



Clam-inspired nanoparticle immobilization method using adhesive tape as microchip substrate

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ABSTRACT

Immobilization of the suspended nanoparticles is essential for many microfluidic applications. This work reports a novel biomimetic method to immobilize nanoparticles by using a common adhesive tape as the substrate of microfluidic chip. It mimics the clams' feeding system that utilizes the mucus (i.e., sticky fluid) to capture small phytoplankton particles in water. This work proves experimentally that this method has a better immobilization effect and a stronger shear stress resistance than the traditional methods using hard glass substrates. Moreover, we have applied this method to immobilize Au nanorods for the detection of R6G of various concentrations using the surface-enhanced Raman scattering (SERS) effect. This method enjoys several major merits: the sticky adhesive tape can seal the microfluidic structure easily, avoiding the bonding process; the immobilization is easy and environmental friendly, without the need for expensive reagents or complex processes; the adhesive tape substrate allows the flexibility of microfluidic chips; and the adhesive tape substrate can be stripped off for off-chip detection and can be replaced easily for the reuse of microfluidic structures. With these, the biomimetic method may find potential applications in environmental sensing, biocatalysis and biosynthesis using microchips.

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1. Introduction

Microfluidics presents great potential for high-throughput biological/chemical detection, catalysis and synthesis due to its inherent advantages, such as large surface-area-to-volume ratio, fast reaction rate, high-precision manipulation and easy flow control [1–5]. The corresponding detection-promoting substances catalysts or carriers are mostly nanoparticles or nanoparticle-involved compounds [6,7]. Actually, nanoparticles are highly preferred to be immobilized on the substrates since their separation and recycling from a mixture are laborious and troublesome [8]. Based on the working principles, the immobilization techniques can be broadly divided into four categories: magnetic attraction, covalent binding, polymer entrapment and surface adsorption. The first three methods function well, but mostly require complicated processes, expensive reagents/equipment, and even harmful chemicals. The surface adsorption method is simple and environmentally

friendly, but the adsorption amount is disappointing and the bond strength is often too weak to resist the high shear stress caused by the laminar flow in microfluidic devices.

Our mother nature has already evolved a highly efficient model of particle immobilization, i.e., the feeding system of clams (as shown in Fig. 1A). In this system, water and food particles (e.g. the phytoplankton) are drawn in through the incurrent siphon, in which the hair-like cilia move the water to the gills. Then, food particles are caught in mucus (sticky fluid) produced by the gills [9]. In this process, mucus plays a very important role in the efficient food particle capture process (Fig. 1B). To simply mimic such an efficient model for nanoparticle immobilization, we propose to use a common adhesive tape as the microfluidic chip substrate, which has sticky polyacrylate surface with surface charges and radicals. Actually, the common adhesive tape has contributed to the Nobel-winning work on graphene [10], triboluminescence [11] and the reduction of metal salts to the corresponding metal nanoparticles [12]. Moreover, other different kinds of tapes, such as double sided adhesive tapes and adhesive transfer tapes have been used in microfluidics for various functions, such as rapid prototyping and PCR [13–15]. The acrylic pressure-sensitive adhesive is typically a “pure polymer” composition (e.g., polyacrylates, the average

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molecular weight from 100,000 to 1,000,000–5,000,000)[16], it can produce adhesion by itself. It is reasonable to expect the nanoparticles to be trapped by the adhesion force, or more specifically, van der Waal's force. This is the same mechanism as what the mucus of clams uses to capture the floating particles [17]. In contrast, the glass slide is traditionally a common substrate for particle immobilization, but often needs complicated surface modifications to anchor linker molecules containing organic groups like amine and thiol, especially when Au nanoparticles are to be trapped [18,19]. In this sense, the adhesive tape substrate is favorable as it avoids the need for surface modifications and expensive reagents.

This biomimetic device presents many advantages. First, the upper layer of microfluidic chip (usually made of polydimethylsiloxane, PDMS) can be sealed by simply pressing it against the adhesive tape, without the use of plasma bonding. The adhesive bond is temporary but strong enough to resist a high flow rate. Second, the adhesive tape can be easily stripped off from the PDMS structure for off-chip detection and can be replaced by a new adhesive tape to reuse the microfabricated PDMS layer, making the device portable, repeatable and inexpensive. Third, similar to the mucus-covered feeding structures, the adhesive tapes immobilize nanoparticles easily and efficiently. Fourth, the combination of PDMS and adhesive tape allows the microfluidic chip to be soft and flexible. In this work, we will experimentally study the immobilization properties of nanoparticles by the adhesive tape substrate and will further investigate the surface-enhanced Raman scattering (SERS) detection of rhodamine 6G (R6G) using the immobilized Au nanoparticles, with the aim to demonstrate the potential for biochemical sensing applications.

