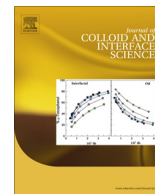




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Gold nanoparticles deposited on linker-free silicon substrate and embedded in aluminum Schottky contact

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ABSTRACT

Given the enormous importance of Au nanoparticles (NPs) deposition on Si substrates as the precursor for various applications, we present an alternative approach to deposit Au NPs on linker-free n- and p-type Si substrates. It is demonstrated that, all conditions being similar, there is a significant difference between densities of the deposited NPs on both substrates. The Zeta-potential and polarity of charges surrounding the hydroxylamine reduced seeded growth Au NPs, are determined by a Zetasizer. To investigate the surface properties of Si substrates, contact angle measurement is performed. Field-emission scanning electron microscope is then utilized to distinguish the NPs density on the substrates. Finally, Al/Si Schottky barrier diodes with embedded Au NPs are fabricated, and their structural and electrical characteristics are further evaluated using an energy-filtered transmission electron microscope and current–voltage measurements, respectively. The results reveal that the density of NPs is significantly higher on n-type Si substrate and consequently has more pronounced effects on the electrical characteristics of the diode. It is concluded that protonation of Si–OH group on Si surface in low pH is responsible for the immobilization of Au NPs, which eventually contributes to the lowering of barrier height and enhances the electrical characteristics.

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1. Introduction

Since the advent and evolution of nanoscience and nanotechnology, metal nanoparticles (NPs) and in particular, gold (Au) NPs have served as one of the most essential building blocks for the fabrication of nanostructure devices [1,2]. Their magnificent optical, electronic, and catalytic properties as well as their controllable size and easily functionalized surface have made the Au NPs the subject of extensive research [1]. These properties render Au NPs applicable in a variety of applications ranging from bioengineered structures to electronic and photonic devices [1–3].

One of the applications of Au NPs in electronic devices is as embedded NPs in metal contacts with different work function to control and tune Schottky barrier height (SBH) in Schottky barrier diodes (SBDs) [4] or to reduce contact resistivity of Ohmic contacts [5]. The work function difference between two metals (Au NPs and metal contact) will setup an electric field, which in turn leads to the facilitation of current conduction through the interface of metal and semiconductor substrate [4,5]. The Au NPs have been previously embedded in metal contacts such as nickel (Ni) [4,6,7], titanium (Ti) [8], palladium (Pd) [9], and tungsten

(W) [5]. Utilizing this method, a two-step process is needed to form the structure on a semiconductor substrate typically on silicon (Si).

The first step to embed Au NPs in metal contact is to deposit the NPs on a substrate followed by the deposition of the metal contact. Au NPs are either being synthesized and deposited directly onto the substrate during the fabrication procedures such as aerosol deposition [6], anodic porous alumina mask [9], ion-implantation [10], and direct deposition self-assembly [5]; or being synthesized separately through reduction of metal ions by reducing agents and building up nano-sized Au metals into the NPs colloidal suspensions. The separately synthesized colloidal Au NPs are then deposited onto the substrates using less expensive methods such as spin coating of the NPs solution onto the substrate or conventionally dipping the substrate into the NPs solution [11–14].

In the case of colloidal nanoparticles, to prevent aggregation and finally coalescence of Au NPs as a result of long-range attractive Van der Waals forces between the NPs, they need to be stabilized by covering with capping agents or stabilizers acting as repulsive agents as opposed to the attractive Van der Waals forces. Electrostatic and steric stabilization or a combination of both, are the most well-known stabilizing mechanisms [15]. Electrostatic stabilization is determined by the interaction of the attractive Van der Waals and repulsive electrical double layer forces that exists between NPs as they approach each other, following the

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Derjaguin–Landau–Verwey–Overbeek (DLVO) theory [16]. Ionic capping agents such as citrate or dodecyl sulfate ions [17] are being introduced during the synthesis process of NPs and having the role of both reducing agent and capping agent, at the same time. Au NPs can then be grown to a desired size by controlling the parameters of synthesis or by seeded growth methods using different reducing agents such as hydroxylamine [18,19].

A number of studies have been carried out on the development of Au NPs deposition on specifically Si substrates [3,20,21]. These methods can be categorized into two general approaches which are summarized in Fig. 1a and b. First one includes the functionalization of the Si substrate with linker molecules followed by the deposition of Au NPs (Fig. 1a). In the second approach, the deposition of NPs takes place on linker-free Si surfaces. The latter can be achieved by simply drying of the NPs on Si surface [22] or acidifying of the NPs solution on H-terminated Si substrates (Fig. 1b) [3,23]. In acidifying process by altering pH of the NPs solution by HF or HCl addition, the citrate ions stabilizing the Au NPs are neutralized and hence, Au NPs are destabilized on H-terminated Si surface [3,23]. Nevertheless, the application of linker molecules to ensure the NPs adhesion to the Si substrates is the most commonly used approach [24–28]. Functionalization of Si substrates with linker molecules are typically carried out by deposition of a self-assembled monolayer (SAM) of polymeric linker molecules on a clean and modified thin silicon dioxide surface with high density of Si–OH (silanol) groups [24]. Chemisorption or physisorption mechanisms between NPs and Si surface have been known to be responsible for the immobilization of NPs on the Si substrate [27–29]. In the chemisorption process the functional groups on NPs and substrate are covalently bonded while in the physisorption process the NPs and functionalized groups on the surface are bonded through Coulombic attraction of negatively charged NPs

