



The electrochemical behavior of tellurium on stainless steel substrate in alkaline solution and the illumination effects



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ABSTRACT

The electrochemical behavior of tellurium on stainless steel plate substrate in alkaline solution and the illumination effects are investigated by utilizing cyclic voltammetry, chronoamperometry and electrochemical impedance spectroscopy. Cyclic voltammetry results indicate that Na_2TeO_3 is firstly reduced to Te and Te is further reduced to Na_2Te . These reduction processes are all controlled by mass transfer. The reductions for tellurium electrodeposition under illumination can be promoted affirming by a more positive reduction potential and a higher reduction current. Photocurrent transient demonstrates that the influence of light on accelerating the deposition rate is owing to electrons excited from the valence band to the conduction band of tellurium. Chronoamperometry measurements indicate that the electrocrystallization of tellurium follows a 3D progressive nucleation and diffusion-limited growth mechanism. Impedance spectroscopy analysis show that the illumination can reduce the Faraday resistance and promote the rate of cathodic reduction owing to the photoconductive effect and the photogenerated electrons injected to the electrolyte. Electrodeposition under illumination can be a good method to produce tellurium in aqueous alkaline solution.

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1. Introduction

Recently, tellurium-based compound semiconductor thin films, such as CdTe and Bi_2Te_3 , have been applied in many fields due to their excellent light sensitivity and thermoelectric property [1–3]. As one of the basic element, tellurium, has also been widely used in metallurgical industry, for example doping in copper to enhance its mechanical property [4]. With the worldwide growing consumption of tellurium, it is necessary to develop a high yield and low energy method to produce tellurium. Electrodeposition is one of the best methods to deposit semiconductor materials because of its economic benefit, easy control of technology parameters and large-scale production [5–8]. However, there are many drawbacks for tellurium electrodeposition as a consequence of its semiconductor characteristics. The increasing resistance and slow deposition rate will lead to the heavy energy-consumption because of the poor electrical conductivity of tellurium. Many semiconductor compounds have been electrodeposited with photo-assistance to accelerate the deposition rate and obtain a better quality of thin films [9–11]. Based on this, we have proposed a novel photoelectrodeposition way to extract the semiconductor elemental tellurium. Firstly the electrochemical behavior of tellurium and the effects of illumination on the electrodeposition and electrocrystallization of tellurium have to be studied in microcosm. Alkaline solution and the stainless steel substrate have been mainly used to electroplate

tellurium in metallurgy industry [12]. There has been plenty of research on the electrochemical behavior of tellurium on different electrodes (such as glassy carbon, Au substrate and p-Si(1 0 0) wafer) in varies of acid aqueous solutions [13–16]. All the studies prove consistently that Te(IV) is firstly reduced to Te(0) by a 4 electron reduction process, and then at more negative potential, Te(0) can be further reduced to Te(–II). However, the investigation on the electrochemical behavior of tellurium in alkaline solution has been little studied. Most research in alkaline systems is focused on the electrodeposition and electrochemical behavior of tellurium-based compound semiconductor instead of the elemental tellurium [17,18]. For instance, Heini Saloniemi et al. [17] have studied the electrodeposition lead telluride thin films on SnO_2 and Cu substrates in aqueous alkaline solution. Kuniaki Murase et al. [18] have reported photoassisted electrodeposition of CdTe layer from ammoniacal basic aqueous solution. Consequently, it is of great significance to fully understand the electrodeposition and electrocrystallization of tellurium on stainless steel plate substrates in alkaline solution.

Tellurium is a p-type semiconductor with a narrow bandgap [19]. Once the tellurium thin film deposited on the substrate, the electrochemical reaction interface would change from conductor/electrolyte to p-type semiconductor/electrolyte. In principle, the Fermi energy of solution is higher than that of p-type semiconductor, and electrons will flow from solution to the semiconductor, causing a negative charge of the surface of semiconductor. When the surface of p-type semiconductor is illuminated by the light, the photons with higher energy than the bandgap of the semiconductor will be absorbed by the

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semiconductor and then generate electron-hole pairs. The photo-generated electrons are injected from semiconductor into solution by the field of space charge region (i.e. depletion layer) to promote the reduction reaction [20,21]. The influence of light on the electrochemical behavior of tellurium is worth investigating in detail.

In this article, for the first time we investigate the nucleation and growth mechanism of tellurium electrodeposition on the stainless steel plate substrate in alkaline solution and the effects of light on the electrochemical behavior of tellurium by employing cyclic voltammetric, chronoamperometric and electrochemical impedance spectroscopy techniques.

2. Experimental

All electrochemical experiments were carried out in a conventional three-electrode cell with a stainless steel plate measuring 1 cm × 1 cm as working electrode, a saturated calomel electrode (SCE) as reference electrode and a purity graphite plate as auxiliary electrode. A Newport 300 W xenon lamp with the light intensity kept at 100 mW/cm² was used as light source to irradiate the surface of stainless steel plate substrates to investigate the influence of light on the electrochemical behavior of tellurium. All potentials in this work were reported versus SCE. Stainless steel plate substrates were cleaned ultrasonically with alcohol and deionized water for 15 min before every electrochemical experiment. The electrolyte solution for study of electrochemical behavior of tellurium consists of 10 mM TeO₂ (99.99%) and 2.5 M NaOH. All experiments were implemented in a quiet bath at room temperature. Princeton Applied Research PARSTAT 4000 Potentiostat was applied for all above experiments. Cyclic voltammetry experiments were performed at a sweep speed of 10 mV s⁻¹ or 5 mV s⁻¹ and scanned firstly in the negative direction. Chronoamperometry measures were carried out from the first peak potential (about -1.1 V) to the second peak reduction potential (about -1.46 V) to avoid the further reduction of tellurium. The initial frequency (I.F.) in electrochemical impedance spectroscopy experiments was 100 kHz, and final frequency (F.F.) was 1 Hz with AC sine wave amplitude of 5 mV. If F.F. was too low, the surface of stainless steel substrates would change strongly as a result of long-time electrodeposition. Therefore a final frequency of 1 Hz was chosen.

