Polyhydroxyalkanoates: The Natural Biopolyester for Future Medical Innovations

Zi-Wei Ren[1, #], Ze-Yu Wang[1, #], Yan-Wen Ding [1, #], Jin-Wei Dao[1,3], Hao-Ru Li[1], Xue Ma[1], Xin-Yu Yang[1], Zi-Qi Zhou[1], Jia-Xuan Liu[1], Chen-Hui Mi[1], Zhe-Chen Gao[2], Hua Pei[4]* Dai-Xu Wei[1,2,4,5*]

[1]Key Laboratory of Resource Biology and Biotechnology in Western China, Ministry of Education, School of Medicine, Department of Life Sciences and Medicine, Northwest University, Xi'an, 710069, China

[2]Department of Orthopedics, Tongren Hospital, Shanghai Jiao Tong University, Shanghai, 200336, China

[3]Dehong Biomedical Engineering Research Center, Dehong Teachers' College, Dehong, 678400, China

[4]Department of Clinical Laboratory, The Second Affiliated Hospital, Hainan Medical University, Haikou, 570311, China

[5]Shaanxi Key Laboratory for Carbon Neutral Technology, Xi'an, 710069, China

[#] Zi-Wei Ren, Ze-Yu Wang and Yan-Wen Ding contributed equally to this work.

*Correspondence: Dai-Xu Wei, weidaixu@nwu.edu.cn;
Key Laboratory of Resource Biology and Biotechnology in Western China, Ministry of Education, School of Medicine, Department of Life Sciences and Medicine, Northwest University, Xi'an, 710069, China

Table S1. Biosynthesis and productivity of PHAs via natural microorganism.

PHAs	Microorganisms	Carbon source	PHA content (%)	Average PHA productivity (g L−1 h−1)	Ref.s
PHB	*Azohydromonas lata*	Sucrose	50.0–88.0	0.050–4.940	[1-3]
	Azohydromonas lata	Fructose, glucose	76.5–79.4	0.121–0.128	[4]
	Azotobacter beijerinckii	Glucose	24.8	0.09	[5]
	Burkholderia cepacia	Xylose	58.4	ND	[6]
	Burkholderia cepacia	Glycerol	31.3	0.103	[7]
	Burkholderia cepacia	Fructose, glucose, sucrose	50.4-59.0	ND	[4]
	Burkholderia sp. USM	Lauric acid, myristic acid, oleic acid, palmitic acid, stearic acid	1.0–69.0	ND	[8]
	Caulobacter vibrioides	Glucose	18.3	0.008	[9]
	Cupriavidus necator H16	Fructose, glucose	67.0–70.5	0.052–0.067	[4]
	Cupriavidus necator H16	4-Hydroxyhexanoic acid	76.3–78.5	ND	[10]
	Cupriavidus necator H16	Corn oil, oleic acid, olive oil, palm oil	79.0–82.0	0.041–0.047	[11]
	Cupriavidus necator H16	CO2	88.9	0.23	[12]
	Cupriavidus necator	4-Hydroxyhexanoic acid	65.8–66.2	ND	[10]
	Cupriavidus necator	4-Hydroxyhexanoic acid	67.2	ND	[10]
	Cupriavidus necator	CO2	60	0.6	[13]
	Cupriavidus necator a	Glucose	76	2.42	[14]
	Cupriavidus necator a	Potato starch, saccharified waste	46	1.47	[15]
	Cupriavidus necator	Molasses	31.0–44.0	0.080–0.120	[16]
	Cupriavidus necator	Waste glycerol	14.8–36.1	0.330–4.200	[17]
	Halomonas boliviensis LC1	Hydrolyzed starch	56	ND	[18]
	Methylobacterium extorquens	Methanol	40.0–46.0	0.250–0.600	[19]
	Methylobacterium extorquens	Methanol	35.0–62.3	0.183–0.980	[20, 21]
	Methylocystis sp. GB25 a	Methane	51	ND	[22]
	Novosphingobium nitrogenifigens Y88	Glucose	81	0.014–0.021	[23]
	Streptomyces spp.	Glucose	1.2–82.0	ND	[24]

