High Refractive Index TiO_2 Nanoparticle/Silicone Composites

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Motivation—There is a great need for moldable and processable high index of refraction (n) optical materials. Optical silicones have excellent material properties but are limited to n ~ 1.5. Increasing the refractive index of optical silicones (and being able to fabricate silicones with a range of refractive indices) will make a dramatic impact in a range of applications including optics for space environments and optical encapsulants. One common application that routinely uses silicone optical encapsulants is high brightness light emitting diodes (LEDs).

Accomplishment—In an effort to increase the index of refraction of optical silicones through the incorporation of inorganic materials, we have developed a new synthesis route for TiO_2 nanoparticles. Nanoparticles are an excellent choice for incorporation into optical encapsulants because their small size will not scatter visible light. Titania's material properties are ideal for this application because it is transparent to visible light and has a high refractive index (n=2.7).

Our synthesis is based on the hydrolysis of titanium isopropoxide which in the past was not a viable method for producing nanoparticles. Through careful control of the reaction conditions and by slowing the hydrolysis rate, we have successfully achieved nanoparticle growth. The nanoparticles can be synthesized with both isopropanol and trimethoxy(7-octen-1-yl)silane (a silane molecule terminated with a vinyl group) surfactants. These ligands can be replaced with other molecules if desired. The TiO_2 nanoparticles produced by this reaction have a small diameter (~ 5 nm) and a narrow size distribution, as can be seen in the transmission electron micrograph displayed in Figure 1. As a consequence of their small size, the particles do not scatter light with a wavelength > 350 nm, as determined from their UV-vis absorption spectrum (inset of Figure 2).

Since these nanoparticles are synthesized using surfactants, they do not form large agglomerates that would scatter visible light. (Commercially synthesized titania nanoparticles, which are typically fabricated through the spray hydrolysis of titanium tetrachloride and without the use of surfactants, contain particles as large as 50 nm in diameter and form agglomerates that can approach a μm in size.) The surfactants on the surface of our nanoparticles have an additional benefit in that they provide useful functionalities which allow the particles to be incorporated directly into a silicone matrix. The vinyl termination group on the trimethoxy(7-octen-1-yl)silane ligands is identical to the termination group on many commercial optical silicones. This allows our nanoparticles to disperse very uniformly and crosslink directly into a silicone matrix. Initial measurements of titania/silicone composites made by spin coating up to 5 wt.% TiO_2 in silicone show that we can achieve refractive indices as high as 1.9. (The final concentration of TiO_2 is higher in the final composite film than in the solution used to cast it.)

Significance—The highest refractive indices achieved in these optical composites could enable us to double the light extraction from high brightness LEDs. Controlling the mass loading of titania nanoparticles in silicone matrices allows for tuning of the composite's refractive index, making this technology also useful in other applications, such as flexible optics.

Sponsors for various phases of this work include: Laboratory Directed Research & Development

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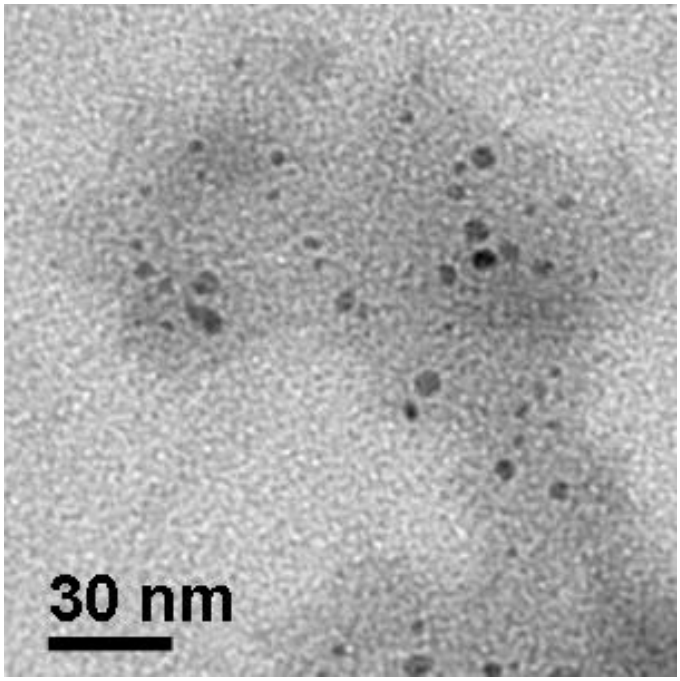

Figure 1. Bright field transmission electron micrograph of TiO_2 nanoparticles (dark spots of the image).

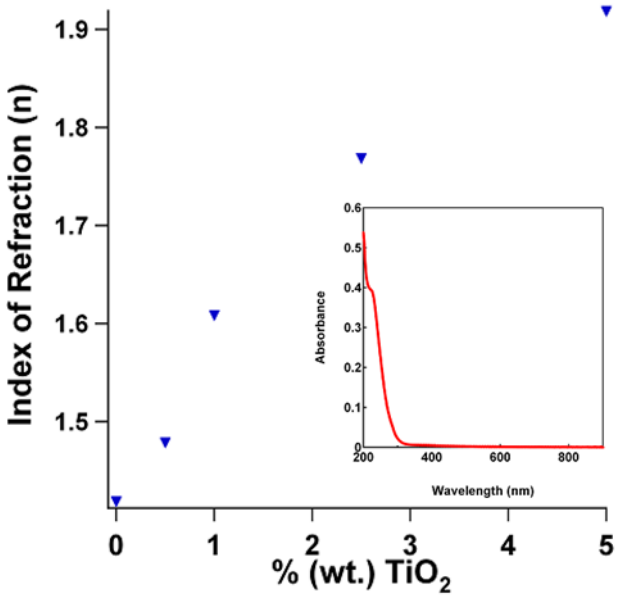

Figure 2. The index of refraction (n) of TiO_2 nanoparticle/silicone composites as a function of % (wt.) of titania in the solution used to spin coat the composites. The inset displays the UV-vis absorption spectrum for TiO_2 nanoparticles used to form the composites.