



Solution-processed fabrication of perfectly (2 0 0)-oriented lead selenide thin films

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ABSTRACT

To the best of our knowledge, the solution-processed fabrication of perfectly (2 0 0)-oriented lead selenide (PbSe) films on various substrates, including glass, transparent conducting glass, and polyethylene terephthalate (PET) substrates, has been achieved for the first time. Herein, the effects of orientation parameters, including the acetate anion effect, crystallization temperature, and chemical composition, on solution-processed PbSe thin films were examined. Particularly, the presence of acetate anions in the precursor solution strongly affected the degree of orientation. X-ray diffraction analysis was employed, and X-ray diffraction pole-figure polar distance was scanned by rotating the sample about the incident beam direction. The experimental results confirmed the formation of perfectly (2 0 0)-oriented PbSe films.

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1. Introduction

Currently, the development of methods for the fabrication of metal chalcogenide thin films is attracting considerable attention because of their broad applications in large-area photodiode arrays [1], solar cells [2], photoconductors [3], X-ray detectors [4], and photosensors [5]. Among the various chalcogenide semiconductors, lead selenide (PbSe) has demonstrated immense potential because of a narrow band gap of 0.26 eV at room temperature and superior electrical conductivity, which are suitable for the operation of optoelectronic devices in the infrared region [6].

PbSe thin films have been fabricated via several techniques, including vacuum deposition [7], electrodeposition [8], chemical bath deposition [9], and successive ionic layer adsorption [10]. The structural, optical, morphological, and electrical characteristics of the fabricated PbSe thin films depend on the preparation methods and the corresponding experimental parameters [11].

In particular, the optical and electrical properties of PbSe are considerably dependent on the crystal orientation and orientation distribution [12]. Therefore, securing technologies for controlling the orientation distribution of PbSe thin films is a crucial challenge for optimizing the electrical and optical properties of thin-film-based functional devices. However, most of the abovementioned techniques have limitations in terms of producing orientation controlled PbSe thin films.

To date, to the best of our knowledge, few studies have reported oriented PbSe thin films obtained using specific vacuum techniques, with no successful report on any axis-oriented PbSe thin film [13]. Furthermore, the vacuum techniques reported so far require the use of expensive procedure, special setup, and/or sophisticated instruments. Solution processing is a promising alternative that can be used for fabricating thin films because of its cost effectiveness, high yield, and potential for large-scale thin-film production [14]. Despite these merits, solution processing has never been proven successful from the perspective of orientation control.

Herein, perfectly (2 0 0)-oriented PbSe thin films were easily prepared by spin coating a PbSe precursor solution that comprises lead acetate and selenourea, and by crystallization at varying temperatures greater than 100 °C. This study provides a widespread method for the formation of perfectly (2 0 0)-oriented PbSe thin films and investigates its effect on the device performance of various PbSe thin films.

2. Experimental section

2.1. Preparation of a PbSe precursor solution

A typical procedure for preparing a PbSe precursor solution is as follows. Lead ion source solution was prepared by introducing Pb(CH₃CHO)₂·3H₂O (0.452 g; 1.2 mmol) into a 3-mL aliquot of 2-methoxyethanol. To separately prepare a selenium source solution, selenourea (0.123 g; 1.5 mmol) was completely dissolved in

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2-methoxyethanol (3 mL) via continuous vigorous stirring and was maintained in an ice bath at $-5\text{ }^{\circ}\text{C}$. Next, the lead source solution was added to the selenium source solution while stirring in the $-5\text{ }^{\circ}\text{C}$ ice bath. The mixture color rapidly changed to transparent yellow. Finally, the mixture was stirred for another 5 min and maintained at $-5\text{ }^{\circ}\text{C}$.

2.2. Fabrication of PbSe thin films

First, a glass substrate was sonicated using absolute ethanol and cleaned using subsequent oxygen plasma treatment for 5 min. The prepared PbSe precursor solution was then spin coated on clean substrates at a speed of 2000 rpm for 120 s. Subsequently, the spin-coated films were placed in a quartz tube under the flow of high-purity argon (99.999%) for 30 min. This tube was then transferred to a preheated tubular furnace at desired temperatures of $100\text{ }^{\circ}\text{C}$, $200\text{ }^{\circ}\text{C}$, $300\text{ }^{\circ}\text{C}$, and $400\text{ }^{\circ}\text{C}$ for 30 min.

2.3. Characterization

X-ray diffraction (XRD) patterns of PbSe thin films were recorded on an X-ray diffractometer (PHILIPS/X'Pert-MPD system) using Ni-filtered monochromatic Cu $K\alpha$ radiation. XRD pole-figure analysis was conducted with high-resolution X-ray diffractometer (PHILIPS/X'pert PRO-MRD). Film-surface morphologies were

observed via atomic force microscopy (AFM; BRUKER/Icon-PT-PLUS). Conductivity measurements were performed using a four-probe system equipped with a Keithley 2182A nanovoltammeter and a 6221 current source.