	Bacillus megaterium	Citric acid, glucose, glycerol, succinic acid	9.0–50.0	ND	
	Pseudomonas aeruginosa	Fructose, Sucrose, glucose, glycerinum	12.4–62.0	0.012–0.110	[25]
	Hydrogenophaga pseudoflava	Lactose, Sucrose	20.2–62.5	0.018–0.117	[26]
	Hydrogenophaga pseudoflava	Hydrolyzed whey and valerate	40	0.05	[27]
	Haloferax mediterranei	Vinasse	50.0–73.0	0.050–0.210	[28]
	Haloferax mediterranei	Whey	72.8	0.09	[29]
	Haloferax mediterranei	Glycerinum	75.0–76.0	0.12	[30]
	Rhodococcus	Acetate, 2-alkenoate, 1,4-butanediol, 5-chlorovalerate, fructose, glucose, hexanoate, 4-hydroxybuytrate, lactate, molasses, succinate, valerate	4.0–53.0	ND	[31]
	Cupriavidus necator	Glucose, propionic acid	80	0.82	[32]
PHV	*Paracoccus denitrificans*	n-Pentanol	22.0–24.0	ND	[33]
PHB & PHBV	Archaeal	Fructose, glucose, glycerinum	0.8–22.9	<0.001–0.021	[34]
scl-mcl-PHA	*Pseudomonas mendocina*	1,3-Butanediol, octanoate	13.5–19.3	ND	[35]
	Pseudomonas oleovorans	4-Hydroxyhexanoic acid	18.6	ND	[10]
	Thermus thermophiles HB8	Whey	35.6	0.024	[36].
scl-mcl-PHA, mcl-PHA	*Pseudomonas marginalis*	1,3-Butanediol, octanoate	11.9–31.4	ND	[35]
	Pseudomonas putida GPo1	n-Alkanoates	5.0–60.0	ND	[37, 38]
mcl-PHA	*Pseudomonas aeruginosa PAO1*	Oil and wax products from PE pyrolysis	25	ND	[39]
	Pseudomonas frederiksbergensis GO23 a	Terephthalic acid from PET pyrolysis	24	0.004	[40]
	Pseudomonas putida CA-3 a	Styrene	31.8	0.063	[41]
	Pseudomonas putida CA-3 a	Styrene from PS pyrolysis	36.4	0.033	[42]

	Pseudomonas putida GO16 a	Terephthalic acid from PET pyrolysis	27	~0.005, 0.008 d	[40]
	Pseudomonas putida GO19 a	Terephthalic acid from PE and PET pyrolysis	23	~0.005, 0.008 d	[40]
	Pseudomonas putida KT2440	Nonanoic acid	26.8–75.4	0.250–1.110	[43]
	Pseudomonas putida KT2440	4-Hydroxyhexanoic acid	25.3–29.8	ND	[10]
	Pseudomonas putida KT2440	Glucose	32.1	0.006	[44]
	Pseudomonas putida F1	Benzene, ethylbenzene, toluene	1.0–22.0	ND	[45]
	Pseudomonas putida mt-2	Toluene, p-xylene	22.0–26.0	ND	[45]

PHB: poly(3-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHV: poly(3-hydroxyvalerate); scl-PHA: short-chain length PHA; mcl-PHA: medium-chain length PHA; scl-mcl-PHA: short-chain length and medium-chain length PHA; PS: polystyrene; PET: polyethylene terephthalate; PE: polyethylene; ND: no data.

Table S2. Medical applications of PHAs for bone tissue engineering.

