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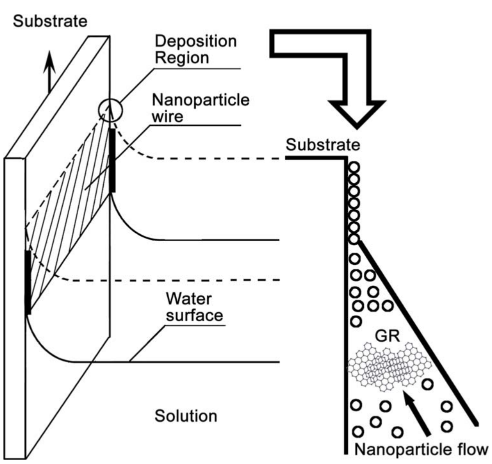
Pristine graphene modulation of vertical colloidal deposition for gold nanoparticle wires

Michael Crowe[a], Zhiqi Tian[a,b], Peng Zhang[c], J.J. Diao[a,*]

[a] Department of Cancer Biology, University of Cincinnati College of Medicine, Cincinnati, OH, 45267, USA
[b] Xi'an Jiaotong University, Xi'an, 710049, China
[c] Department of Chemistry, University of Cincinnati, Cincinnati, OH, 45221, USA

GRAPHICAL ABSTRACT



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ABSTRACT

Vertical colloidal deposition has emerged as an eco-friendly method for assembling nanoparticles into functional bulk structures. This technique is based on the coffee-ring effect, a natural dewetting phenomenon by which suspended particles in solution aggregate at a solid-liquid-gas interface. Significant research efforts have been devoted to developing and characterizing methods of modulating the coffee-ring effect, which offer additional degrees of control for vertical colloidal deposition. Functional nanoparticle conglomerates are often composed of gold nanoparticles, and studies of the coffee-ring effect have featured graphene materials. Moreover, some newly proposed technologies integrate graphene and gold nanoparticles. Here, we examine the effects of pristine graphene nanoflakes on vertical colloidal deposition for gold nanowires. Absorption measurements, for probing electrostatic changes, of colloidal mixtures of gold and graphene nanoparticles are presented, and the structural and electronic properties of the resulting wires are characterized. Our results suggest that pristine graphene modulates colloidal deposition of gold nanoparticles through jamming.

* Corresponding author.
E-mail address: jiajie.diao@uc.edu (J.J. Diao).

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1. Introduction

Nanoparticle research has garnered significant interest for the unique properties exhibited by these materials such as increased surface area and higher reactivity [1–3]. Synthesizing bulk structures from nanomaterials may extend such desirable properties to the macroscale for real applications [4,5]. For example, conglomerate structures have been developed for ultrasensitive biomolecule detection [6–8]. Electronic measurements with these systems are much easier to perform relative to biomolecule sequence detection modalities. More established approaches to nanoparticle assembly include lithography and inkjet printing, which allow for amalgamation into patterned geometries [9–13]. Single-step assembly methods like dielectrophoresis are attractive; however, they afford limited control over the resulting structure and may be experimentally onerous, as the dielectric constant between nanoparticles and the solvent must be precisely tuned for suitable results [14].

While these nanoparticle assembly techniques have their merits, environmental concerns have increased the demand for green manufacturing practices in order to reduce the use or generation of hazardous substances [15]. A more eco-friendly approach, known as vertical colloidal deposition (VCD), has emerged for the assembly of wires, thin films, and integrated arrays among other structures [16–18]. This method is based on the coffee-ring effect, a natural dewetting phenomenon by which suspended particles in solution tend to aggregate at a solid-liquid-gas interface as evaporation occurs [19] (Fig. 1a,b). Colloidal gold nanoparticles (AuNPs) have been used extensively in VCD studies because of their biocompatibility and the dependence of the electronic properties on capping molecules [20,21]. Even though hazardous substances may be used or generated to synthesize nanoparticle suspensions, VCD is regarded as more environmentally friendly as no additional hazardous chemicals or experimental conditions are required for bulk assembly.

Despite its simplicity, VCD offers precise control over the resulting structure. The width and height of the assembled nanoparticle wires are dependent on the temperature, deposition time, and other parameters that the user can easily manipulate [22]. Meanwhile, modulation and suppression of the coffee-ring effect have been researched extensively [23]. For example, using differently shaped nanoparticles may result in more uniform, film-like deposition rather than the coffee-ring effect [24]. The addition of surfactants like SDS can dramatically affect deposition patterns [25,26]. Light pulses [27], electrowetting [28], and polymer additives [29] are just a few of the other ways investigators have sought to modulate the coffee-ring effect. Adapting such techniques for VCD could confer additional degrees of control and other desirable traits for real applications.