3. Results and discussion

3.1. Cyclic voltammetry

Cyclic voltammetry measurements were carried out in the potential range of 0.2 V to -1.5 V to confirm the existence of the

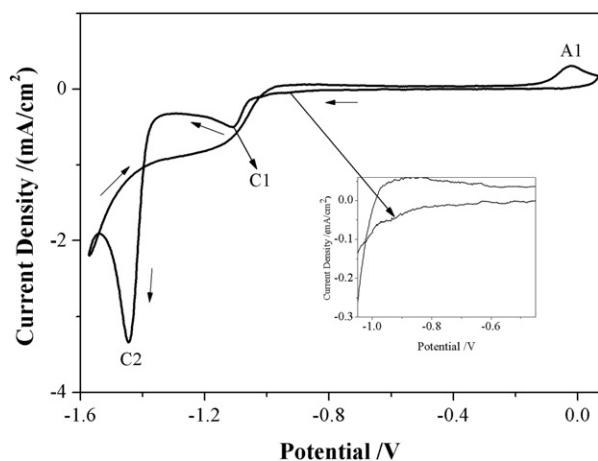
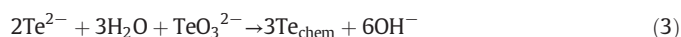
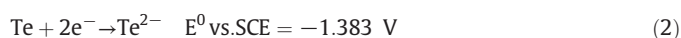


Fig. 1. Cyclic voltammogram for tellurium electrodeposition on stainless steel electrode in 10 mM TeO₂ + 2.5 M NaOH solution. Sweep rate is 10 mV s⁻¹.

electrodeposition of tellurium thin films. Fig. 1 shows cycle voltammogram of the stainless steel plate electrode in 10 mM TeO₂ + 2.5 M NaOH solution with a sweep speed of 10 mV s⁻¹. The curve exhibits two obvious cathodic peaks (marked peak C1 and C2) and one anodic peak (marked peak A1). It is proved that C1 at about -1.1 V is assigned to the four-electron reduction process of Te(IV) to Te(0) according to Eq. (1). Peak C2 at about -1.46 V is owing to Te(0)/Te(-II) reduction couple corresponding to Eq. (2). It is worth mentioning that once the Te(-II) is formed, it will react with Te(IV) resulting in the chemical formation of Te(0) through a comproportionation reaction in solution (Eq. (3)). From the inset we can see clearly that there is an initial reductive characteristic in the potential range of -0.8 V to -1.0 V affirmed by the very low reductive current. This indicates the formation of the first monolayer tellurium thin film before the bulk deposition. Anodic peak A1 can be assigned to the oxidation of the bulk tellurium deposits formed in the course of cathodic reduction process. There is an apparent crossover between the cathodic scan and the reverse anodic scan, which is called the “nucleation loop” [22], and this indicates typical tellurium nuclei formation and growth on the stainless steel plate substrate.



A series of linear sweep voltammograms at different sweep rate are carried out to figure out the controlling procedure during tellurium electrodeposition and shown in Fig. 2. With the increase of sweep rate, electrodeposition of tellurium commences at more negative potential and the cathodic peak current increases sharply. The relationship between the peak current density of C1 and C2 and the square root of scan rate (V^{1/2}) are presented in Fig. 3. The plots exhibited a very good linear relationship, showing that the reduction process for C1 and C2 is partially controlled by mass transfer [23]. Actually, peak C2 should be assigned to the following reduction: TeO₃²⁻ + 3H₂O + 6e⁻ → Te²⁻ + 6OH⁻, since only when the tellurium is formed can the reaction (2) be going on. So it is the above-mentioned reduction of peak C2 that is controlled by mass transfer.

Fig. 4a shows cyclic voltammograms in the dark and under illumination respectively, obtained at the scan rate of 5 mV s⁻¹. It is observed that the potential of the cathodic peak C1 and C2 shifts to more positive values and the peak current increases when the surface of working electrode is illuminated by the lamp. Fig. 4b shows the cathodic photocurrent transient under chopped illumination at the potential of -1.0 V. It displays a sharp increase in the absolute value of the current density

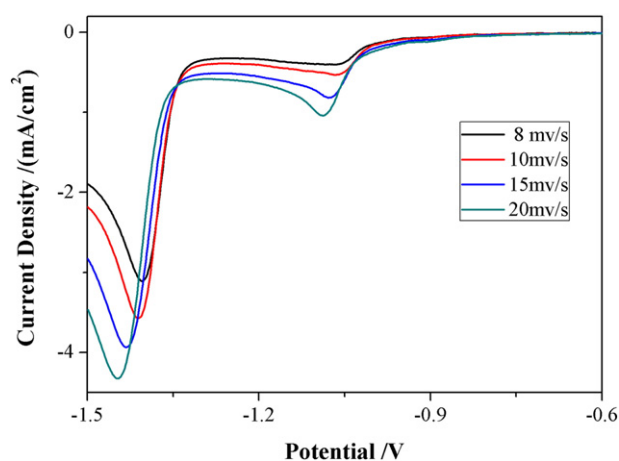


Fig. 2. Linear sweep voltammograms for tellurium electrodeposition on stainless steel electrode in 10 mM TeO₂ + 2.5 M NaOH solution at various scan rates.

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