PHAs	Other materials	Methods	Type of devices	Cell types	Animal models	Laden drugs	Ref.s
PHB			Patches		Minipigs		[46]
PHB				CRL-1543			[47]
PHB			Cylindrical pins		Rats		[48]
PHB		Electrospinning	Films			Levofloxacin	[49]
PHB		Ultrasonic emulsion	Nanoparticles	OCT-1		BMP2	[50]
PHB	HA	Injection molding			Rabbits		[51]
PHB	HA	Solvent casting	Microspheres				[52]
PHB	HA		Films		Rabbits		[53]
PHB	HA		Films	Rat stromal osteoblastic cells	Rats		[54]
PHB	HA	Phase separation	Scaffolds	MC3T3-E1			[55]
PHB	HA	Solvent casting	Films	MC3T3-E1			[56]
PHB	nHA	Salt leaching	Scaffolds	MG-63			[57]
PHB	nHA	Electrospinning	Scaffolds	MSCs	Mice		[58]
PHB	HA&Gel	Electrospinning-electrospraying	Scaffolds	hMSCs			[59]

PHB	HA&Gel	Electrospinning	Scaffolds	MC3T3-E1		[60]
PHB	BG	Solvent casting	Films	MG-63		[61]
PHB	HA& BG	Injection molding			Rabbits	[62]
PHB	CS& Al_2O_3	Electrospinning	Scaffolds	MG-63		[63]
PHB	CMWCNT	Electrospinning	Scaffolds	MG-63		[64]
PHB	CNT	Electrospinning	Scaffolds	MG-63		[65]
PHB	MCNT	Electrospinning	Scaffolds	SMSCs	Rats	[66]
PHB	β-TCP	Foam replication	Scaffolds			[67]
PHB	Ag& β-TCP	Coating	Microspheres			[68]
PHB	Coral					[69]
PHB	MMT	Solvent casting	Films	Human blood lymphocyte cells		[70]
PHB	PEG	Solvent casting	Films	CHL fibroblasts		[71]
PHB	PEG	Electrospinning	Scaffolds			[72]
PHB	PANi& rGO	Electrospinning	Scaffolds		Rats	[73]
PHB	Starch	Electrospinning	Scaffolds	MG-63		[74]

P4HB		Solvent evaporation	Microspheres	ADSCs	Rats	[75]
PHBV		Phase separation	Foams	Rat stromal osteoblastic cells		[76]
PHBV		Solvent evaporation& solute leaching	Foams	Rat stromal osteoblastic cells	Rats	[77]
PHBV		Electrospinning	Films& fibers	MG-63& NIH 3T3		[78]
PHBV	HA	Injection molding	Plates			[79]
PHBV	HA	Injection molding	Scaffolds		Rabbits	[80]
PHBV	HA	Electrospinning	Scaffolds	MG-63		[81]
PHBV	HA& TCP	Melt casting	Films			[82]
PHBV	HA& β-TCP	MAPLE	Films	hMSCs		[83]
PHBV	BG	Solvent casting-particulate leaching	Scaffolds	HUVECs& HBMSCs	Mice	[84]

PHBV	HA, SGBG& TCP	Salt leaching	Scaffolds	Rabbit primary osteoblast cells			[85]
PHBV	HA&PLA	Electrospinning	Membranes				[86]
PHBV	nHA	Melt processing&solvent casting	Films	Macrophages& osteoclasts	Mice		[87]
PHBV	nHA& nD	Solvent evaporation & solvent casting	Nanoparticles	RAW 264.7		Vancomycin	[88]
PHBV	nHA& SF	Electrospinning	Scaffolds	HOB			[89]
PHBV	nHA& CS	Electrospinning	Scaffolds	hFOB			[90]
PHBV	nHA& Col	Salt leaching	Scaffolds	MC3T3-E1			[91]
PHBV	n-HA& PCL	Solvent-casting& particulate-leaching	Scaffolds	Saos-2			[92]
PHBV	BW	Solvent-casting& particulate-leaching	Scaffolds				[93]

