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To Study the Effect of Temperature Variation in Optical and Structural Properties of Nano-Composites Formed by 10% Doping of Bismuth Oxide in Copper Oxide

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ABSTRACT: CuO-doped Bi_2O_3 and undoped Bi_2O_3 were prepared using tartaric acid assisted co-precipitation to be applied as nano-photocatalysts for shape controlling Al_2O_3 ceramics. The prepared powders annealed at 300 °C were characterized using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), ultraviolet–visible absorbance spectroscopy (UV–vis) and photoluminescence (PL) spectrophotometry. According to the XRD results the undoped sample consisted of pure $\beta\text{-Bi}_2\text{O}_3$ while the doped one consisted of $\beta\text{-Bi}_2\text{O}_3\text{-CuBi}_2\text{O}_4$. The prepared Bi_2O_3 powders were characterized by band gap energy values ranging from 3.0 to 2.3 eV. The developed CuBi_2O_4 was characterized by band gap energy of 1.3 eV. Al_2O_3 ceramics were prepared using a modified polyol method. A marked variation in the microstructure of Al_2O_3 ceramics treated with the different prepared nano-photocatalysts was observed. This variation was referred to the free motions of photocarriers that were the backbone in varying the size, composition and morphology of the particles.

KEYWORDS: nanocomposites, doping, bismuth oxide, copper oxide, temperature, optical

I. INTRODUCTION

In recent decades, semiconductor nano-photocatalysis has attracted increasing attention for the solar energy harvest and environmental remediation [1]. Among these semiconductors, bismuth trioxide (Bi_2O_3) and Bi-based oxides have been studied due to their unique properties like large band gap, high oxygen ion conductivity, remarkable photoconductivity and good photoluminescence properties [2]. Bi_2O_3 has been widely used in gas sensors, varistors solid oxide fuel cells, optical coating, electronic ceramics, optoelectronics equipment's, high temperature superconductors and catalysts [1], [3], [4], [5], [6]. Bi_2O_3 has five main polymorphic forms denoted by: $\alpha\text{-Bi}_2\text{O}_3$ (monoclinic), $\beta\text{-Bi}_2\text{O}_3$ (tetragonal), $\gamma\text{-Bi}_2\text{O}_3$ (body centered cubic), $\delta\text{-Bi}_2\text{O}_3$ (cubic) and $\omega\text{-Bi}_2\text{O}_3$ (triclinic) [2], [7]. Among these polymorphic forms $\beta\text{-Bi}_2\text{O}_3$ has the strongest absorption in the visible light region because it has the smallest band gap (~2.4 eV) and has demonstrated better photocatalytic performance than other phases [7], [8]. As the photocatalytic activity of photocatalysts closely interrelates with their sizes, morphologies and microstructures, therefore it is highly desirable to fabricate both doped and undoped Bi_2O_3 with high activity and controllable morphologies that are cost effective and allow large scale production from the practical point of view [9], [10], [11]. In line with the philosophy of green chemistry, an increase in demand was needed to replace the hazardous chemicals by more environmentally friendly alternatives [12]. So it was preferred to prepare Bi_2O_3 , a non-toxic material [13], using green methods. Co-precipitation method could be considered as one of the methods that is easy to process, energy saving, environmentally friendly and cost effective as compared with the other methods [14]. There were several technical challenges for wide applications of photocatalysts, including: 1-catalyst optimization to improve quantum yield or to utilize visible light, 2-efficient photocatalytic reactor design and catalyst recovery/immobilization techniques, 3-better reaction selectivity [15]. Especially the practical applications were mainly restricted to photocatalytic water/air purification, hydrogen production from splitting water and high efficiency/low cost solar cells [16]. In the past decades, shape control of nanostructures has been proved extremely important because a wide range of physical and chemical properties of nanostructures depended primarily on their sizes and shapes [17]. The use of Bi_2O_3 as a green photocatalyst for modifying nano-ceramic microstructures has not been discussed before and I tried to spotlight on this new trend. In this study doped Bi_2O_3 with CuO and undoped Bi_2O_3 were prepared using tartaric-assisted co-precipitation technique and applied as photocatalysts for modifying the microstructure of Al_2O_3 ceramics.



