. A methodology for nanoparticles release from the coated surfaces and their transfer to other substrates was demonstrated by the simple crock meter rubbing in the presence of sweat and protease. A high amount of material can be transferred and released to other substrates, such as textiles and skin, through the synergistic effect of sweat/protease and abrasion.

Enzymatic-assisted reactions were explored as means to increase new functionalities, like drug release, to polymeric materials. Improved performance of chitosan (CS) films was achieved by grafting with the antimicrobial catechin (CA) via *in-situ* laccase-assisted oxidation [8]. The CS films prepared by enzymatic grafting presented a high antioxidant activity compared to native CS films. Kim and co-workers [8] also highlight that the type of molecules loaded into the films, the preparation manner of films and the release medium are decisive parameters affecting the release rate. Under this scope, a work related with enzyme-catalysed functionalization of poly(L-lactic acid) for drug delivery applications is also presented by Pellis et al. [9]. They report a new poly(lactic acid) (PLA)-based drug delivery system in which the cationic chemotherapeutic drug doxorubicin was adsorbed via ionic interactions.

The hydrolysis of long-chain triglycerides such as olive oil and the synthesis reactions such as biodiesel production, lipid modification and production of different kind of drugs such as prostaglandins and non-steroidal anti-inflammatories make it attractive to improve and test the catalytic properties of lipases. For this, Rivera et al. [10] successfully cloned CpLip1 lipase from *Carica papaya* into pGAPZ\_B plasmid and functionally expressed for the first time using *P. pastoris* as expression system.

The modification and functionalization of wood surfaces for increasing the hydrophobicity have been also a subject of research. A new approach for wood functionalization is described by the group of Guebitz [11]. They mechanistically investigate for the first time chemo-enzymatic and enzymatic coupling of fatty acids onto lignin model substrates. They demonstrate the ability of laccase to mediate hydrophobization of wood veneers.

Different works presented in this special issue are good examples of the recent developments in medical and pharmacological areas. The surface of urinary catheters composed of polydimethylsiloxane (PDMS) was functionalized with antibacterial aminocellulose nanospheres (ACNSs) using the epoxy/amine grafting chemistry [12]. The silicone surface decorated with these NSs demonstrated efficient inhibition of *E. coli* biofilms, reducing the total biomass when compared with pristine silicone material. As the authors established, the functionalization of silicone-based materials with ACNSs are promising potential platforms for the prevention of biofilm-associated infections caused by *E. coli*. The role of cyclodextrines (CD) as inclusion complexes was evaluated for fibromyalgia treatment by complexation with a steroidal acetylated-sapogenin, hecogenin acetate (HA) [13]. The aim was to improve the anti-hyperalgesic effect of HA in a non-inflammatory animal model of musculoskeletal pain (considered to be a suitable animal model for FM), evaluating its effectiveness by examining its effect on substance P (SP) in the spinal cord, and its influence on the opioid system. Their findings suggest that CD can improve the anti-hyperalgesic effect of HA in an animal model of musculoskeletal pain.

A review article presenting a summary of the progresses on the surface engineering of naturally produced particles is also included [14]. The concept underlying this surface engineering as well as the fundamentals of the synthesis and recovery of surface-engineered particles are addressed. The authors highlight the surface engineering aspects that will facilitate further development of these nanoparticles for future applications.

The exploitation of polymers production was assessed and described in the last two papers of this special section. Blunt et al. optimized the conditions of oxidative synthesis of medium chain length polyhydroxyalkanoate from fatty acids [15]. Yim et al. developed a new type of eco-friendly and sustainable fabric by using tea as nitrogen source for bacterial cellulose production [16].

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