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Charge-selective gate of arrayed MWCNTs for ultra high-efficient biomolecule enrichment by nano-electrostatic sieving (NES)

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ABSTRACT

We report a rapid and highly-efficient biomolecule preconcentrating device based on nano-electrostatic sieving (NES) mechanism that is facilitated by multi-nanofluidic channels operated in parallel. The opening of these nanochannels is regulated by tunable charges that are generated on arrayed multi-walled carbon nanotubes (MWCNTs) gate. The NES device is fabricated by standard photolithography and plasma-enhanced chemical vapor deposition (PECVD) techniques, followed by subsequent deposition of parylene (poly(p-xylylene))-C on vertically grown MWCNTs in order to obtain arrayed multi-nanochannels with mean pore sizes that are comparable to the thickness of an electrical double layer (EDL). The enrichment efficiency for charged analytes is dependent on electrostatic repulsion, which is regulated by the distribution of the local electric field on the MWCNTs gate. The NES device exhibits polarity selectivity on the analytes and performs efficient collection and separation of biomolecules by probing the surface charge density dependence on the applied gate field. A tunable gate of the parylene-MWCNT nanochannels was used as size sieving devices for nano-scale biomolecules. The experimental results for the collection of FITC-labeled bovine serum albumin (BSA, 0.033 nM) were as high as nearly 10^6 fold after only 45 min. These data are attributed to the in-parallel molecule sieving process as conducted by the many nanochannels formed among the MWCNTs. This device allows uncharged polar molecules, such as water, to rapidly pass through thus enable highly efficient bio-molecule concentration for the application to ultra-high sensitive biosensing.

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1. Introduction

Current efforts to establish microscale analytical systems are complemented by implementing nanometer-sized fluidic components (Kovarik and Jacobson, 2009; Perry and Kandlikar, 2006; Mijatovic et al., 2005) for providing a higher surface-to-volume ratio and confining geometry to reduce dispersion, which is beneficial for high-sensitivity analysis while maintaining a high-throughput operation (Han et al., 2008). As a result, considerations of surface charge density (Schoch and Renaud, 2005), nanopore size and shape (Abgrall and Nguyen, 2008), mobile phase ionic strength (Holtzel and Tallarek, 2007), and mass-to-charge ratio of ionic species (Garcia et al., 2005) for the nano-dimensional sample pre-treatments are of particular interest for improving bio-sensing performance. On the other hand, when encountering scarce sample amount within nanochannels, it is necessary to increase the sample concentration before analysis in order to enhance the signal-to-noise ratio and reliability for better detection (Napoli et al., 2010; Kim et al., 2010).

Therefore, several preconcentration schemes have been developed for capillary electrophoresis (CE) and microfluidic chips (Sueyoshi et al., 2008; Lin et al., 2010; Aranas et al., 2009). Among these preconcentration protocols, electrokinetic trapping is advantageous for molecule concentration due to its generality in charge polarity and molecule/particle sizes, and capability for effectively capturing and concentrating samples in diverse buffer conditions. In the past years, a number of electrokinetic trapping methods have been developed. For example, isoelectric focusing (Kim and Terabe, 2003) field amplified sample stacking (Sustarich et al., 2010), and isotachophoresis (Hirokawa et al., 2003) have been widely adopted to electrokinetically concentrate samples into a local equilibrium state. However, the aforementioned methods have suggested several mechanisms involving specific buffer systems or reagents for concentration, which may not be generally useful for fluidic systems that require different buffer conditions.

Recently, there has been increased interest in using nanochannels for the charge-selective transport of ionic species (Plecis

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et al., 2008; Karnik et al., 2007). Indeed, when the size of a microfluidic channel approaches the order of tens of nanometers, the surface charge induces an ion-exclusion effect (Bocquet and Charlaix, 2010; Jin et al., 2007), i.e., co-ions of the surface charges are excluded from the nanochannel to enrich counterions. So far, the best preconcentration rate has been achieved by Prof. Han's group (Wang et al., 2005), a million-fold preconcentration of GFP in 150 min using a micro/nanofluidic device. Kim et al. (2006) also demonstrated an impressive 10^5-fold preconcentration in 30 min using field-induced nanochannels. Other approaches relying on induced ion depletion (Karnik et al., 2005) and ion-permselectivity exclusion (Hlushkou et al., 2008) were reported to concentrate macromolecules by employing a porous membrane that excludes the species of interest from its membrane pores with electrokinetic transport (Foote et al., 2005; Hatch et al., 2006). However, most of these techniques are limited by Joule heating and bubble generation, which is induced by the highly concentrated electric field, as well as a long period of preconcentrating process limited by using only a single nano-passage.