Al₂O₃ ceramics were prepared using a modified polyol method that is considered as one of the most important derivatives of sol-gel method and has the advantages of simplicity, low cost, relatively low reaction temperatures, and a green method [18]. The interaction of the different prepared photocatalysts with the prepared Al₂O₃ gel under UV light was investigated. A novel mechanism explaining this interaction was also interpreted.

II. DISCUSSION

Metal-oxide nanomaterials (MONs) have gained considerable interest in the construction of flexible/wearable sensors due to their tunable band gap, low cost, large specific area, and ease of manufacturing. Furthermore, MONs are in high demand for applications, such as gas leakage alarms, environmental protection, health tracking, and smart devices integrated with another system. In this Review, we introduce a comprehensive investigation of factors to boost the sensitivity of MON-based sensors in environmental indicators and health monitoring. Finally, the challenges and perspectives of MON-based flexible/wearable sensors are considered.

Recently, flexible electronics including flexible sensors,¹⁻⁴ power sources,⁵ and conductors^{6,7} have been studied eagerly due to significant advantages in terms of reduced weight, flexibility, and durability to sustain multideformation (i.e., stretching, twisting, bending, and compression) compared with their rigid counterparts.⁸⁻¹⁰ During this vigorous development, flexible/wearable sensory systems have also been studied with various sensing materials including carbon materials,^{2,11-13} biomolecules,^{14,15} conductive polymers,^{2,16,17} and inorganic nanomaterials^{1,18,19} to improve their sensitivity, functionality, and stability via diverse strategies.

Among these materials, metal-oxide nanomaterials (MONs) are promising candidates in diverse areas of chemistry, materials science, physics, and biotechnology.²⁰ The unique electronic structure determines the conductor, semiconductor, and insulator properties of nanomaterials. Transition metal ions typically possess unfilled d-shells, allowing for reactive electronic transitions, wide band gaps, superior electrical characteristics, and high dielectric constants.²¹ As a result, MONs have exceptional and adjustable optoelectronic, optical, electrical, thermal, magnetic, catalytic, mechanical, and photochemical properties. Furthermore, MONs have a wide range of applications: sensors, fuel cells, batteries, actuators, supercapacitors, optical devices, pyroelectrics, piezoelectrics, ferroelectrics, and random access memory due to their exceptional shape and size, which manifest distinctive physicochemical properties. Potential applications also exist due to the high density of surface sites and nanostructures.²¹

Especially, for sensing materials, MONs, mainly copper(II) oxide (CuO), copper(I) oxide (Cu₂O), tin(II) oxide (SnO), tin(IV) oxide (SnO₂), zinc oxide (ZnO), nickel oxide (NiO), indium oxide (In₂O₃), and tungsten oxide (WO₃), are promising candidates that manifest high sensitivity, fast response/recovery (res/rec) time, excellent reproducibility and stability, and cost-effectiveness with simple fabrication processes.^{18,19} Conventional MON-based sensors are generally fabricated on rigid substrates due to their high manufacturing temperature, which increases energy consumption and manufacturing cost as well as imposes limitations in expanding applications.^{15,16,17} However, numerous studies on building MON-based sensors on flexible and stretchable substrates have been reported as a result of rapid advances in fabrication techniques in the past decade. As substrates for MON-based flexible sensors, polyimide (PI), polyethylene naphthalene (PEN), poly(ether sulfone) (PES), polyether ether ketone (PEEK), polycarbonate, polydimethylsiloxane (PDMS), and polyethylene terephthalate (PET) are preferred. The thermal and chemical resistance, flexibility, and transparency of materials should be addressed for pragmatic sensing applications.³³ Conductive features are essential to the operation of electrical devices, enabling the connection between various components.^{12,13,14}

We present an overview of MON-based flexible/wearable sensors in this Review. First, we go through some of the most prevalent sensing materials. Then we discuss the diverse manufacturing technologies for fabricating flexible/wearable sensors. The applications of flexible/wearable sensors in the field of environmental indicators (e.g., gas, chemical, pH, and humidity sensors) and health monitoring (e.g., biomaterial, respiration, temperature, and mechanical sensors) are introduced to gain a comprehensive understanding of sensing efficiency via the design strategies and advanced characterization techniques. Finally, we discuss the challenges that flexible/wearable sensors are currently facing as well as their potential development in the coming years.



