Nanocomposites for novel holographic applications

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Nanozeolites incorporated into photopolymerizable holographic recording materials can be used for holographic sensing, data storage, and optical systems design.

Recent advances in the design of lasers and of novel adaptable holographic recording materials have contributed to the rapid growth of new holography applications. Examples include holographic sensors and optical memories, holographic optical systems for the non-destructive testing of mechanical and biological objects, as well as holographic electro-optical switchable and security devices, all of which are expected to significantly impact our daily lives.

In this context, the study of photopolymerizable materials has long generated major interest, due to their high sensitivity, high spatial frequency response, and ease of processing into layers. Recently, various investigators have demonstrated that the addition of solid inorganic nanoparticles to different photopolymer materials improves the holographic properties of the resulting nanocomposite. A main drawback of these materials, however, is their significant scattering. By using porous nanoparticles, we have recently obtained layers characterized by relatively low scattering and improved properties.

Holographic recording in photopolymerizable materials is accompanied by light-induced mass transport of photopolymer components. This well-known effect prompted several research groups to base new approaches for improving material properties on the underlying processes. A basic formulation for a photopolymerizable nanocomposite requires introducing nano-dopants with significantly different refractive index from that of the host material. Redistribution of the nano-dopants during the holographic recording phase must also be achieved with significant increase in the ultimate light-induced refractive index modulation (see Figure 1). The presence of nano-dopants also leads to a significant decrease in the shrinkage of the nanocomposite during holographic recording.

**Figure 1.** The light-induced redistribution of photopolymer components is caused by the diffusion of monomers driven by a concentration gradient from dark to bright fringe areas and by the diffusion of short polymer chains in the opposite direction.

**Figure 2.** Scanning electron microscopy image of the Si-MFI nanoparticles used in our studies. The bar size is 20nm and the openings in the zeolite structure are 5.3 x 5.6 Å; 5.1 x 5.5 Å.

Our nanocomposite is an acrylamide photopolymer doped with nanozeolites, which are porous materials with a well-defined...
crystalline structure and pore size whose chemical structure is easily modified. In our studies, we use silicalite nanozeolites with a MFI-type structure (see Figure 2) that have a refractive index significantly lower than that of the photopolymer. The properties of this nanocomposite were studied by real-time monitoring of the holographic recording process. Shrinkage was measured by evaluating the properties of slanted gratings.

Figure 3 shows the real-time growth of the refractive index modulation induced during the recording of a grating with a spatial frequency of 1000 lines/mm in a 40µm-thick photopolymer layer doped with nanozeolites characterized by an average diameter of 60nm. A 6wt.% concentration of the nanoparticles doubled the refractive index modulation from 1.8x10^{-3} to 3.5x10^{-3}. Nanocomposite shrinkage was reduced and found to depend on the thickness of the layers. A shrinkage of 0.1% was observed in a 650µm-thick nanocomposite layer (see Figure 4).

We also performed energy dispersive X-ray spectroscopy studies on our holographic gratings. Results show that a redistribution of zeolite nanoparticles takes place during the recording process. The occurrence of redistribution effects suggests that these materials could be used for the design of novel holographic sensors since the local concentration of nanoparticle dopant could be controlled on a sub-micron scale.

Zeolites can adsorb different chemicals to change their refractive index. Such a grating could change its diffraction efficiency when the zeolite nanoparticles adsorb a specific chemical as illustrated in Figure 5.

Figure 3. Dynamics of recording in 40µm-thick layers at a spatial frequency of 1000 lines/mm with a recording intensity of 5mW/cm². The concentrations of the Si-MFI nanoparticles are as follows: 0wt.% (dash-dotted black), 1.5wt.% (solid light grey), 5wt.% (solid dark grey), 4.5wt.% (solid grey), and 6wt.% (solid black).

Figure 4. Angular selectivity curves for a photopolymer containing 4.5wt.% of Si-MFI in the dry layer (black trace) and for a non-doped sample (grey). Sample thickness is ~650µm.

Figure 5. Operational principle of a holographic sensor recorded in a zeolite nanocomposite.

Increased dynamic range and suppressed shrinkage, combined with a relatively low (<6% in 40µm-thick layers) level of scattering, make zeolite nanocomposites promising candidates for the development of holographic memories. Nanozeolite redistribution can also be utilized in holographic sensor design. To determine the exact mechanism responsible for the improved nanocomposite properties, we are planning to study the interaction between the photopolymer host and the zeolite nanoparticles using Raman spectroscopy. Transmission and scanning electron microscopy are also being used to study nanoparticle redistribution in the material volume and nanocomposite surface.

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