to the positively charged functionalized Si surface [26,28,30], or vice versa. There are some drawbacks to both methods when applied for the fabrication of electronic devices such as Schottky barrier diodes making the NPs deposition on the substrate a costly and time-consuming step. The functionalization of the substrate is normally time consuming and will end up with polymeric linker molecules attached to the Si substrate, which cannot be removed from the surface later, and will eventually affect the current conduction mechanism of the diode. Moreover, a relatively high volume amount of NPs solution is required for the immobilization of the NPs on Si surface if the substrates are immersed into the NPs solution. Similarly in acidifying process, due to the hydrophobic properties of H-terminated Si surface, it may require a relatively high volume of NPs solution in order to cover the surface of the substrate uniformly. On the other hand, different types (n- and p-type) of Si substrate behave substantially different due to their specific surface properties (especially substrates with high doping concentration) [31]. In spite of this fact, the effect of this difference to the final density of deposited NPs on n- and p-type Si substrates have not been addressed in literature.

In this study, an alternative approach to deposit Au NPs with a relatively low amount of colloid (5 μ l) on hydrophilic linker-free Si substrates is presented (Fig. 1c). This is carried out without even acidification and destabilization of the NPs during the deposition process. Moreover, we demonstrate the effect of the difference between n- and p-type Si surface properties on final density of the deposited Au NPs on the substrates, which is not previously reported. Our approach is based on the formation of a pure native oxide at the final step of RCA (Radio Corporation of America) cleaning process, which is rich in Si–OH groups and then deposition of more than one monolayer of seeded growth Au NPs [32] by spin-coating technique. The immobilization of Au NPs on Si substrate is due to

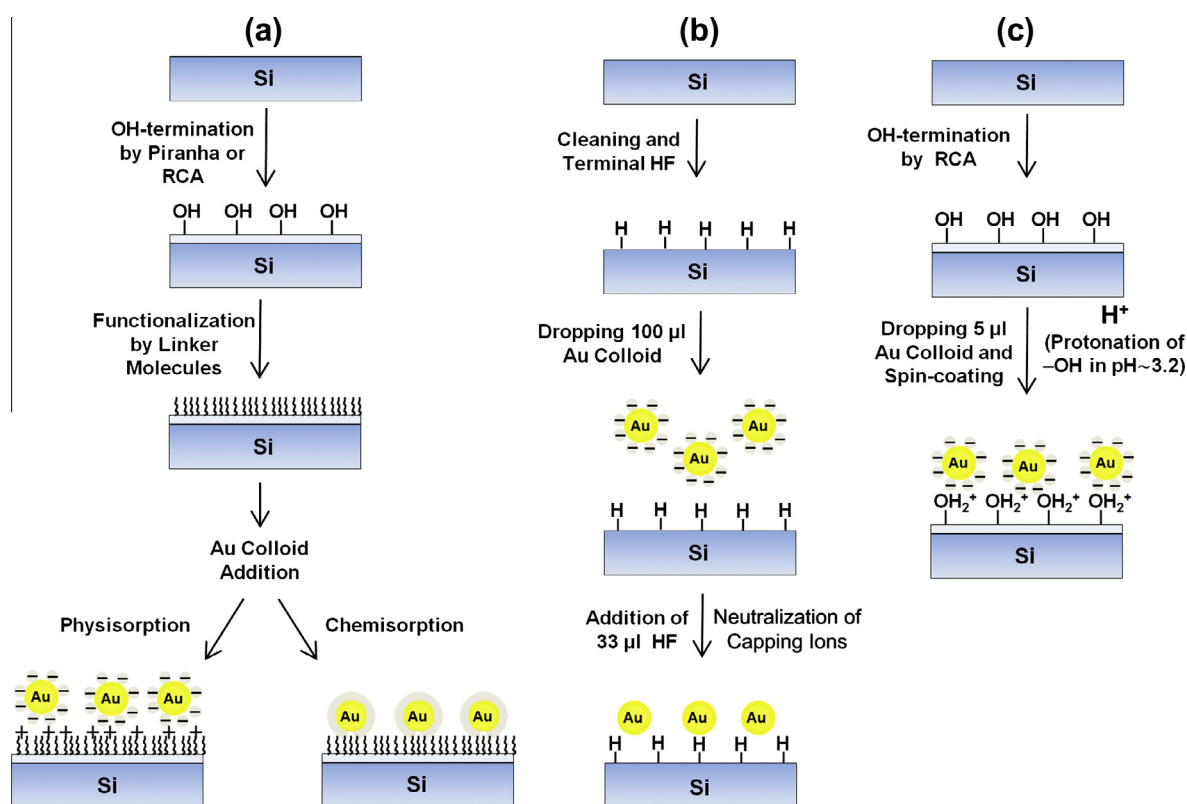


Fig. 1. Schematics of the colloidal Au NPs deposition routes on Si substrate using (a) functionalization process, (b) lowering the pH of colloidal Au NPs, and (c) which was introduced in this study, is the protonation mechanism of Si–OH groups on Si surface after dropping seeded growth Au NPs solution with pH~3.2, and subsequent spin-coating.

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