PHBV	Wollastonite	Thermoplastic method	Scaffolds	Osteoblasts			[94]
PHBV	MBGN& CIN	Emulsion solvent extraction& evaporation method	Microspheres	MG-63			[95]
PHBV	PG		Films		Dogs		[96]
PHBV	PCL	Solvent casting	Membranes	rMSCs			[97]
PHBV	PCL	Injection molding	Scaffolds	Vero cells& hMSCs			[98]
PHBV	PCL, GO&CP	Electrospinning	Scaffolds				[99]
PHBV	PCL-pullulan& DS	Electrospinning	Scaffolds	Saos-2		CA	[100]
PHBV	ELR-REDV	Wet spinning	Scaffolds	rBMSCs			[101]
PHBV	Aloe vera gel	Electrospinning	Scaffolds	iPSCs			[102]
PHBV	G-BR	Electrospinning	Nanofibrous	hFOB			[103]

PHBV	BR&FG	Electrospinning	Membranes	hFOB		[104]
PHBV	CaSH& CS	3D Printing	Scaffolds	rBMSCs	Rats	[105]
PHBV	ZnO	3D Printing	Scaffolds	MG-63		[106]
P34HB		Electrospinning	Scaffolds	rBMSCs	Rats	[107]
P34HB	PEG	Electrospinning	Membranes			[108]
P34HB	PVA	Electrospinning	Scaffolds	hBMSCs	Mice	[109]
PHAs	β-TCP	3D printing	Scaffolds	MC3T3-E1		[110]
PHBHHx		Solvent evaporation	Films	MC3T3-E1		[111]
PHBHHx		Salt leaching	Scaffolds	Rabbit bone marrow cells		[112]
PHBHHx		Electrospinning	Scaffolds	MSCs		[113]
PHBHHx		Solvent evaporation	Films	MSCs		[114]
PHBHHx	SF	Electrospinning	Films	hUC-MSCs		[115]

PHBHHx	MBG	3D printing	Scaffolds	hMSCs			[116]
PHBHHx	MBG	3D printing	Scaffolds	hBMSCs	Rats		[117]
PHBVHHx		Method of gas-in-oil-in-water double emulsion	Microspheres	hMSCs	Mice		[118]
PHBVHHx	PLA& SL	Emulsion-solvent evaporation	Nanoparticles	hADSCs		BMP-2& BMP-7	[119]
P(3HB-4HB-3HHx)		Solvent evaporation	Films	hMSCs			[120]
PHO	TCP	Coating					[121]
PHB& PHBV		Electrospinning	Scaffolds	HDFs			[122]
PHB & PHBV	$CaCO_3$	Electrospinning	Scaffolds	MC3T3-E1			[123]
PHB & PHBHHx	HA	Salt leaching	Scaffolds & films	Rabbit bone marrow cells			[124]

