Supplementary Information

Additional Experimental Methods

Differential Scanning Calorimetry (DSC) Thermal properties of poly(vinyl) PVAc films and tunicate whisker/poly(vinly) alcohol (TW/PVAc) composites were investigated using a TA Q100 heat-flux differential scanning calorimeter. Samples of mass 7.5 ± 0.5 mg, enclosed in hermetic aluminium pans, were used and were heated and then cooled and then heated again in the range 25 - 200 °C at 10 °C min⁻¹ under a 50 mL min⁻¹ nitrogen purge gas flow.

Dynamic Mechanical Analysis (DMA) The mechanical properties of the nanocomposites were characterized by Dynamic Mechanical Analysis (DMA, TA instruments Model Q800). Tests were conducted in tensile mode using a temperature sweep method (23 - 100 °C) at a fixed frequency of 1 Hz, and a strain amplitude of 15 μ m. All samples were dried in vacuum (65°C, -80 KPa, 16 - 18 h) prior to DMA testing (unless swollen materials were measured). In order to determine the tensile properties of the nanocomposite films in the wet state, samples were swollen in water at 25 °C for 1 week and DMA experiments were conducted using the above set up with a submersion clamp, which allowed measurements while the samples were immersed in water; in this case, temperature sweeps were conducted from 20 - 50°C.



(b)



(c)



Figure 1(a) Dynamic mechanical analysis (DMA) of tunicate whisker/poly (vinyl acetate) (TW/PVAc) composites over the temperature range 25 to 90°C. Two repeats are shown (Sample 1 and 2); (b) the intensity of the 1095 cm⁻¹ Raman band as a function of rotation angle (polar scale) of a 12.2% v/v tunicate whisker/poly(vinyl acetate) (TW/PVAc) nanocomposite film that that was produced by solution-casting from DMF without compression moulding; (c) Stress-strain curves for pressed (dry), solution cast (dry) and wet TW/PVAc composite samples obtained during deformation under the Raman microscope.