Rapid synthesis of multi-layered & multi-functional polymer nanocomposite films

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Polymer nanocomposites consist of a polymer matrix with nano-sized fillers. They may exhibit different properties than the pure polymer or a polymer with micron-sized fillers (Hagleitner, 2001). This change comes forth due to the unique material properties of nanoparticles as well as their increased specific surface area. An increase in mechanical durability (Camenzind, 2010), greater thermal stability (Kashiwagi, 2005) is common. Incorporation of superparamagnetic, plasmonic or phosphorescence properties is also possible by (Sotiriou, adjusting the filler material 2013). Agglomeration of the fillers, however, seriously hinders this task (Schulz, 2008). Nanocomposite homogeneity is achieved usually at the expense of lower filler loading even though fillers are surface functionalized prior to their embedding in the polymer (Suter, 2011). With this step their loading can be enhanced (e.g. up to 20%) while maintaining dispersion homogeneity (Schulz, 2008). This, however, increases the cost and complicates the processing.

Here a new route for synthesis of nanocomposite polymer films was invented. Superparamagnetic, phosphorescent and plasmonic polymer nanocomposites were made by directly depositing and integrating flamemade Fe₂O₃, Eu/Tb-doped Y_2O_3 and Ag nanoparticles, respectively, into a poly-methyl methacrylate (PMMA) matrix. Such properties allow the polymer film to serve as a sensor, actuator, display or electromagnetic shield (Sotiriou, 2013). Such a multitude of functionalities in one film is desired in miniaturized devices such as labon-a-chip (Suter, 2011).

The entire manufacturing procedure consists of dry synthesis of nanoparticles by flame-spray pyrolysis (Strobel, 2007) while directly depositing them on a cooled substrate and subsequently spin-coating with a polymer solution (Sotiriou, 2013). By varying the particle deposition parameters, *in-situ* flame annealing conditions and the spin-coating speed, the particle film thickness and filler loading (i.e. vol%) can be selected.

The nanoparticle loading in the polymer films is determined by thermogravimetric analysis. These loadings are higher than conventional methodologies without affecting film flexibility. Figure 1A shows the response of such a film to an external magnet. Imaging of this nanocomposite film by electron microscopy reveals a homogeneous layered particle distribution throughout the polymer (Figure 1B).

A free-standing thin flexible film (Fig. 1A) can be achieved by coating the deposition substrate with a sacrificial layer prior to manufacturing. Furthermore multi-layered structures with the same and/or different nanoparticles are easily achieved (Sotiriou, 2013). The substrate for deposition consists of a pre-existing polymer nanocomposite film. This allows one to easily incorporate sequentially additional functionalities.

Overall the multitude of polymer – nanoparticle combination possibilities achievable by scalable flamespray pyrolysis (Strobel, 2007) and high filler fractions with homogeneous distribution distinguish the current nanocomposite synthesis technique.



Figure 1. Magnetically-actuated multi-layer superparamagnetic nanocomposite film by a magnet at the top right (A). Vertical cut of that film showing the 3 homogeneous nanostructured layers of Fe_2O_3 (light grey) within the PMMA (dark grey) matrix (B).

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