

# Circular Development of Sludge Cellulose/Biopolymer Composites from Municipal Wastes

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## Introduction

This work focuses on the development of biocomposites by the use of biodegradable resources (i.e. bioplastics and cellulosic materials) derived from municipal wastewater, aiming to reduce the resource competition and environmental impact of wood plastic composites (WPC) industry.

The prior issue to be addressed in the development of cellulosic polymer composites is the constituent compatibility, i.e. the large number of hydroxyl groups existed in the structure of cellulose makes it incompatible with hydrophobic and non-polar bioplastics, leading to poor interfacial bonding and thus mechanical properties.

This work concentrates on the interfacial optimisation of sludge cellulose (SC)/polyhydroxyalkanoate (PHA) composites (SPC) by the incorporation of various coupling agents, and the effect of improved interface on the mechanical properties of the composites. For

comparison purposes, the conventional WPC and pure cellulose fibre (CF) based PHA composites were also developed.

## Materials and Methods

Recovered sludge cellulose was provided by CirTec BV (Netherlands), PHA, i.e. poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), was obtained from TianAn Biologic Materials Co., Ltd (China). Recycled wood flour (WF) and polyethylene (PE) were supplied by Rettenmeier Holding AG (Germany) and JFC Plastics Ltd (UK) respectively. Coupling agents including maleic anhydride (MA),  $\gamma$ -aminopropyltriethoxysilane (APS) and vinyltrimethoxysilane (VTMS) were purchased from Sigma-Aldrich UK.

A Brabender Plastograph twin-screw mixer was used for the compounding of the materials and the formulation method was hot-press compression moulding. The formulation of SC/PHBV composites (SPC) and comparison composites with specific ratio were summarised in Table 1.

**Table 1 Formulation of SPC**

Composites	Polymer matrix		Cellulosic filler			Lubricant and Compatibiliser		Initiator	Coupling agent		
	PHBV (%)	PE (%)	SC (%)	WF (%)	CF (%)	TPW 709 (%)	12HSA (%)	Peroxide (%)	MA (%)	APS (%)	VTMS (%)
PHA	100	0	0	0	0	0	0	0	0	0	0
PE	0	100	0	0	0	0	0	0	0	0	0
PLA	0	0	0	0	0	0	0	0	0	0	0
Untreated SPC	55	0	40	0	0	2.5	2.5	0	0	0	0
MA treated SPC	52	0	40	0	0	2.5	2.5	0.3	2.7	0	0
APS treated SPC	52	0	40	0	0	2.5	2.5	0.3	0	2.7	0
VTMS treated SPC	52	0	40	0	0	2.5	2.5	0.3	0	0	2.7
MA treated SC-PE	0	52	40	0	0	2.5	2.5	0.3	2.7	0	0
MA treated WF-PE	0	52	0	40	0	2.5	2.5	0.3	2.7	0	0
MA treated WF-PHBV	52	0	0	40	0	2.5	2.5	0.3	2.7	0	0
Untreated CF-PHBV	55	0	0	0	40	2.5	2.5	0	0	0	0
MA treated CF-PHBV	52	0	0	0	40	2.5	2.5	0.3	2.7	0	0
APS treated CF-PHBV	52	0	0	0	40	2.5	2.5	0.3	0	2.7	0
VTMS treated CF-PHBV	52	0	0	0	40	2.5	2.5	0.3	0	0	2.7

**Table 2 Mechanical properties of matrices, SMART-Plant SPC, WPC, and CF-PHBV composites**

Matrix/Composite	Tensile stress (MPa)	Tensile strain (%)	Flexural stress (MPa)	Flexural extension (mm)
PHBV	26.06	2.46	64.62	8.46
PE	23.26	8.61	21.95	11.17
PLA	39.11	1.50	43.69	2.46
Untreated SPC	5.81	0.31	3.73	0.25
MA treated SPC	10.17	0.45	8.65	0.40
APS treated SPC	6.30	0.39	2.61	0.20
VTMS treated SPC	9.55	0.45	9.54	0.39
SC-PE	13.58	1.69	23.91	3.80
WF-PE	17.15	2.48	24.80	3.40
WF-PHBV	8.40	0.37	4.25	0.13
Untreated CF-PHBV	9.22	0.50	9.88	0.44
MA treated CF-PHBV	15.73	0.67	16.40	0.65
APS treated CF-PHBV	8.78	0.40	6.94	0.30
VTMS treated CF-PHBV	12.36	0.49	21.46	0.95

## Results and Discussion

The mechanical properties of polymer matrices, SPC, WPC, and CF-PHBV composites were summarised in Table 2.

PHBV exhibited a tensile stress of 26.06 MPa which is slightly higher than that of PE but lower than that of PLA. PHBV was more ductile than PLA by showing a higher tensile strain (2.46%), and PE showed much better ductility as expected. It was shown that the incorporation of the coupling agents increased the tensile stress of the composites by 75.0% (MA), 8.4% (APS) and 64.4% (VTMS) compared to the untreated composite (5.81 MPa). In addition, the tensile strain of the composites was also increased by 45.2% (MA), 25.8% (APS) and 45.2% (VTMS). These results suggested that the coupling agent treatments, especially MA and VTMS treatments significantly stiffened and strengthened the composites mainly due to the considerable increase of the constituent compatibility (SC/PHBV), interfacial adhesion and bonding.

PHBV exhibited a maximum flexure stress of 64.62 MPa, which is around 200% and 50% higher than that of PE and PLA while remains a flexure extension of 8.46 mm. As per poor extension showed in tension, PHBV appeared to be an anisotropic polymer. PE and PLA seemed to be isotropic polymers since PE exhibited outstanding extensions in both tension and bending, while PLA showed the poorest extensions in both circumstances.

Regarding the coupling agent treatments, the introduction of MA and VTMS led to a significant increase of the flexure stress of the composites by 131.9% and 155.8% respectively, along with the increase of flexure extension by 60% and 56% respectively. These results are consistent with the aforementioned tensile property analysis, confirming that MA and VTMS coupling agents were capable to enhance the miscibility and compatibility of the constituents, improve the bonding quality and consequently the mechanical properties of SPC.

Noteworthy that PHBV has a superior tensile stress over PE, but the tensile stress of PHBV based composites, such as MA treated SPC and WF-PHBV, was lower than that of PE based counterparts, i.e. SC-PE and WF-PE. This might suggest that PE was more compatible with cellulosic fillers than PHBV.

## Conclusions

It is feasible to develop a class of composites based on sludge cellulose and PHA derived from municipal wastewater. It was concluded that MA and VTMS coupling agents could considerably enhance the compatibility of sludge cellulose and PHA, leading to a significant increase of mechanical properties.

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