Some studies of the coffee-ring effect have featured the carbon allotrope graphene [30,31]. This material consists of a monolayer of sp^2-hybridized carbon atoms arranged in a hexagonal lattice. Graphene has received considerable research interest in the last decade for its singular properties such as mechanical flexibility, strength, and high conductivity [15,32,33]. Numerous applications of graphene have emerged in medicine, optics, electronics, and other industrial fields [32,34–36]. Some of these proposed technologies integrate graphene and AuNPs [37–40]. However, no studies have examined the effects of pristine graphene (GR) on VCD-conducting nanoparticle wires from AuNP/GR dispersions.

In this letter, we examine the effects of GR nanoflakes on the

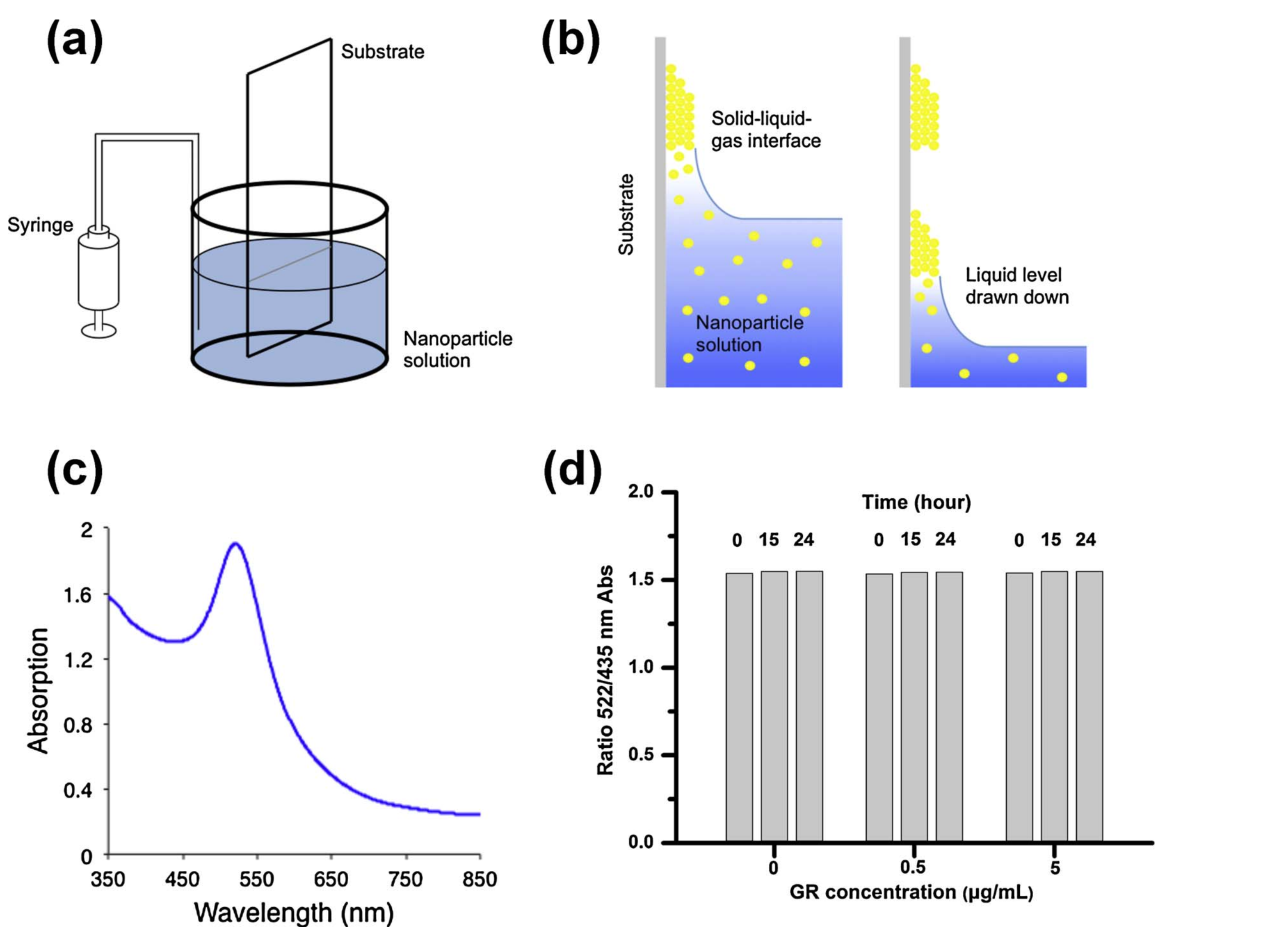

Fig. 1. (a) Illustrations of the general experimental setup for VCD and (b) a cross-sectional view of the substrate at the solid-liquid-gas interface as deposition occurs (c) Absorption spectrum of colloidal AuNPs. The peak at 522 nm is sensitive to colloidal aggregation while absorption at 435 nm is a background effect unaffected by aggregation. (d) The ratio of absorption at 522 nm to that at 435 nm for AuNP solutions containing GR. Over the course of 24 h, the ratio consistently remains slightly above 1.5 for all samples.