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Effects of high-lignin-loading on thermal, mechanical, and morphological properties of bioplastic composites

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1 **Effects of high-lignin-loading on thermal, mechanical, and morphological properties of bioplastic**
2 **composites**

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15 **Abstract**

16 The present study investigates the effects of high-lignin-loading on properties of lignin/polyethylene-*co*-
17 vinyl acetate (EVA) rubber composites. Results from mechanical (quasi-static and cyclic) and rheological
18 investigations revealed a brittle-ductile transition around a lignin volume fraction of $\varphi_L = 0.59$ coinciding
19 with a two-fold increase in steady-shear viscosity. Towards higher lignin contents, a 36 % increase in
20 dynamic stiffness C_{dyn} from $\varphi_L = 0.59$ ($C_{\text{dyn}} \approx 350 \text{ N mm}^{-1}$) to $\varphi_L = 0.71$ ($C_{\text{dyn}} \approx 550 \text{ N mm}^{-1}$) was
21 observed by load increase tests (LIT). In addition, analyses of the ultra-micro-hardness revealed less
22 indentation creep towards high-lignin-loading. At $\varphi_L = 0.59$, a pronounced relaxation endotherm
23 superimposed on the glass transition (T_g) was observed, which was ascribed to molecular confinement to
24 occur at highly loaded composites. At this point, the molecular weight (M_w) of lignin increased
25 considerably. These results were explained by the different role of lignin at high volume fraction, i.e. a
26 change from lignin as mere stiffness-inducing filler to a strength-imparting and fatigue-resistant matrix
27 component which was supported by morphological analysis.

28

29 **Keywords**

30 Polymer matrix composites (PMC), Lignin composites, Extrusion, Mechanical testing, Fatigue.

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