



Sensing Applications Using High-Throughput Heterogeneous Integration of Diverse Nanomaterials on a Single Chip

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Abstract

Nanomaterials come in a wide range, and each one has distinct electrical, optical, and sensing capabilities. However, there is currently no paradigm for the low-cost, high-throughput integration of several nanomaterials on a single chip. To produce a scalable array of individually addressable assemblies of graphene, carbon nanotubes, metal oxide nanowires, and conductive polymers on a single chip, we describe a high throughput integration technique based on successive spatially controlled dielectrophoresis [1]. This is the first time that a single chip has had such a variety of nanomaterials built on the same layer. The size and spacing of the underlying electrodes on the chip utilised for assembly are the only factors that restrict the resolution of assembly, which can range from mesoscale to microscale. The utility of such an array is illustrated with an example application of a chemical sensor array for detection of volatile organic molecules below parts-per-million sensitivity, despite the fact that many applications are feasible [2].

Keywords: Nanomaterials; gGapheme; Graphene oxide nanoplatelets; Polypyrrole

Introduction

Due to their potential to be used to create highly effective electronic and optical devices as well as ultrasensitive biological, chemical, and physical sensors, nanomaterials have received a lot of interest in recent years. Extreme dimensionality, or very high surface-to-volume ratios, is characteristics of nanomaterials. As a result, the material can be more sensitive than bulk materials because interactions at the material's surface, such as those with gas molecules, can change the material's overall properties [3]. Despite their sensitivity, it is challenging to arrange several arrays of variously manufactured nanomaterials on a single substrate due to the considerably varied synthesis processes of nanomaterials. The three most common categories of chemiresistive nanomaterials—organic and inorganic—are combined onto a single chip in this work. The nanomaterials used in this work have a variety of forms, including copper oxide nanowires, reduced graphene oxide nanoplatelets, carbon nanotubes, and nanostructured polypyrrole (PPy). It has never before been possible to integrate a heterogeneous mixture of such disparate nanomaterials onto a single chip-scale substrate [4]. While there are a variety of applications that could make use of this paradigm of high throughput assembly of several nanomaterials on a single chip, we show how useful it is for realizing a chemical sensor array. The food service industry, the medical field, environmental monitoring, and the military/security industries would all be very interested in such a sensor array chip [5].

In this section, we give a brief summary of the four distinct nanomaterials that were used to create the chemical sensor array in this study. Two of the four sensing nanomaterials used are carbon nanotubes and graphene nanoplatelets. With potential uses in energy storage, catalysis, electronic devices, and sensing, graphene is a promising two-dimensional allotrope of carbon in which the carbon atoms are organised in a hexagonal lattice. Reduced graphene oxide (rGO), which is produced chemically, has a surface made up of several oxygen atoms that are dangling and other functional sites like alcohol and carboxyl groups. The most common explanation for the sensing action is that the gas phase analyte acts as an electron donor or receiver at the impurity sites, modulating the carrier concentration and, consequently, the material's conductivity [6]. Carbon nanotubes (CNTs), a 1-dimensional analogue of graphene, have been long been

studied for the role of defects and electron exchange with and without functionalization, and reported sensitivities in the PPB range have been shown. The most prevalent type of chemiresistive sensor is made of metal oxides; tin oxide sensors, also known as Taguchi sensors, have been in use for a long time in a variety of applications. In this study, the chemiresistive characteristics of copper oxide nanowires at room temperature are investigated. There have been few research on the naturally p-type semiconductor copper oxide [7]. Long used as chemiresistors are conductive polymers, such as polypyrrole (PPy), and more recently, nanostructured conductive polymers have been found. Due to their inherent non-specificity and ability to react to a wide range of gases, conductive polymers are particularly common components in cross-reactive sensor arrays. This wide-ranging family of nanomaterials can be heterogeneously integrated in a single chip-scale platform to offer the diversity and selectivity required for cross-reactive chemical sensing of numerous target analytes. This was previously accomplished by functionalizing and/or altering components from the same family of chemiresistors (i.e. carbon, metal oxides, or conductive polymers). We recently displayed a sensing platform with various metal oxide nanowire kinds for gas sensing [8].

Materials and Methods

Carbon nanomaterial preparation

The CNCs were set up as previously mentioned. In a nutshell, a graphitic cathode and anode were placed within an arc chamber together with an inert gas (helium, argon, or nitrogen). The chamber was then given a current with a voltage (10–30 V) high enough to cause a carbon arc reaction. The chamber pressure was kept between 1 and

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2 atm, while the inert gas flow rate was kept between 60 and 90 cm³/min. The deposit on the cathode was collected and purified using a 0.22 m filter using a pulse current (50-70 Hz; 50-500 A). Approximately 70% of the deposits were CNCs before purification, while at least 95% were CNCs after purification. Similar MWCNTs were made utilising the arc-discharge technique in an argon environment, as previously mentioned. Deposits from the cathode were collected and purified using a direct current electric field. The deposits were about 50% pure when discovered, but following purification, they were more than 95% pure. C60 was bought from Sigma Aldrich, and SWCNTs from SES research (Texas, USA) (Missouri, USA). All carbon nanomaterials were dissolved in 5–10 mg/ml of 1 weight percent polyvinyl alcohol (PVA). The nanomaterial dispersions were sonicated for one hour right before injection (E60H, Elma Ultrasonics, Germany).