PHB& P(3HO-*co*-3HD-*co*-3HDD)	HA	Solvent casting	Films	125

PHB: poly(3-hydroxybutyrate); P4HB: poly(4-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); P34HB: poly(3-hydroxybutyrate-co-4-hydroxybutyrate); PHBVHHx: poly(3-hydroxybutyrate-co-3-hydroxyvalerate-co-3-hydroxyhexanoate); PHO: poly(3-hydroxyoctanoate); P(3HB-4HB-3HHX): poly(3-hydroxybutyrate-co-4-hydroxybutyrate-co-3-hydroxyhexanoate); P(3HO-*co*-3HD-*co*-3HDD): P(3-hydroxyoctanoate-co-3-hydroxydecanoate-co-3-hydroxydodecanoate); HA: hydroxyapatite; nHA: nano scale HA; Gel: gelatin; BG: bioactive glass; CS: chitosan; CMWCNT: carboxyl multi-walled carbon nanotubes; CNT: carbon nanotubes; MCNT: Multiwalled carbon nanotubes; *β*-TCP: β-tricalcium; MMT: modified montmorillonite; PANi: polyaniline; PEG: polyethylene glycol; rGO: reduced graphene oxide; TCP: tricalcium phosphate; SGBG: sol-gel bioglass; PLA: polylactic acid; nD: nanodiamond; SF: silk fibroin; CS: chitosan; Col: collagen; PCL: polycaprolactone; BW: bioactive wollastonite; MBGN: mesoporous bioactive glass nanoparticles; CIN: cinnamaldehyde; PG: Polylactin 910; GO: Graphene oxide; CP: Calcium phosphate; DS: diatom shell; ELR-REDV: Elastin-like recombinamer (ELR) with the REDV sequence (R: L-Arginine, E: L-Glutamic acid, D: L-Aspartic acid, and V: L-Valine); BR: Bredigite; G-BR: 3-glycidoxypropyltrimethoxysilane (GPTMS)-modified BR; FG: fibrinogen; PVA: polyvinyl alcohol; MBG: mesoporous bioactive glass; SL: soybean lecithin; MAPLE: matrix assisted pulsed laser evaporation; CHL: Chinese Hamster Lung

Table S3. Medical applications of PHAs for cartilage tissue engineering.

PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
PHB	CS	Melt casting	Scaffolds	rMSCs	Sheep		[126]
PHB	CS	Electrospinning	Scaffolds	Rabbit chondrocyte cells	Rabbits		[127]
PHB	CS& MWNTs	Electrospinning	Scaffolds	Rabbit chondrocyte cells	Rabbits		[128]
PHBV		Solvent casting-particulate leaching	Scaffolds	Chondrocytes, BMSCs& cartilage progenitor cells	Mice		[129]
PHBV	BG	Stirring emulsification	Scaffolds	Rabbit chondrocyte cells	Rabbits		[130]
PHBV	CMCht & SF	Electrospinning	Scaffolds	rBMSCs	Rats		[131]
PHBV	QUE	Electrospinning	Scaffolds	Rabbit chondrocyte cells	Nude mice		[132]
PHBHHx		Solvent casting	Scaffolds		Rabbits	Chondrocytes	[133]
PHB& PHBHHx		Solvent casting	Scaffolds	Rabbit chondrocytes, BM-MSCs & ASCs	Rabbits		[134]

P34HB& PHBHHx	Solvent casting	Films	Rabbit chondrocyte cells	[135]

PHB: poly(3-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); P34HB: poly(3-hydroxybutyrate-co-4-hydroxybutyrate); CS: chitosan; MWNTs: multi-walled carbon nanotubes; BG: bioactive glass; CMCht: carboxymethyl chitosan; SF: silk fibroin; QUE: Quercetin

Table S4. Medical applications of PHAs for skin tissue engineering.

PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
PHB		Electrospinning	Films	ADSCs		Cur	[136]
PHB	CNT	Electrospinning	Scaffolds	Human PDLSCs	Rats		[137]
PHB	CS	Coprecipitation	Scaffolds	L929			[138]
PHB	O-CS	Electrospinning	Films	L930			[139]
PHB	Gel	Electrospinning	Scaffolds	Human dermal fibroblasts			[140]
PHB	Gel	Electrospinning	Scaffolds	Balb/3T3	Rats		[141]
PHB	BC	Solvent casting	Films	L929			[142]
PHB	BC	Solvent casting	Scaffolds	CHL fibroblast			[143]
PHB	CA	Electrospinning	Scaffolds	3T3			[144]
PHB	SPN	Electrospinning	Scaffolds	NIH3T3			[145]
PHB	GO& SA	Solvent casting	Scaffolds	WS1 human skin fibroblast cells		CUR&GS	[146]