3. Results and discussion

PbSe thin films were fabricated by solution processing via the spin coating of a precursor solution on various substrates, including glass, conducting indium tin oxide-coated glass, and PET substrates. First, the PbSe precursor solution was easily prepared by mixing with solutions in which the lead acetate and selenourea were dissolved in 2-methoxyethanol (Fig. 1a). During thermal treatment under argon flow for crystallizing the precursor spin-coated film, the coated thin film rapidly changed color from colorless to dark brown (Fig. 1b and c).

The UV-vis-NIR absorption spectra and the Tauc plot of $(\alpha h\nu)^2$ versus $h\nu$ of PbSe thin film were shown in Fig. 1d. The linearity of the plots of $(\alpha h\nu)^2$ versus $h\nu$ indicated the direct transition band gap, and the direct transition band gap value of resultant PbSe thin film was estimated to be 1.0 eV by extra polluting, which is higher than the reported bulk PbSe value of 0.27 eV [1,2]. The increase band gap is attributed to quantum size effect.

AFM analysis was performed for observing the surface topography and grain sizes. Fig. 1e shows the AFM images of the

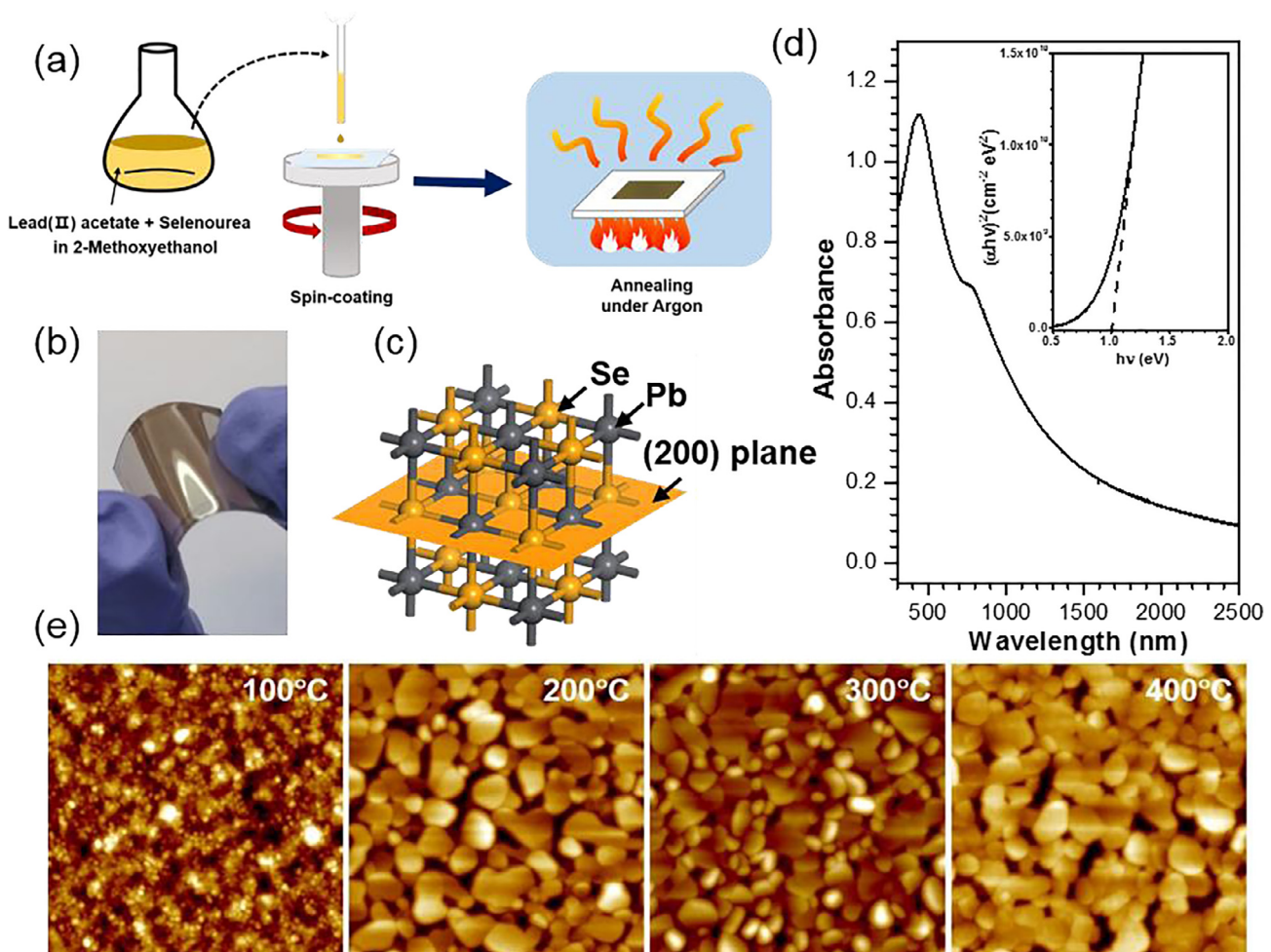


Fig. 1. (a) Schematic illustration of fabrication procedure, (b) photograph image of PbSe thin film fabricated on the PET substrate (c) crystal structure of PbSe with (2 0 0) plane, (d) absorption spectra and the corresponding Tauc plot for band gap determination of PbSe thin film fabricated on the glass substrate and (e) AFM images of PbSe thin film fabricated on the glass substrate with different crystallization temperature as indicated.

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