NIRT: Highly Integrated Optical Nanoparticle-Based Sensing Systems Based on Nanoparticle Synthesis, Assembly, and Integration

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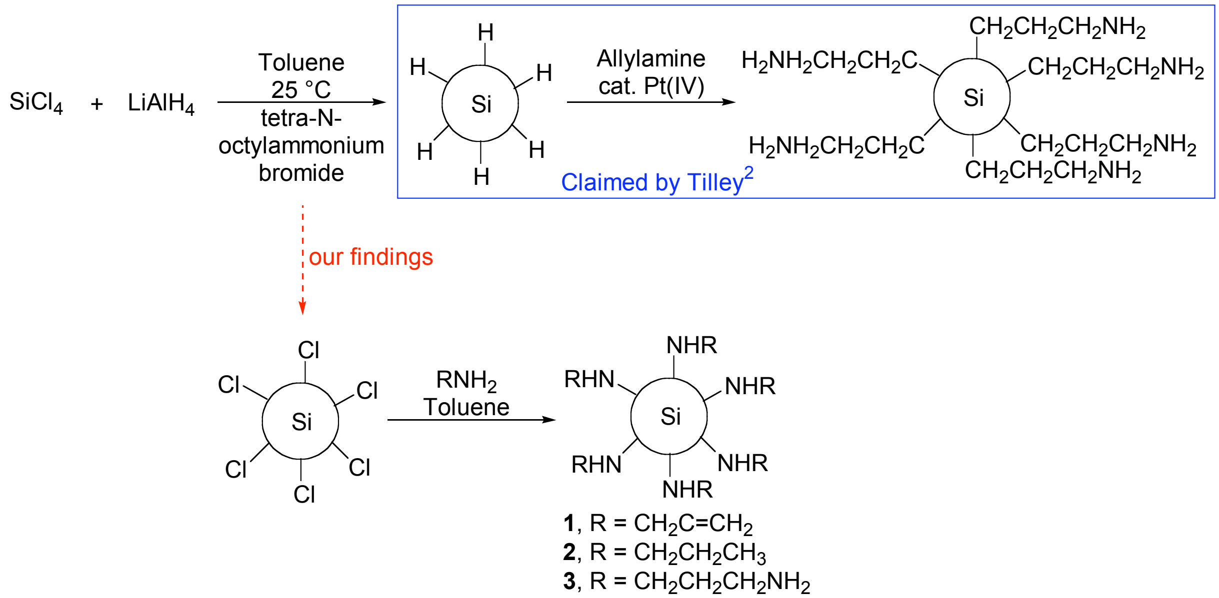
Biological and chemical sensor systems are of great importance for monitoring environmental conditions, warfare threats and medical conditions. While great strides have been made in sensor technology over recent years, sensors are still too large, too expensive and too analyte - specific for widespread and easy use. We are currently investigating the underlying science and system engineering to create new sensor-based microsystems. These chip-scale systems build upon new means of nanoparticle, compound semiconductor, and silicon (Si) materials integration, and will yield improvements in performance gained from on-chip integration of multiple functions, including advanced signal processing.

Our research is aimed at creating a planar, integrated optical and electrical system, which integrates thin film light emitting diodes (LEDs), dielectric channel waveguides, Si nanoparticles and photodetectors onto a Si CMOS signal processing electronics platform. Various applications can be addressed with fluorescence-base sensing as a key example. Materials science advances are required to achieve device-quality materials that can be integrated onto Si and within waveguides, including strain engineering and defect control. In this work, we are exploiting nanostructured materials as both multi-spectral emitters and robust targeted probes. Nanoparticles are increasingly important for sensing and offer a range of optical modalities. Optical signals from nanoparticles are often significantly enhanced over bulk materials allowing for miniaturization of the entire sensing system. Nanoparticles will be used in different functions: as light emitters to enable sensing and as molecular beacons to enhance signals generated during sensing. The efficacy of this integrated approach will be studied the use of these sensing\detection media to recognize tagged molecular and endogenous fluorescence targets. In addition, our work will significantly advance GaN materials and devices technology by enabling the integration of GaN multi-quantum well (MQW) based devices directly onto Si and directly into polymer waveguides for planar lightwave circuit applications.

One major goal in the past year (Winter) was to develop synthetic approaches to monodisperse silicon nanoparticles that luminescence in the visible region with a high quantum efficiency. In addition, the synthetic route should allow desired surface functional groups to be introduced easily. Scheme 1 outlines our synthetic approach to the silicon nanocrystals that is based upon the original report of Tilley.[2] We have found that the synthesis of the silicon nanocrystals does not proceed by intermediate hydride-terminated silicon nanocrystals as suggested by Tilley (shown in a blue box in Scheme 1), and instead appears to proceed through chloride-terminated silicon nanocrystals. Our findings greatly simplify the synthesis of the silicon nanocrystals and allow a range of surface groups to be easily introduced. For example, we have prepared silicon nanocrystals containing 1-amidopropyl (**2**) and 1-amido-3-aminopropyl (**3**) groups by using the parent amines in the general synthesis. Initial TEM and luminescence spectra of silicon nanocrystals **2** and **3** demonstrate ~2 nm silicon nanocrystals cores with luminescence maxima at 473-475 nm. Thus, silicon nanocrystals **1**-**3** appear to have identical inorganic cores and differ only in the surface organic groups.

Scheme 1. Synthesis of Si Nanoparticles



Our modification of the Tilley silicon nanocrystals synthesis allows us to prepare 1.5-2 nm nanocrystals that are highly luminescent and possess a narrow luminescence band. In addition, the particles are air stable, water stable, and water soluble. The surface organic groups can be easily introduced, and the approach allows for wide variations in the nature of the surface groups

The design of the waveguide structures (Kuech) was initially carried out using a commercial modeling program. The light power distribution in the multilayer waveguide is determined by the thickness and the refractive index of each layer. The photonic simulation software was utilized to simulate the optical power distribution within the waveguide to optimize multilayer waveguide structure. The waveguide is designed such that most of the optical power is confined in the layer with higher index (ZrO_2 or HfO_2) where the nanoparticles will be embedded. In other words, the waveguide will be designed to produce the maximum coupling between the nanoparticles within the waveguide. A sol-gel technique was used to fabricate the multilayer waveguide. The thickness of the film is controlled by the spinning parameters (spin rate and spin time), which will also have an effect on the uniformity of the film. The refractive index of the film is controlled by the aging time of the sol-gel solution. There are many Si-compatible oxides which are transparent over the visible to the near-UV and possess a range of refractive index spanning from 1.48 for SiO_2 to over 2.3. We can change the composition of the sol-gel to get a series of films with different refractive indices. We have used commercially available CdS-based nanoparticles as a test system. The efficiency of the nanoparticle luminescence drops with processed in the sol-gel relative to the nanoparticles in toluene solution. The surface protection on the CdS particles appears not to be sufficiently robust to survive in a polar solvent. We have begun to use Si-based particles as well.

Finally, we are also focusing efforts on the creation of integration processes (Brown, Jokerst, Brooke) for removing GaN LEDs from growth substrates. These techniques include photoelectrochemical (PEC) wet etching of n-GaN to remove it from sapphire and the synthesis of GaN on Lithium Aluminate: a substrate that is readily directly etched from GaN.