Electronic Supplementary Information.

Microstructural and property changes in high pressure treated carbon nanotube/polybutadiene composites

Junchun Yu[1], Bounphanh Tonpheng[1], Gerhard Gröbner[2], and Ove Andersson[1]*

[1]*Department of Physics, Umeå University, 901 87 Umeå, Sweden*
[2]*Department of Chemistry, Umeå University, 90187 Umeå, Sweden*

*Corresponding author. E-mail: ove.andersson@physics.umu.se

Results for 5wt% SWCNT and MWCNT filled polyisoprene (PI) show similar behaviour in glass transition temperature (T_g) as that discussed in the paper. As shown in Fig. S1, the T_g of 5 wt% SWCNT[1] and 5 wt% MWCNT filled PI are almost identical: 216.4 and 216.6 K, respectively, at atmospheric pressure, which is ~7 K higher than that of pure PI[2] (209.4 K). After treatment at 1.0 GPa and 513 K for 4 h, T_g at atmospheric pressure differed ~2 K between the two composites: 237.6 K and 235.3 K for SWCNT/PI[1] and MWCNT/PI, respectively, which increased to as much as ~10 K in the 0.5 to 1 GPa range. The more pronounced raise in T_g for the SWCNT/PI composite may thus be due to the slightly larger SSA of SWCNT (The similarly treated pure PI showed a T_g of 225.4 K[2]).

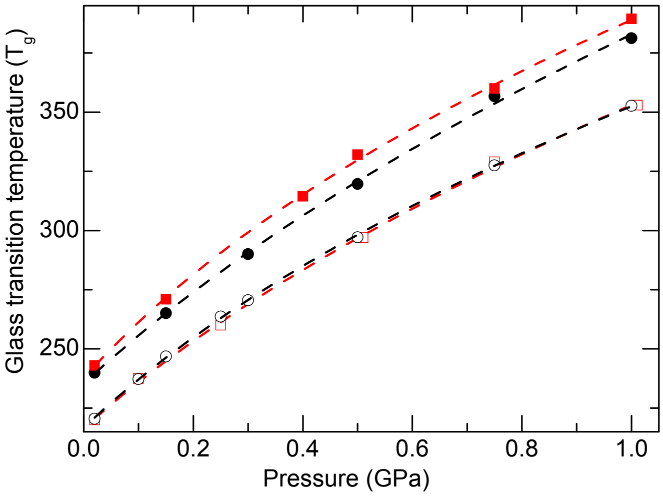

Fig. S1. Glass transition temperature plotted against pressure: untreated 5 wt% SWCNT/PI (□) from ref 1, MWCNT/PI (○) and 1.0 GPa treated 5 wt% SWCNT/PI (■) from ref 1, MWCNT/PI (●). The dashed lines represent fits of eq 1.

As depicted in Fig. S2, the MWCNTs in 1.5 GPa treated PB33MWCOOH formed a network structure. The web-like extended structures are observed in the height and amplitude images as well as in the phase image. The structures have lengths exceeding 5 μm, which is a limit set by the size of the scan.

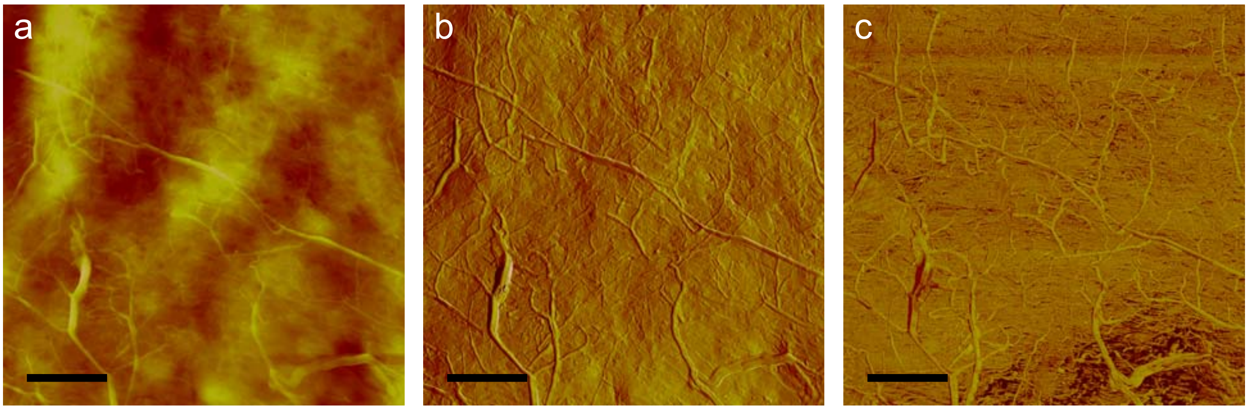

Fig. S2. Morphology analysis of the PB33MWCOOH composite by AFM: (a) Height image; (b) amplitude image; and (c) phase image of a surface oriented perpendicular to the applied pressure. The scale bars in all images are 1 μm and *z* range=120° in (c).

As shown in Fig. S3, the D^*- mode Raman peak, using argon-ion laser excitation, is at about the same wave number for the MWCNT-SH and PB3MWSH samples, but occurs at ~19 cm^{-1} lower wave number for the 1.5 GPa treated PB3MWSH sample. This difference is about the same as observed using He-Ne laser excitation (~16 cm^{-1}).

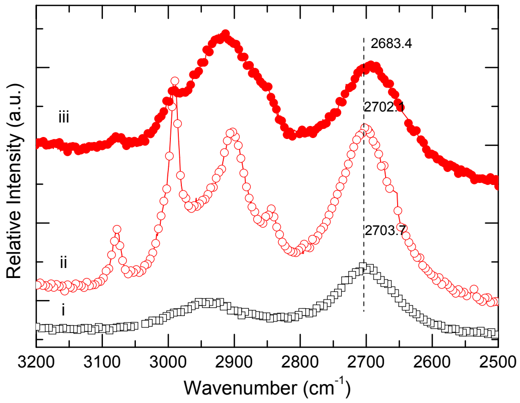

Fig. S3. (a) Raman spectra of the D* vibration of PB based composites obtained using argon-ion laser excitation: (i) MW-SH, (ii) PB3MWSH, and (iii) 1.5 GPa treated PB3MWSH.

Reference:

1. B. Tonpheng, J. Yu and O. Andersson, *Macromolecules*, 2009, **42**, 9295.

2. B. Tonpheng and O. Andersson, *Eur. Polym. J.*, 2008, **44**, 2865.