III. RESULTS

Nanoparticles composed of metallic cores encapsulated in oxide shells emerged in the last decade as an attractive class of nanocomposite materials due to their high stability and unique properties provided by the high contact area between the metal and oxide components. Diverse metal-oxide interactions in metal@oxide core@shell nanoparticles enable tuning their electronic structure, spectroscopic properties, and surface reactivity for applications in sensing, electrochemistry, batteries as well as thermal and photocatalysis. Herein, we review the recent literature on the synthesis, characterization, simulations, and applications of metal@oxide nanocomposites. In particular, we discuss how the properties of metal@oxide nanoparticles can be tuned for a given application by changing the size of the metal core, the thickness and porosity of the oxide shell, as well as their composition, e.g. by alloying the core or doping the shell. Understanding of structure-property relations in metal@oxide systems provides vast opportunities for the rational design of advanced metal@oxide nanocomposites, making this class of materials promising for a wide range of applications.

Metal nanoparticles that can serve as cores for metal@oxide nanocomposites can be prepared through various well-known bottom-up strategies. For example, metal cores can be synthesized in the solution phase via the reduction of their corresponding metal salts, either by a reducing agent (e.g. NaBH_4) or by redox transmetalation of metal precursors (e.g. reduction of Ag salts by Cu nanoparticles to yield bimetallic Cu-Ag nanoparticles). Since nanoparticles have lower thermodynamic stability than the corresponding bulks, their formation is kinetically controlled. Hence, careful control of synthesis variables (e.g. temperature, reducing agent, and solvent) are often necessary to obtain nanoparticles of desired size and morphology. For instance, the size of Rh nanoparticles generated by the reduction of dissolved Rh(III) salts was shown to be directly related to the reduction potential of the reducing agent. In turn, tuning the precursor/seed concentration ratio allows one to control the branch length of branched Ni nanoparticles, whereas higher pressure during synthesis may change the structure of nanoparticles to a metastable hexagonal closed-packed phase.

One of the most common physical methods to synthesize metal@oxide core-shell nanocomposites is sputtering. For example, sputtering in an ionic liquid environment enabled the coating of small (around 2.5 nm) noble metal nanoparticles with ultrathin indium oxide shells with a thickness of around 1.5 nm. Another case is magnetron sputtering, where Ar^+ ions collide with a target material under a magnetic field that promotes Ar ionization and increases the deposition rate. The ability to control the plasma source, reactive environment, target materials, and geometry of the sputtering chamber provides multiple handles to control the properties of the final product. Similarly, in cluster beam deposition, metal nanoparticles are created by vaporization of the target through contact with a molecular beam, followed by condensation and growth in the gas phase. Alternatively, physical adhesion can be used to encapsulate catalysts with zeolites, which could be promising if sufficient control of the shell size and applicability to various core sizes is achieved. Beyond the deposition of metal nanoparticles onto a substrate, physical vapor deposition can simultaneously deposit metals and oxides of different materials by combining multiple targets in an inert gas environment. This technique can also produce thin films of metal nanoparticles embedded in ceramic oxides^{10,11,12}.

In precipitation and sol-gel syntheses, the metal core nanoparticles nucleate the growth of oxide shells from soluble precursors that gradually precipitate out of the solution. The rate and the extent of shell growth, and hence the uniformity and thickness of the resulting shells, are affected by the reaction time and temperature, concentrations of the nanoparticle and the precursor, as well as pH. Additionally, the success of this approach depends on the strength of electrostatic or chemical interactions and the extent of lattice parameters mismatch between the metal core and the growing oxide shell. If the interactions are too weak, shell growth will not occur unless the surface of the metal core is modified by surface primers. For example, silane coupling agents are required to facilitate the growth of a SiO_2 on citrate-stabilized gold nanoparticles. Conversely, if the interactions are too strong, inhomogeneous nucleation can occur leading to inconsistent coatings. This effect can be mitigated by deliberately slowing the rate of precipitation, for instance by employing a less reactive precursor.