In the present work, a charge-selective and preconcentrating device (Fig. 1(a)) is proposed to eliminate the requirement for special buffer conditions to obtain a high-performance molecule concentration by nano-electrostatic sieving (NES). The NES device consists of insulating layer coated MWCNTs array for not only the electrostatic gating of charged molecules but also the sieving of different size molecules. In the NES device, the electrical double layers (EDL) generated near the surfaces of the insulating layer coated MWCNTs array can overlap with one another to allow electrokinetic expulsion of a selected polarity of molecules. Due to the surface charge density can be further increased/manipulated by an external applied field on the CNTs gate electrode, the induced electrical double layer ($\lambda_{D\text{-}NES}$) with variable-length can be employed to control the nanochannel radius (Ro), i.e. the passages among MWCNTs, when λ_{EDL} is close to Ro. As a result, counter- and small ions have a higher transport rate than co- and large ions, as shown in Fig. 1(b), which would not be limited by the concentration of the bulk electrolyte ($\lambda_{D\text{-inherent}}$). When an electric fields for EOF is on, the EDL has the ability to fully overlap thus no co-ions can be leaked out to the other side of the nanochannel by exclusion-enrichment effect (EEE) while the counter-ions and water can be transported through the nano-channel rapidly by EOF. Like in a polarity-switching nanochannel (Nishizawa et al., 1995; Ku and Stroeve, 2004), the polarity of the MWCNTs gate can be switched in-situ to concentrate counter ions as shown in Fig. S1(a), opening up a new way for ion concentration which cannot be easily achieved by only counting on the inherent surface charges of the nanochannel. In contrast to the ion depletion effect only by applied bias on bulk ion concentration (Fig. S1(b)), the current CNTs gate allows more efficient control of the polarization and surface charge density on the gate surface (not in the bulk solution) than those depending only on inherent/fixed gate surface charge density.

Due to the use of a MWCNTs array, we accomplish this process at an electric field strength (75 V/cm) for operating an nanofluidic channel (Kim et al., 2006; Hlushkou et al., 2008; Hatch et al., 2006), therefore Joule heating and bubbling problems can be significantly reduced. Substantially un-insulated silicon microchip can normally only withstand a few hundred volts, before short circuiting (Harrison et al., 1993). In addition, the arrayed virtual nanochannels formed among the vertically aligned MWCNTs provide a much larger pathway (due to the large porosity ratio, empty area/occupied area in cross section, ~80% for MWCNT array (Wu et al., 2009; Majumder et al., 2010) the mobility of EOF can reach $9 \times 10^{-4}\,cm^2\,V^{-1}\,s^{-1}$ in our system) for water molecule passing through; thus, the MWCNTs-NES

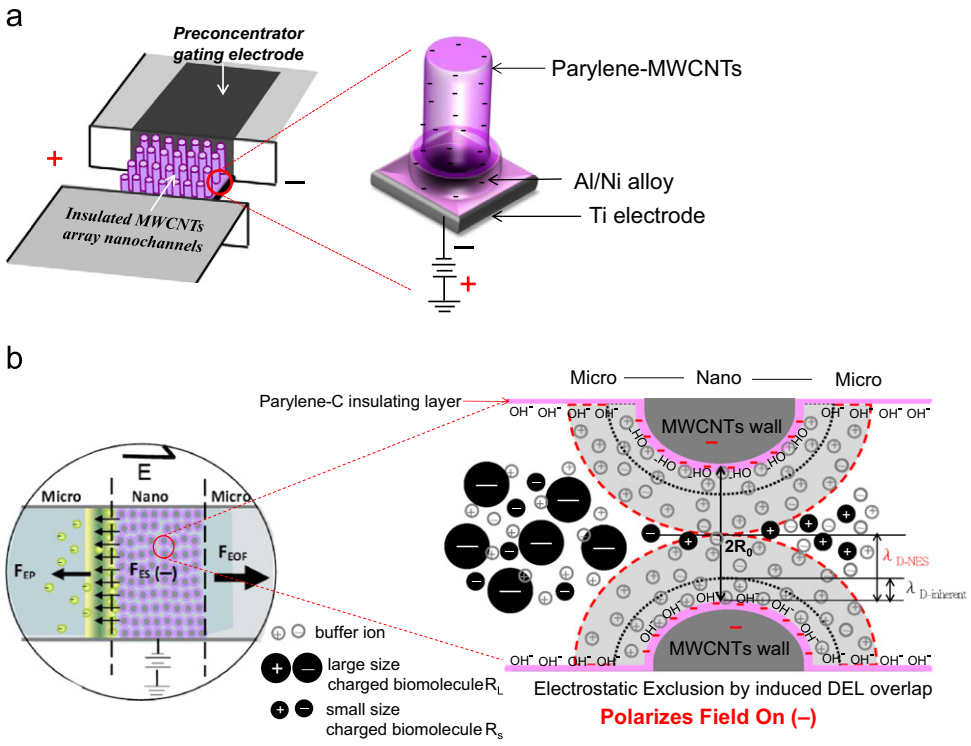

Fig. 1. Schematic illustration of (a) an electrokinetic concentrator realized by parylene-MWCNTs multi-nanochannel, (b) tunable EDL overlapped status before (black dashline, surface charge density of bulk solution) and after (red deshline, polarization charged density of NES) the application of a gating voltage on the arrayed MWCNTs gate electrode, and the mechanism of electrostatic exclusion for molecule concentration, (FEP: driving force by electrophoresis; FES: driving force by electrostatic repellent; FEOF: driving force by electroosmotic flow). (For interpretation of references to color in this figure legend, the reader is referred to the web version of this article.)

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