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In-situ scrutiny of the transition melting of pristine (Au) and metal oxide (Au-gallia)-supported plasmonic nanostructures

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ABSTRACT

Keywords: Thermal stability In-situ monitoring Plasmonic nanostructures Theoretical models Gold and gallia Morphological and compositional stabilities of pristine and metal-oxide-supported gold/gold-gallia nanostructures were determined at elevated temperatures (800 °C) for the first time. Emphasis was on the size and shape dependence of the transitional melting temperatures, from which conclusions were drawn predominantly from monitoring in-situ changes to plasmonic properties of the nanostructures. The morphological, crystalline, optical and elemental analyses supported and revealed new insights into temperature-driven morphological transformations. The experimentally-derived melting temperature of pristine nanostructures was corroborated using theoretical melting models, with the best results for the liquid skin melting model. The shape deformations and thermal expansion of pristine nanostructures were precluded by reinforcement of the plasmonic Au nanostructures with a matrix material of gallium oxide, which gave markedly different results in terms of enhancing thermal stability at temperatures where the pristine nanostructures were completely vaporized. The interfacial energies were likely the reason for the enhanced stability against shape transitions and/or melting, wherein it was found that the formative steps of gallia were insufficient to achieve these effects and that complete phase formation was necessary for stabilization. The insights concerning the reasons for the enhancements, the supportive role of the metal oxide matrix, the plasmonic stabilities of the Au nanoparticles, the shape dependence of the melting transitions and the crystalline stabilities of the composites in this report will help in predictive composite design for high-temperature applications such as sensing, catalysis, energy storage and design of compatible optical communication devices.

1. Introduction

"There's plenty of room at the bottom," a notable lecture by Richard Feynman, has led to progress in the scientific community regarding nanotechnology [1]. Today we are able to tailor individual atoms and molecules seemingly at will, with a wide range of beneficial applications for society such as in energy [2], environment [3], quantum computing [4] and space research [5] due to the many exceptional physical, chemical, electronic and optical properties [6]. Among the various divisions of nanoparticle chemistry, noble metal nanoparticles (MNPs: Ag, Au, Pt and Pd) have unique magnetic, optical and electronic properties [7]. Among the many properties, light manipulation in these nanoparticles made possible over a wide gamut of wavelengths due mainly to the property of surface plasmon resonance makes them interesting, as confirmed by multiple studies [8]. Our studies have demonstrated precision control of this property through patterning [9], as well as pioneering observations on harvesting of radiant energy present in a high-temperature environment of 500 °C [10] and even down to 300 °C [11,12] with the prospect of realizing optical sensors without a separate light source. Owing to this, they have promise in multiple fields such as catalysis [13], imaging [14], drug delivery [15] and sensors [16] that can operate from room temperature (RT) to >500 °C. Among these applications and among plasmonic MNPs, gold nanoparticles (AuNPs) are often of interest in sensing due to their catalytic activity, stability against chemical poisoning and established procedures for morphology control - hence they suitable even for harsh-environment sensing applications as in combustion and environmental monitoring [17]. However, at elevated temperatures, due to their small size, even AuNPs are prone to agglomeration and inter-material diffusion, leading to loss or reduction of reactivity [18]. In this report we present a study on the changes in melting point and the enhancements to stability (against changes in morphology and/or melting) of gold nanorods (AuNRs) and gold nanospheres (AuNSs) when present alone or as a composite with support from a metal oxide such as gallium oxide (GaO).

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