TEM analysis

For TEM examination with an H-7500 TEM, carbon nanoparticles were distributed in 1% PVA onto Formvar/carbon-coated 200 mesh copper grids from Ted Pella Inc. in California, USA (Hitachi, Japan). The materials were photographed by a skilled technician after being lyophilized for 24 hours.

Results

The original search as mentioned above yielded a total of 90 citations. The analysis of the bibliographies of the first 90 unique citations led to the discovery of an additional 40 citations. 38 of these 130 references were disregarded as duplicates. As a result, 92 references in all were evaluated before being included in the evaluation. Eight of them were disqualified based on the titles and abstracts. Each of these publications' specific exclusion from further evaluation is listed in the S1 Table. To determine eligibility, the remaining 84 citations were examined in full-text. 77 of them were disqualified. Consequently, a total of seven studies were included.

A total of 46 studies were eliminated because they failed to evaluate or describe a surveillance or screening programme in a population of workers who had been exposed to nanoparticles. 26 studies were also disregarded because they failed to provide defined parameters for biological or clinical monitoring. Four studies were disregarded because they either didn't provide enough details to identify the nanomaterials workers were exposed to (two), or they didn't provide details on the procedures followed to take regular measurements for surveillance reasons.

Discussion

All electrodes where assembly for all four types of nanomaterials was attempted produced resistive assemblies with a 100% yield. Due to process problems like lithography, packaging, or soldering faults, which may be easily fixed in the future, the yield of the functional microassemblies was lower at 65%. For instance, we observe a linked network of nanomaterials bridging the gap in some instances where the lengths of nanomaterials were lower than the electrode gap. The smaller CuO nanowires that are constructed on only one side of the electrode pair in the case of CuO nanowires (see figure) do not contribute to chemiresistive responses because they do not bridge the electrodes [9]. Lack of wire bond integrity, lithographic mistakes, and electrode corrosion during assembly are some causes of faulty sensors. In the case of rGO, the DEP assemblies were poisoned and became less sensitive when submerged in the PPy dispersions for subsequent assembly. One of the main advantages of the suggested approach is the absence of any evidence of cross-contamination or spurious assembly

of undesirable nanomaterials at nonspecific electrodes. Impedance measurements between all of the electrode pairs in between the successive processes of assembling various nanomaterials were used to determine the cross-contamination measurement [10]. The impedance of the samples not chosen for DEP did not change (measured within the instrument's error of 102 ohms). This includes measuring electrodes that have had previous assemblies attached to them. This property can be explained by two factors: first, the electric field is only applied locally to the electrodes where assembly is required, and second, the dielectrophoretic force only affects the dispersed nanoparticles, not the nanomaterials that have already been assembled. Furthermore, it was not seen that preassembled nanomaterials would breakdown during future assembly because Van der Waals forces strongly hold them to their electrodes. It should be noted that one cannot legitimately assert that there is absolutely no cross-contamination because no pictures or material characterization were performed at each electrode site in between assembly [11]. However, the impedance measurement showed no influence of any cross-contamination from the standpoint of a sensing application.

The electrode spacing determines the assembly resolution, which can be increased to nanoscale levels. The goal of future work might be a precise description of the lowest resolution that is currently attainable [12].

Our prior research using scanning probe microscopy to in situ monitor DEP constructed rGO has demonstrated that the main mode of sensing is due to redox reactions of the gas species modifying the charge carriers in the semiconductor assembly [40]. The same research demonstrates that the contact resistance does not significantly affect the sensor's overall resistance [13]. Although it is hypothesised that the same mechanism is in charge of the sensing activity in the other assemblies, more research is required and is the subject of ongoing investigations.

Conclusion

In order to realise a scalable array of individually addressable assemblies of graphene, carbon nanotubes, metal oxide nanowires, and conductive polymers on a single chip, this work illustrates a throughput integration approach based on spatially controlled dielectrophoresis executed sequentially for each nanomaterial type. The constructed array was used to detect a variety of gas species at PPB concentrations, demonstrating the usefulness and potential of such a platform. This integration platform can also be used with nanomaterials that are optically, physiologically, or mechanically sensitive to create image sensors, bioassays, or tactile sensors, respectively. The platform's future directions include enlarging the array dimensions to accommodate additional elements, including a wider range of sensor components, and adding a pattern recognition engine to enable the identification and measurement of gases. For additional applications, the technological challenge of contact formation with other nanomaterials will also need to be solved. In addition, the technology will make it possible to integrate hybrid CMOS-nano devices, which is presently not conceivable with other methods like microcontact or nanocontact printing or nanoimprint lithography. This includes top-down produced CMOS-dies. The foundation for all future effort will be this.

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