Reactive sputter-driven synthesis of transition metal nanoparticles and nanofluids

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Human dependence on non-renewable energy sources is causing a global energy crisis. Possible solutions include the transition to renewable energy sources such as the wind and the sun. Here, we present the gas-phase synthesis of low-dimensional materials based on transition metal nitride nanoparticles (MeN NPs, Me = Ti, Zr, Hf, Ta, and Cu) that might be potentially suitable for solar harvesting or plasmonic sensing [1-3]. In contrast to conventional thin-film deposition, we show that reactive magnetron sputtering of metal targets in Ar/N_2 can be adapted to synthesize MeN NPs in the gas phase, avoiding high temperatures and toxic chemicals. Under proper conditions, highly crystalline and stoichiometric MeN NPs can be produced, with optoelectronic properties suitable for use in solar-driven electrochemical water splitting and as direct solar absorbers. Both planar and cylindrical magnetrons can be used for this purpose. Furthermore, we show that this method allows for direct deposition of MeN NPs into vacuum-compatible liquids to produce linker- and residual-free plasmonic nanofluids.

The plasmonic sensitivity of MeN NPs was found to exceed that of Au; however, they suffer from partial oxidation after contact with air. To overcome this issue, we developed a method to passivate Me NPs using rf magnetron sputtering of Si_3N_4 for the in-flight deposition of SiN protective layers over MeN NPs without contact with air. The SiN shell tunes the position of localized plasmon resonance (LSPR) of the MeN NPs from 580 to 850 nm by tuning the porosity and, consequently, the effective refractive index of SiN. Although the plasmonic sensitivity becomes attenuated, the SiN layer protects MeN NPs from post-deposition oxidation in air and preserves LSPR at temperatures above 400°C. Thus, our method offers an environmentally benign, annealing-free route to MeN NPs and nanofluids with controllable optical properties, enhanced thermal stability, and promising features for novel applications.

References

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