2. Materials and methods

The PDMS layer (width 1 mm and height 40 μm) with a simple pattern of serpentine microchannel was fabricated using standard soft lithography techniques (see Fig. 1C) [20]. Different from the normal plasma bonding process on a glass substrate, the PDMS

layer was sealed directly by pressing it onto the transparent adhesive tape (3M Company), without the use of plasma cleaners. As the tape has tacky substance on the surface, it can be adhered to another surface by applying a light pressure. The recommended bonding pressure is 14.5–29 psi (i.e., 100–200 kPa). Optical images show that the fabricated microfluidic chips are still transparent. Liquid with blue color is flowed along the microchannel smoothly and the flexibility of these chips is good (see Fig. 1D and the inset).

The bond strength between the PDMS and the adhesive tape substrate was characterized by two methods, *channel deformation test* and *direct interface bond strength test* using a digital force gauge (Aigu, Hong Kong). In the first test, the flow injected into the microchannel was slowly increased until a clear channel deformation was observed, and this flow rate was recorded as the critical flow rate. Here, we consider the channel deformation as an important factor but not the leakage since the stability of channel shape is important to many microfluidic applications, such as droplets, bubbles and laminar flow formation [21–25]. In the second test, a PDMS layer (5 mm \times 5 mm) adhered to a transparent adhesive tape was pulled by a digital force gauge. With the increase of the tension offered by the force gauge, the PDMS layer was separated from the adhesive tape. This force read from the gauge is the bond strength between the PDMS and the adhesive tape substrate. Each test was repeated for five times.

The immobilization stage was processed by the introduction of TiO_2 nanoparticle–water or TiO_2 nanoparticle–ethanol suspension into the microfluidic channels (50 mg/mL). The TiO_2 nanoparticles were P25 from Sigma–Aldrich, with the average size of 21 nm. During this immobilization process, the adhesive tapes trapped the TiO_2 nanoparticles tightly. To assess the immobilization effect, we compared the immobilization density on the adhesive tape with that on the glass slide using the same method. To investigate the adhesion strength of nanoparticles, we performed a shear stress test by flushing the microchannel at the critical pressure for ten minutes. During this test, we captured the optical images once every minute, followed by the calculation of TiO_2 nanoparticle

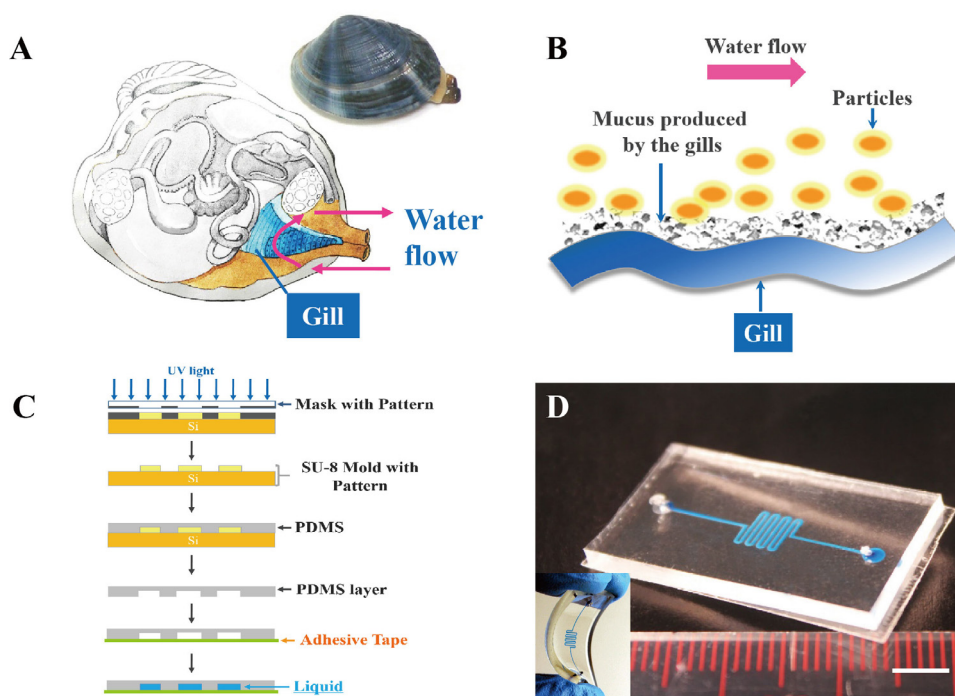


Fig. 1. Nanoparticle immobilization of the adhesive tape substrate by mimicking the clam. (A) Clam internal anatomic diagram. (B) Capture of floating particles by the gill secreted mucus. (C) Process flow of the PDMS microfluidic device using soft lithography. (D) Optical image of the fabricated microfluidic chips. PDMS layer was sealed simply pressing against the adhesive tape, without the need for the bonding process by plasma cleaners. The inset illustrates the flexibility of this chip. The scale bar is 5 mm.

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