PHB	PVA	Electrospinning	Scaffolds	HaCaT			[147]
PHB	PANi	Electrospinning	Scaffolds	L929			[148]
PHBV		Electrospinning	Nanofibers	HaCaT			[149]
PHBV		Electrospinning	Nanofibers	BM-MSCs			[150]
PHBV		Electrospinning	Scaffolds	HDF	Mice		[151]
PHBV	CS	Electrospinning	Scaffolds	L929	Rats		[152]
PHBV	Col	Electrospinning	Scaffolds		Rats	USSC	[153]
PHBV	Col&Gel	Electrospinning	Nanofibers	Human DS cells	Mice		[154]
PHBV	Col& GO	Electrospinning	Scaffolds	3T3-L			[155]
PHBV	GO & CNFs	Solvent casting	Films	ASC			[156]
PHBV	Pullulan	Electrospinning	Scaffolds	L929			[157]
PHBV	PDX	Electrospinning	Scaffolds	L929			[158]
PHBV	Gel & polyethylene oxide	Electrospinning	Electrospun Mats	L929			[159]

P34HB	Col	Electrospinning	Nanofibers	L929	Rats	[160]
P34HB	PCL	3D printing	Scaffolds	HUVECs& BMSCs	Rats	[161]
PHBVHHx		Solvent casting& solvent-extraction	Films& nanoparticles	HaCaT		[162]
PHB& P4HB	HA, CS, pectin& alginic acid	Solvent casting	Films	HaCaT		[163]
PHB& P(3HO-*co*-3HD)		Electrospinning	Nanofibril	Human keratinocytes		[164]
PHBV& PHBHHx	PLA	Melt spinning	Fibers		Rats	[165]

PHB: poly(3-hydroxybutyrate); P4HB: poly(4-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); P34HB: poly(3-hydroxybutyrate-co-4-hydroxybutyrate); PHBVHHx: poly(3-hydroxybutyrate-co-3-hydroxyvalerate-co-3-hydroxyhexanoate); CNT: carbon nanotubes; CS: chitosan; O-CS: organic-soluble chitosan; Gel: gelatin; BC: bacterial cellulose; CA: cellulose acetate; SPN: soybean protein nanoparticles; GO: graphene oxide; SA: sodium alginate; PVA: polyvinyl alcohol; PANi: polyaniline; Col: collagen; CNFs: carbon nanofibers; PDX: polydioxanone; PCL: polycaprolactone; PLA: polylactic acid; CHL: Chinese Hamster Lung; CUR: curcumin; CS: chitosan; GS: gymnema sylvestre; USSC: unrestricted somatic stem cells

Table S5. Medical applications of PHAs for muscle and esophagus tissue engineering.

Applications	PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
Muscle	PHB		High-speed melt spinning& spin drawing process	Scaffolds		Rats		[166]
	PHB		High-speed melt spinning& spin drawing process	Scaffolds		Rats		[167]
	PHB		Electrospinning	Scaffolds	C2C12& H9c2			[168]
Esophagus	PHBV	Gel	Electrospinning	Scaffolds	HEEpiC			[169]
	PHBHHx	PLGA&SIS	Solution casing	Membrane& scaffolds	MSCs	Rats		[170]

PHB: poly(3-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); Gel: gelatin; PLGA: poly (lactic-co-glycolic acid); SIS: small intestinal submucosa

Table S6. Medical applications of PHAs for nerve tissue engineering.

PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
PHB			Conduits		Rabbits	GGF	[171]
PHB			Conduits		Rats		[172]
PHB			Conduits		Rats		[173]
PHB			Conduits		Rats		[174]
PHB			Conduits		Rabbits		[175]
PHB			Conduits	SCs	Rats		[176]
PHB		Coating	Conduits	MSCs& SCs	Rats		[177]
PHB		Coating	Conduits		Rabbits	GGF	[178]
PHB			Scaffolds		Rats		[179]
PHB	PCL	Salt leaching	Scaffolds	iPS			[180]
PHBV		Solvent evaporation	Microspheres	Neuro2a cells			[181]
PHBV		Solvent evaporation	Microspheres	PC12, CNs& NPCs			[182]
PHBV	Col	Electrospinning	Scaffolds	PC12			[183]

PHBV	CS	Electrospinning	Scaffolds	SCs	Rats	[184]
PHBHHx		Particle leaching	Conduits		Rats	[185]
PHBHHx		Solvent casting	Films&scaffolds	NSCs		[186]
PHBHHx	PDLLA	Solvent casting	Films			[187]
PHBHHx	rGO& Au	Electrospinning	Scaffolds	SCs		[188]
PHBVHHx		Solvent casting& phase separation	Membranes& scaffolds	hBMSCs		[189]
P(3HO-3HD)	PCL	Solvent casting	Conduits	NG108-15& SCs	Rats	[190]
PHB& PHBV		Electrospinning	Scaffolds	SCs		[191]
PHB& PHO		Solvent casting	Conduits	SCs		[192]
PHB & PHO		Electrospinning	Scaffolds	NG108-15 neuronal cells & RN22 Schwann cells		[193]

				NG108-15 neuronal cells & RN22	
PHB&PHO	BG	Solvent casting	Films	Schwann cells	[194]

PHB: poly(3-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); PHBVHHx: poly(3-hydroxybutyrate-co-3-hydroxyvalerate-co-3-hydroxyhexanoate); PHO: poly(3-hydroxyoctanoate); P(3HO-3HD): poly(3-hydroxyoctanoate-co-3-hydroxydecanoate); PCL: polycaprolactone; Col: collagen; GS: gymnema sylvestre; PDLLA: poly (DL-lactide); rGO: reduced Graphene oxide; BG: bioactive glass; GGF: glial growth factor

Table S7. Medical applications of PHAs for cardiovascular tissue engineering.

PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
PHB		Solvent casting	Patches	L929	Rabbits& sheep		[195]
PHB		Electrospinning	Nanofibers	MSCs, cardiomyocytes & cardiac fibroblasts	Rats		[196]
PHB	ePTFE	Electrospinning	Vascular Disease Detector	HUVECs			[197]
P4HB		Salt leaching	Scaffolds	Sheep SMCs, endothelial cells & fibroblasts			[198]
P4HB		Salt leaching & solvent evaporation	Patches	Endothelial cells, smooth muscle cells & fibroblasts	Lambs		[199]
P4HB	PGA	Solvent evaporation	Scaffolds	Myofibroblasts	Lambs		[200]
P4HB	PGA	Coating	Patches	Ovine EPCs & MSCs	Sheep		[201]
PHBV		Electrospinning	Scaffolds	HDFs & HDMECs			[202]

PHBV		Electrospinning	Scaffolds		Sheep	VEGF, bFGF, SDF-1α, surface coated with heparin& iloprost	203
PHBV	PCL	Electrospinning	Scaffolds		Rats		204
PHBV	PCL	Electrospinning	Scaffolds		Rats	VEGF, bFGF&SDF-1α	205
PHBV	PCL	Electrospinning	Scaffolds		Rats	VEGF& RGD	206
PHBV	PCL	Electrospinning	Scaffolds		Sheep	VEGF、bFGF、SDF-1α、heparin& iloprost	207
PHBV	PCL	Electrospinning	Patches		Rats	RGD	208
PHBV	PCL	Electrospinning	Patches		Rats	VEGF	209
PHBHHx		Solvent casting	Patches		Rabbits		210
PHBHHx		Electrospinning	Scaffolds				211
PHBHHx		Electrospinning	Scaffolds	PIECs			212

