Aerosol synthesis of silicon germanium hybrid and alloy nanoparticles

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Nanocrystalline germanium (Ge) and silicon (Si), as well as the GeSi alloy are promising materials for the application in printable electronics. Dispersed in organic liquids and deposited as thin functional film the nanocrystals (NCs) can be utilized in electronics, optoelectronics or photovoltaics. The synthesis of freestanding NCs is challenging and can for example be achieved in a nonthermal plasma reactor (Pi and Kortshagen, 2009). We report on our progress in synthesizing Si and GeSi NCs in a reactor system utilizing two consecutive hot-wall reactors.

The setup allows to produce silicon nanoparticles (NPs), GeSi alloy NPs and anisotropic hybrid particles from Ge and Si (Ge@Si NPs). The first reactor stage (HWR I) can be used for Si NP synthesis via monosilane (SiH₄) pyrolysis in Argon carrier gas. GeSi alloy particles of variable composition are produced by feeding SiH₄ and monogermane (GeH₄) into the reactor simultaneously in varying ratios. After the first reactor stage the aerosol is quenched by additional argon. Prior to the second reactor stage (HWR II) GeH₄ can be fed into the system. As a consequence, Ge growth at the NPs from the first stage (e.g. Si NPs) takes place at temperatures of 700°C to 900°C. At the reactor exit the aerosol is quenched by nitrogen. The particles are collected with a membrane filter or sampled via a low pressure impactor by deposition on TEM grids or silicon wafers.

Figure 1 shows SEM images of Ge@Si NPs. GeSi-700-b is synthesized at 700 °C, GeSi-600-c at 600 °C in HWR II. Ge appears bright due to its higher density compared to silicon (dark material). It is observed, that at lower temperature the Ge grows in several patches, while at higher temperatures predominantly one patch is observed at the Si NP surface.



Figure 1. STEM image of Ge@Si NPs. Insets show higher magnification

This is explained by differences in surface diffusion. At higher temperatures higher diffusion leads to decreasing surface supersaturation of Ge on the SiNP surface in a zone around a Ge patch, thus supressing nucleation of further patches. At lower temperatures surface diffusion is lower and several patches can nucleate (Markov, 1971).

Figure 2 shows a high resolution TEM image of exemplary GeSi alloy particles. They are spherical and monocrystalline. Their mean size is 26 nm with a geometric standard deviation (GSD) of 1.25. They are composed of 1.3 at% Si and 97.7 at% Ge, as XRD and EDX measurements show in good agreement. Since Ge and Si are fully miscible over the whole composition range, particles of every composition are accessible by varying the ratio of GeH₄ and SiH₄ in the precursor feed.



Figure 2. HRTEM image of GeSi alloy NPs

The progress on the synthesis of the germaniumsilicon hybrid particles will be demonstrated, regarding the influence of the process parameters on their morphology, composition and growth. Furthermore the tailoring of GeSi alloy nanoparticle morphology and composition as function of process parameters mentioned above will be demonstrated.

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