In contrast, hydrothermal synthesis relies on continuous crystallization and dissolution yielding rather uniform core@shell structures. Typically, a sealed vessel (e.g. polytetrafluoroethylene-lined autoclave) containing a mixture of water, the core nanoparticles, and the shell precursors is heated over an extended period to create high autogenous



pressure. Under these conditions, sparingly soluble materials continuously grow, dissolve and recrystallize analogously to the geological formation of silicate minerals. This process of continuous and controlled equilibration allows highly uniform materials to be obtained in a simple and scalable, albeit sometimes slow, manner. For example, Pt@CeO₂ can be synthesized by hydrothermal treatment of an aqueous solution containing citrate-coated Pt nanoparticles, CeCl₃, and urea in an autoclave at 90°C for 24 hours. Although clear structure-property relationships remain elusive, the flexibility of the hydrothermal approach permits a wide variety of catalyst morphologies to be accessed through the addition of suitable structure-directing agents and/or in situ transformations. For example, the synthesis of metal nanoparticles encapsulated in hollow hierarchical single-crystal zeolite of ZSM-5 was recently achieved through two consecutive hydrothermal steps involving the formation of the zeolite from plain silica nanospheres, followed by in situ reconstruction into a metal@zeolite structure^{9,10,11}.

Physical methods such as atomic layer deposition use self-limited surface reactions under discontinuous gas flow to deposit oxide films layer by layer. For example, to deposit thin TiO₂ films, a Ti precursor such as TiCl₄ is first adsorbed onto a substrate or core; the excess gaseous precursor is purged with an inert gas before the same substrate is subjected to another gas (e.g. H₂O) that reacts with the adsorbed TiCl₄ to form the TiO₂ film. The primary advantage of atomic layer deposition over wet-chemical methods, despite its higher cost, is the linear relationship between shell thickness and the number of cycles, which facilitates obtaining highly uniform shells with precisely controlled thicknesses even at sub-monolayer coverages. Atomically dispersed oxides can also be selectively deposited on metal nanoparticles using sterically hindered precursors, leading to significantly improved selectivities, as demonstrated in the cases of Pt@FeO_x/SiO₂ for CO oxidation, and Rh@MoO_x/SiO₂ in the conversion of CO and H₂ into alcohols. Finally, as discussed above, wet-chemistry syntheses can often be adapted to alter the structure of the resulting product. For example, porogens such as decyltrimethylammonium bromide and cetyltrimethylammonium bromide can be added to introduce shell porosity in a controllable fashion, which can be desirable when exposing sites of the metal core is beneficial, as in catalysis. Additionally, sufficiently thick oxide shells can be doped to further tune their properties. Restructuring of thick shells upon heat treatment driven by a thermodynamically favorable interaction at the core-shell interface can also serve as an approach to access unique morphologies that are not easily obtainable by other methods. For example, Ni@SiO₂ with thick SiO₂ shells (around 11 nm) can be transformed by calcination in air and subsequent reduction in H₂ into a yolk-satellite-shell structure with voids between Ni and SiO₂ and concomitant formation of smaller satellite Ni particles. This transformation was found to be driven by the interfacial formation of nickel phyllosilicate that promotes the diffusion of Ni from the core throughout the shell to increase the available Ni-SiO₂ interface^{4,5,6}.

Characterization of metal@oxide nanocomposites

The available synthetic techniques enable the preparation of metal@oxide particles with a wide variety of structures, which can be understood through detailed experimental and computational characterization. Moreover, the characterization of metal@oxide nanocomposites allows one to establish structure-property relations, which are crucial to the understanding of the properties and application potential of the developed materials. However, metal@oxide systems are challenging to study with both experimental and computational techniques due to the intricacy of their structures, as well as processes and interactions on the metal-oxide interface. The wide range of experimental techniques available to characterize nanoparticles was reviewed recently. Here, we briefly discuss well-established as well as emerging techniques particularly relevant to the characterization of metal@oxide core-shell interfaces and challenges in their application, followed by the main strategies and recent advances from computational studies of metal@oxide particles.

Experimental characterization of metal@oxide nanoparticles

To visually confirm the existence of a core@shell morphology and determine the core size and shell thickness, transmission electron microscopy (TEM) has become a routine technique. Furthermore, high-resolution TEM provides information about the composition of crystalline materials through the measurement of d-spacings in their lattice. Coupling of TEM with energy dispersive X-ray spectroscopy^{1,2,3} (EDX), commonly referred to as TEM-EDX mapping, can provide spatially resolved number densities of each element for semiquantitative elemental analysis. This analysis is particularly useful when the visual contrast between core and shell materials is unclear from the micrographs, or when the presence of the expected elements in multi-component cores or shells needs to be confirmed. More recently,



precise mappings of the full strain tensor as well as the 3D coordinates and identities of individual atoms in a single nanoparticle have been achieved by tomographic reconstruction of TEM images .

