



Review

The chemomechanical properties of microbial polyhydroxyalkanoates



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ABSTRACT

Microbially produced polyhydroxyalkanoates (PHAs) are fully biodegradable biopolyesters that have attracted much attention recently as alternative polymeric materials that can be produced from biorenewable and biowaste resources. The properties of these biological polymers are affected by the same fundamental principles as those of fossil-fuel derived polyolefins, with a broad range of compositions available based on the incorporation of different monomers into the PHA polymer structure, and with this broad range tailoring subsequent properties. This review comprehensively covers current understanding with respect to PHA biosynthesis and crystallinity, and the effect of composition, microstructure and supramacromolecular structures on chemomechanical properties. While polymer composition and microstructure are shown to affect these properties, the review also finds that a key driver for determining polymer performance properties is compositional distribution. From this review it follows that PHA–PHA blend compositions are industrially important, and the performance properties of such blends are discussed. A particular need is identified for further research into the effect of chemical compositional distribution on macromolecular structure and end-use properties, advanced modeling of the PHA accumulation process and chain growth kinetics for better process control.

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1. Introduction

Bacterial polyhydroxyalkanoates (PHAs) are a unique family of polymers that act as a carbon/energy store for more than 300 species of Gram-positive and Gram-negative bacteria as well as a wide range of archaea [1]. Synthesized intracellularly as insoluble cytoplasmic inclusions in the presence of excess carbon when other essential nutrients such as oxygen, phosphorous or nitrogen are limited, these polymeric materials are able to be stored at high concentrations within the cell since they do not substantially alter its osmotic state [2]. The resulting polymers are piezoelectric and perfectly isotactic/optically active (having only the *R*-configuration). They are hydrophobic, water-insoluble, inert and indefinitely stable in air, and are also thermoplastic and/or elastomeric, non-toxic and

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Nomenclature

2D	two-dimensional
3D	three-dimensional
¹³ C	carbon-13
3HB	3-hydroxybutyrate
3HD	3-hydroxydecanoate
3HDD	3-hydroxydodecanoate
3HHx	3-hydroxyhexanoate
3H2MB	3-hydroxy-2-methylbutyrate
3H2MV	3-hydroxy-2-methylvalerate
3H4MV	3-hydroxy-4-methylvalerate
3HO	3-hydroxyoctanoate
3HP	3-hydroxypropionate
3HPE	3-hydroxy-4-pentenoate
3HTD	3-hydroxytetradecanoate
3HV	3-hydroxyvalerate
4HB	4-hydroxybutyrate
ACP	acyl carrier protein
Ae	aerobic
ADF	aerodynamic feeding
AFM	atomic force microscopy
An	anaerobic
BOD	biochemical oxygen demand
CCD	chemical compositional distribution
CH ₂	methylene group
CoA	coenzyme A
COD	chemical oxygen demand
Da	Dalton
DMTA	dynamic mechanical thermal analysis
DO	dissolved oxygen
DOC	dissolved organic carbon concentration
DSC	differential scanning calorimetry
DP	degree of polymerization
<i>E_a</i>	activation energy
EBPR	enhanced biological phosphorus removal
ESI-MS	electrospray ionization multistage mass spectrometry
<i>F_{BB}</i>	fraction of the BB diad sequence in a P(3HB-co-3HV) copolymer
<i>F_{BV}</i>	fraction of the BV diad sequence in a P(3HB-co-3HV) copolymer
<i>F_{VB}</i>	fraction of the VB diad sequence in a P(3HB-co-3HV) copolymer
<i>F_{VV}</i>	fraction of the VV diad sequence in a P(3HB-co-3HV) copolymer
FAB-MS	fast atom bombardment mass spectroscopy
FT-IR	Fourier-transform infrared
g	gram
G	growth rate of spherulites
GAOs	glycogen accumulating organisms
GPa	gigapascals
GPC	gel permeation chromatography
ΔH_f	heat of fusion of the melting peak
$\Delta H_{f100\%}$	heat of fusion of a 100% crystalline material
HB	hydroxybutyrate
HDPE	high density polyethylene
HV	hydroxyvalerate
<i>k_A</i>	crystallization rate constant

kDa	kilodalton
<i>l_c</i>	lamellar core distance
<i>L_p</i>	long period distance
<i>M_n</i>	number-average molecular weight
<i>M_w</i>	weight-average molecular weight
mcl	medium chain length
MDa	megadalton
MPa	megapascals
μ m	micrometer
<i>n_A</i>	Avrami index
NADH	nicotinamide adenine dinucleotide (reduced form)
NADPH	nicotinamide adenine dinucleotide phosphate (reduced form)
nm	nanometer
NMR	nuclear magnetic resonance
P(3HB)	poly(3-hydroxybutyrate)
P(3HB-co-3HV)	poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
P(3HB-co-4HB)	poly(3-hydroxybutyrate-co-4-hydroxybutyrate)
P(3HB-co-3HHx)	poly(3-hydroxybutyrate-co-3-hydroxyhexanoate)
PAOs	polyphosphate accumulating organisms
PDI	polydispersity index
PEG	polyethylene glycol
PHA	polyhydroxyalkanoate
PHO	polyhydroxyoctanoate
<i>P_n</i>	number average degree of polymerization
R	R enantiomeric form
RNA	ribonucleic acid
SBR	sequencing batch reactor
scl	short chain length
SDS-PAGE	sodium dodecyl sulfate polyacrylamide gel electrophoresis
<i>t</i>	time
<i>T_c</i>	crystallization temperature
<i>T_{cc}</i>	cold crystallization temperature
<i>T_g</i>	glass transition temperature
<i>T_m</i>	melting temperature
<i>T_m⁰</i>	equilibrium melting temperature
TCA	tricarboxylic acid cycle
TEM	transmission electron microscopy
UHMW	ultra high molecular weight
UV	ultraviolet
VFAs	volatile fatty acids
WAXD	wide angle X-ray diffraction
WAXS	wide angle X-ray scattering
WWTP	wastewater treatment plant
<i>X_t</i>	percent crystallinity
XRD	X-ray diffraction

have very high purity within the cell [3–8]. PHA has a much better resistance to UV degradation than polypropylene but is less solvent resistant. Most importantly, these biopolymers are completely biodegradable. To date more than 150 different monomer units have been incorporated into biological PHA [3–8], and the polymer

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