PHBHHx	SF		Scaffolds	Human fibroblasts, hSMCs & HUVECs			[213]
PHBHHx	Decellularized porcine aortic valves	Coating	Patches& conduits		Rabbits& sheep		[214]
PHBHHx	Plasma treatment &fibronectin coating	Solvent casting	Films	HUVECs & SMCs			[215].
P34HB		Solvent casting	Films& scaffolds	RaSMCs			[216]
P34HB		Ultrasonic spray-coating	Films	L929			[217]
PHO		Salt leaching	Scaffolds	Ovine vascular cells	Lambs		[218]
PHO		Electrospinning	Nanofibers	Rat cardiomyocytes		VEGF& RGD	[219]
PHO	PGA		Scaffolds	Eendothelial cells, smooth muscle cells & fibroblasts	Lambs		[220]
PHB & PHBV		Solvent casting & Electrospinning	Films	3T3, L929, HUVECs & EPCs			[221]

PHB, PHBV & PHBHHx		Solvent casting	Films	HUVECs	[222]
PHB, PHBV& PHO		Solvent casting& electrospinning	Films & scaffolds		[223]
PHO & P(3HN-co-3HHP)			Scaffolds	NVRM	[224]
MCL-PHA	PCL	Solvent casting	Films	CPCs Mice	[225]
PHA		Salt leaching	Scaffolds	Ovine vascular cells	[226]
PHA		Salt leaching	Scaffolds	Ovine vascular cells	[227]

PHB: poly(3-hydroxybutyrate); P4HB: poly(4-hydroxybutyrate); PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); P34HB: poly(3-hydroxybutyrate-co-4-hydroxybutyrate); PHO: poly(3-hydroxyoctanoate); P(3HN-co-3HHP): poly (3-hydroxynonanoate-co-3-hydroxyheptanoate); ePTFE: an expanded polytetrafluoroethylene; PGA: polyglycolic acid; PCL: polycaprolactone; SF: silk fibroin; RGD: Arg-Gly-Asp; VEGF: vascular endothelial growth factor; bFGF: basic fibroblast growth factor; SDF-1α: stromal cell-derived factor 1

Table S8. Medical applications of PHAs for liver, tendon, eyes and eyelid tissue engineering.

Applications	PHAs	Other materials	Method	Type of devices	Cell type	Animal model	Drug delivery	Ref.s
Liver	PHBV		Solvent evaporation	Microspheres	HepG2& Hep3B			[228]
Liver	PHBV	PLGA	Solvent evaporation	Microspheres	Rat primary hepatocytes		HGF	[229]
Liver	PHBVHHx			Scaffolds	UC-MSCs	Mice		[230]
Tendon	PHBHHx	Col	Particle leaching & extrusion methods	Tubes & fibers		Rats	Tenocyte	[231]
Tendon	PHB, PHBV, PHUA, PHUE, PHOUE PHOUE-POSS, & PHUE-O3,		Solvent casting & phase separation	Films	L929			[232]
Eyes	PHBV		Solvent evaporation	Microspheres		Mice	Rapamycin	[233]
Eyelid	PHBHHx		Solvent casting	Scaffolds		Rats		[234]
Eyes	PHBHHx	PEG& PPG	Chemical synthesis	Thermogels		Rabbits		[235]

PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBHHx: poly (3-hydroxybutyrate-co-3-hydroxyhexanoate); PHBVHHx: poly(3-hydroxybutyrate-co-3-hydroxyvalerate-co-3-hydroxyhexanoate); PHUA: poly(3-hydroxyundecanoate); PHUE: poly(3-hydroxy-10-undecenoate); PHOUE: poly(3-hydroxyoctanoate-co-3-hydroxy-10-undecenoate; PHOUE-POSS: PHOU (50/50) derivatised with polyhedral oligomeric silsesquioxane; PHUE-O3: ozone treated PHUE with unknown degree of oxidation, exact formula not known; PLGA: poly(lactic-co-glycolic acid); Col: collagen; PEG: polyethylene glycol; PPG: poly(propylene glycol); HGF: hepatocyte growth factor

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