Characterizing spectroscopic properties is important to understand key phenomena connected to the application of the core@shell particles, such as plasmonic resonance and light absorption . For example, the dependency of light scattering on the nanoparticle size can help to characterize the particle size distribution in the obtained sample and other features in the nanocomposite structure .Spectroscopic techniques can also be used to characterize the chemical environment and electronic properties of heavy atoms within the core@shell nanostructure. The most common spectroscopic techniques are X-ray photoelectron spectroscopy (XPS) and X-ray absorption spectroscopy (XAS), which can be further broken down into extended X-ray absorption fine structure (EXAFS) and X-ray absorption near-edge structure (XANES) depending on the energy of the incident X-ray. All these techniques can quantify individual contributions from multiple components by comparison to suitable reference materials and subsequent deconvolution of the spectra by linear combination analysis. XPS is often used to distinguish and quantify oxidation states and coordination environments. For example, small fractions of unreduced nickel phyllosilicate detectable by XPS have been used to explain the improved stability of phyllosilicate-based core@shell catalysts [Citation95]. In another example, XPS could reveal the increasing extent of formation of an interfacial 2:1 nickel phyllosilicate phase (blue deconvoluted peak) with increasing SiO₂ shell thickness .In some cases, XPS can also be employed to quantify average shell thicknesses (< 10 nm) in metal@oxide nanoparticles .Finally, charge transfer between core and shell can also be observed by XPS through shifts of the binding energies of various peaks .

Solid-state nuclear magnetic resonance (NMR) spectroscopy, aided by dynamic nuclear polarization to improve sensitivity, can be employed to determine the structures of nanoparticles or their ligands if the NMR-sensitive nuclei are present in the sample .Another promising nuclei-based technique is Mössbauer spectroscopy, which can provide information on the local environment and magnetic field around the nuclei. For example, this technique helped to characterize the structure of ligand-capped Sn@SnO_x nanoparticles containing concentric amorphous SnO and SnO₂ layers that could not be spatially resolved by other techniques^{5,6,7} .

For metal@oxide nanoparticles that are synthesized by the reduction of a metal oxide precursor, the reducibility of the precursor measured by temperature-programmed reduction under flowing hydrogen can indirectly provide qualitative information on core-shell (or metal-support) interactions. A significant shift relative to the bulk oxide in either direction is usually indicative of intimate contact between the oxide precursor and another component. A decrease in reduction temperature can be rationalized by close contact with a metal that has a high capacity for hydrogen activation and can catalyze the reduction of the oxide by hydrogen spillover .In contrast, an increase in reduction temperature in the presence of an oxophilic oxide such as silica, alumina, or a rare earth oxide can often be attributed to the formation of a metal silicate aluminate or mixed oxide phase with highly ionic and poorly reducible metal-oxygen bonds. The reduction of such material usually yields strongly bound, well-dispersed, metal nanoparticles.

However, the powerful characterization techniques discussed above also have some limitations. For TEM analyses, the main pitfalls that can lead to erroneous conclusions include (1) electron beam-induced sample degradation, and (2) human bias in extrapolating single-particle measurements to average properties of the entire particle set. The former can be mitigated by a judicious selection of measurement parameters or by coating the sample with a protective layer , whereas high-throughput image processing algorithms have been developed to avoid the latter .

In turn, the major limitation of XPS is the requirement of a high-vacuum environment. Although near-ambient pressure XPS measurements (under about 1 mbar pressure) have increasingly been explored, the issue of the XPS ‘pressure gap’ remains to be fully overcome . XANES generally provides information similar to that from XPS, except that it is a bulk-sensitive instead of a surface-sensitive technique, and can be performed fully in operando or in situ and time-resolved manners. For thick samples, discrepancies between XPS and XANES results can shed light on sample heterogeneity as a function of depth .Finally, EXAFS measurements can be fitted to yield detailed structural information, including the average coordination numbers and average interatomic distances. For instance, EXAFS can confirm phase assignments and provide insights into the interactions between core and shell materials, e.g. by quantifying the extent of interfacial site formation through the coordination numbers of metal and oxygen atoms . EXAFS analysis where the shell is shown to undergo different degrees of reduction upon application of an overpotential depending on its combination with a monometallic or a bimetallic core^{8,9,10} .



