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ELECTRONIC AND OPTICAL HYBRID MATERIALS VIA SELF-ASSEMBLY AND NANOIMPRINT LITHOGRAPHY

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Polymer/nanoparticle hybrid materials organized at the local level by additive driven self-assembly and patterned at the device level using nanoimprint lithography offer versatile approaches to the solution-based fabrication of nanostructured devices. Block copolymers have long held promise for templating periodic functional materials, however the ability to fabricate well-ordered composites with high loadings of nanoparticles and with domain sizes large enough to be useful for the manipulation of visible light has remained challenging. We demonstrate the formation of well-ordered nanocomposites with tunable magnetic and optical characteristics containing up to 70 wt. % of metal, metal oxide and/or semiconducting nanoparticles through phase specific interactions of the particles with the block copolymer templates. In one example metallodielectric 1D photonic crystals with controlled domain spacings between 120 nm and 260 nm were prepared using amphiphilic (polynorbornene-g-polystyrene)-b-(polynorbornene-g-polyethylene oxide) brush BCPs as the templates and hydrogen bonding as a driving force for the selective incorporation of gold NPs into hydrophilic domains at gold core loadings of up to 50 wt.%. In a second example, we prepared composites with tunable magnetic permeability and high Verdet constants by phase selective incorporation of FePt and ZrO2 nanoparticles in poly(styrene-block-2-vinylpyridine) (PS-b-PVP) diblock copolymer templates in collaboration with the Norwood Group at the University of Arizona. The magnetic character of the composite was tuned by controlling the FePt particle size (2-12 nm) and loading within the PVP domains while the refractive index of the composite was tuned by controlling the loading of zirconia nanoparticles. The small size and excellent dispersion of the NPs in the block copolymer domains provide very good optical transmission. Applications include high performance magnetic field imaging sensors and optical isolators. Nanoimprint lithography (NIL) offers high precision patterning of structures as small as 50 nm using wafer-based or roll-to-roll process platforms, however current resist systems offer little functionality. We developed hybrid UV-NIL resists containing up to 90 wt. % nanoparticles with excellent optical transparency for direct patterning of device structures including a readily scalable print, lift, and stack approach for producing large-area, 3D photonic crystal (PC) structures and optical gratings. Grating structures composed of highly filled NP polymer composite resists with tunable refractive indices (RI) between 1.58 and 1.92 at 800 nm were prepared using hybrid resists composed of anatase titania (TiO2) NPs, between 5 and 30 nm in diameter, and a UV-curable acrylate host. The gratings were robust and upon release from a support substrate were oriented and stacked to yield 3D PCs. The grating structure dimensions, line width, depth, and pitch were easily varied by simply changing the imprint mold. A six-layer log-pile stack was prepared using a composite resist containing 50 wt. % TiO2 NPs with an RI of 1.72 and yielded up to 72% reflection at 840 nm. Extension of these approaches to other materials systems and applications will be discussed.

References: 1. Song, D-P; Lin, Y; Gai, Y; Colella, N; Li, C; Liu, X-H; Gido, S; Watkins, J.J. “Controlled Supramolecular Self-Assembly of Large Nanoparticles in Amphiphilic Brush Block Copolymers” *JACS* **2015**, 137, 3711. **2.** Song, D. P.; Li, C.; Colella, N. S.; Lu, X.; Lee, H.-W.; Watkins, J. J. “Thermally Tunable Metallodielectric Photonic Crystals from Self-assembly of Brush Block Copolymers and Gold Nanoparticles” *Advanced Optical Materials*, in press **2015** DOI: 10.1002/adom.201500116. **3.** Howell, I.R; Li, C; Colella, N; Ito, K; Watkins, J.J. “Strain-Tunable One Dimensional Photonic Crystals Based on Zirconium Dioxide/Slide-Ring Elastomer Nanocomposites for Mechanochromic Sensing” *ACS Applied Mats & Interfaces* **2015** 7(6), 3641-3646. **4.** Beaulieu, M.R.; Hendricks, N.R.; Watkins, J.J.; “Large-Area Printing of Optical Gratings and 3D Photonic Crystals Using Solution-Processable Nanoparticle/Polymer Composites” *ACS Photonics* **2014** 1,(9) 799-805.