Atomistic modeling of metal@oxide nanoparticles

The number of computational studies of metal@oxide nanocomposites is rather limited due to their challenging nature. Currently, our theoretical understanding of metal@oxide particles is based on more established computational studies of periodic oxide films on single-crystal metal surfaces, which can represent very large metal@oxide nanoparticles. Furthermore, islands or patches of oxides supported on metal surfaces can model oxide-metal boundaries in core@shell particles with porous oxides or satellite structures. However, as particle size decreases and the nanoconfinement effects intensify, it becomes necessary to use discrete models including curvature, edges, corners, and other nanoscale features. The significant size of such models required to reliably represent metal@oxide nanocomposites represents one of the most significant challenges for computational studies of these materials. There are common challenges faced in the design of initial structures of metal@oxide particles, which are often obtained based on the best structures known for pure metal and oxide materials.

The available computational studies suggest that sizeable metal cores should favor polyhedral shapes obtained through Wulff construction or global optimization. These computational approaches also seem valid for metallic nanoalloys. In turn, the modeling of oxide nanoparticles is complicated by the presence of more than one element, which may lead to the formation of oxygen or metal vacancies in the nanoparticle as well as atoms with dangling bonds. Recently a few software packages have been developed to address these problems and obtain more realistic nanoparticle models for binary materials such as oxides^{11,12,13}.

Although generating an oxide shell with the desired stoichiometry and without low-coordinated atoms remains a challenge, the issue may be alleviated by the presence of the metallic core providing or accepting electrons to ensure charge balance within the nanocomposite. Additional challenges in the simulations of metal@oxide nanocomposites are related to the design of realistic interfacial structures, which can be rigorously addressed using computationally expensive global optimization techniques. To circumvent such high costs, machine-learning interatomic potentials can be developed for simulations of oxide-metal interfaces, although the high dimensionality of the fitting may complicate their application to multi-component materials. While applications of such methods to metal-oxide nanocomposites are scarce, their promising potential was shown in recent studies of heterostructures formed by metallic Cu and Hf with their oxides. Moreover, cores and shells with amorphous structures can be obtained from molecular dynamics simulations. For small particles, molecular dynamics simulations followed by quantum-mechanical optimization were used to study the optical properties of M@ZnO (M = Fe, Co, and Ni), where the oxide shell was created by removing atoms from a preestablished magic-size cluster of ZnO.

IV. CONCLUSION

The available computational studies of metal@oxide nanocomposites further illustrate the intricacies of modeling these materials. Indeed, the structures of oxide shells can differ substantially from those of bulk oxides or periodic oxide films. For example, ultra-thin Ag@ZnO films prepared via atomic layer deposition exhibit shells with wurtzite structure rather than the graphitic phase observed for ultra-thin films supported on extended Ag(111) single-crystal surfaces. Furthermore, the shell in metal@oxide nanocomposites can show an overall contraction or expansion, accompanied by displacement of certain atoms toward the core or vacuum region, as exemplified in calculations for Ag@ZnO. This seems analogous to the rumpling observed for periodic films; however, the distortion of the nanoparticle shell is further complicated by intricate atomic arrangements around the low-coordinated atoms at the edges and corners of the nanoparticle. Another complication is that the functionalization of the metal@oxide nanoparticles often produces structural discontinuities, such as pores. These defects can create further changes in the particle, such as the incorporation of atoms from the core into the vacancies of the shell region, which was noted on Pd@Fe₃O₄ particles.

Some atomistic insights into the structure and properties of interfaces in metal@oxide nanocomposites can be obtained from recent studies of partially oxidized metal nanoparticles composed of metallic cores encapsulated in shells of their native oxides. For example, the interactions between Al cores and Al₂O₃ shells were calculated to depend on the nanoparticle size and involve the diffusion of O and Al atoms between the core and the shell, which could be induced by the heating and melting of the nanocomposites. Furthermore, the thickness of Al₂O₃ shells on Al cores was shown to strongly affect the diffusivity of Al and O atoms on the interface as well as the melting point of the shell. Later studies showed that O mobility in M@MO_x (M = Fe, Ni, and Cu) depends strongly on the identity of the metal M and



its propensity to be oxidized. Thus, the mobility of O atoms is calculated to be one of the key factors for the formation and stability of metal@oxide nanocomposites with cores prone to oxidation.^{19,20,21}

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