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Influence of Substitution Group of the Side Chain of Polypeptide on the Morphology and Its Hysteresis Property as a Ferroelectric Memory Device

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1. Introduction

Organic materials have been attracting significant interest for their potential use as inks in low-cost printable electronics device such as flexible displays [1-3], electronic papers [4-7], RFID tags [8], and sensors [9]. Printable nonvolatile memories have high applicability in electric devices, but promising techniques for the fabrication of these memories have not been developed thus far. Since an organic thin-film transistor [OTFT] fabricated using ferroelectric materials as a gate dielectric shows memory property, the development of a new ferroelectric material has great significance from the viewpoint of all printing device fabrication. Polyvinylidene fluoride (PVdF) and its derivatives are typical ferroelectric materials used as dielectric layers in OTFTs memory because of the high ferroelectricity. However, in order to generate ferroelectric behavior using PVdF, it is necessary to subject the thin film to special treatments such as stretching, annealing, and poling [10].

We have reported the fabrication of a printable OTFT memory using α-helical poly (γ-methyl-L-glutamate) [PMLG] as a ferroelectric layer [11]. The use of a PMLG film is advantageous because it exhibits ferroelectric behavior without requiring any special treatment. Further, we have shown that the memory device shows good performance when the molecular axis of PMLG is arranged parallel to the film surface. PMLG has a rigid rod-like, i.e., α-helix, structure, and it behaves as a mesogen. Therefore, the molecular axis can be arranged easily by carrying out only the printing process.

However, the alignment of PMLG was strongly depended on the preparation condition. For example, when a PMLG of large molecular weight was used as an ink of OTFT memory, the PMLG was easily crystallized in fibrous because of the strong molecular interaction between PMLG molecules. In such case, since the alignment of PMLG molecules on the substrate was broken, the ferroelectricity of the device was not observed. Therefore, it is important to control the alignment of molecules. In this paper, we investigate the influence of side chain of a polypeptide on morphology of the film and its hysteresis property in the memory device. In a previous study, a ferroelectric memory device was fabricated with a PMLG which have methyl group as a end group of the side chain of poly (L-glutamate). In this study, poly (γ-benzyl-L-glutamate) [PBLG], which have benzyl group as an end group, was used in order to investigate the influence of substituent group.

2. Experimental

Preparation

A PMLG (Kyowa Hakko Co., Ltd., degree of polymerization: 185) film and PBLG film (Sigma Aldrich Co., degree of polymerization: ca. 180) was prepared by dipping method or spin-coating method with a 1,2-dichloroethane (Kanto Kagaku Co.) solution containing PMLG and PBLG. In the experiment for the effect of polymer blend, poly (methyl methacrylate) [PMMA] was used as reserved (Nakalai Tesque, Inc.).

Device fabrication and measurements

During OTFT fabrication, the abovementioned solution was spread on an indium thin oxide (ITO) glass substrate (sheet resistance: 10Ω/square) by dipping method and then dried in solvent atmosphere. A pentacene (Naad Co., 98% purity) layer (thickness: 50 nm) to be used as the semiconductor layer and Au layer to be used as the source/drain electrode (W/L = 5 mm/20 μm) were prepared by vacuum evaporation. The structure of the OTFT is shown in Fig.1. In all dielectric films, OTFT was fabricated by the abovementioned method. All electrical measurements were carried out using a Keithley 2400 and 6430 SourceMeter in a vacuum.

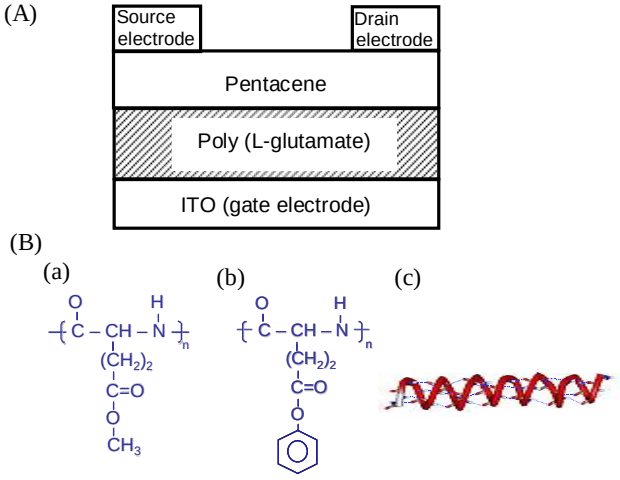

Fig.1 Schematic representation of cross-sectional view of device structure of OTFT memory (A) and molecular structure (B) of PMLG (a), PBLG (b) and these secondary structure (c), i.e., α-helix structure .

3. Results and Discussion

The memory behavior can be confirmed by measuring the hysteresis property in the transfer characteristic of OTFT device. If the OTFT device shows memory property, large hysteresis, which attributed to ferroelectricity of the gate dielectric layer, is observed. Fig.2 shows the transfer characteristic of OTFT fabricated using PMLG films (a) and PBLG film (b) as a gate dielectric. The larger and smaller hysteresis curves were observed in cases (a) and (b), respectively. In the case of PMLG, the AFM image shows nano-scale domain due to ferroelectric phase [Fig.2 (a)]. On the other hand, the AFM image of the surface of PBLG film shows fibrous crystalline structure. It indicates that the crystallinity of poly (glutamate) was increased by having benzyl group instead of methyl group as an end group of side chain. It can be considered that the interaction between main chains of poly (glutamate) was increased by replacing methyl group to benzyl group. Probably, the benzyl group accelerated the packing of side chains in the neighborhood of poly (glutamate). It leads to strong interaction between main chains, i.e., high crystallinity. If the crystallization can be prevented, large hysteresis in the transfer characteristic may be observed.

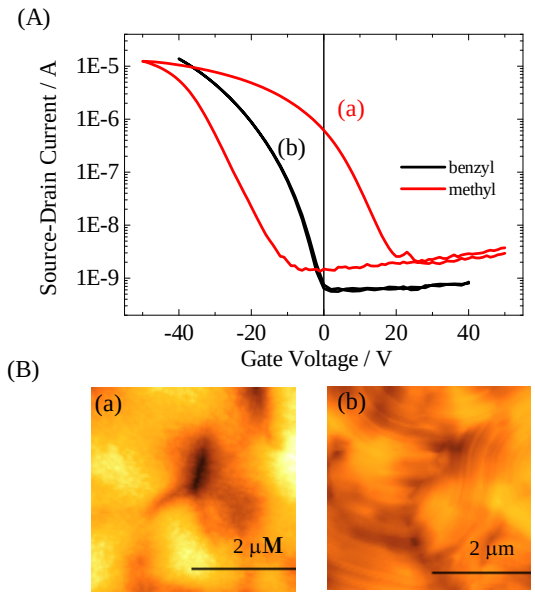

Fig. 2 Transfer characteristics of OTFT (A) fabricated with PMLG (a) and PBLG(b) as a gate dielectric and AFM images of these surface of PMLG film (a) and PBLG film (b).

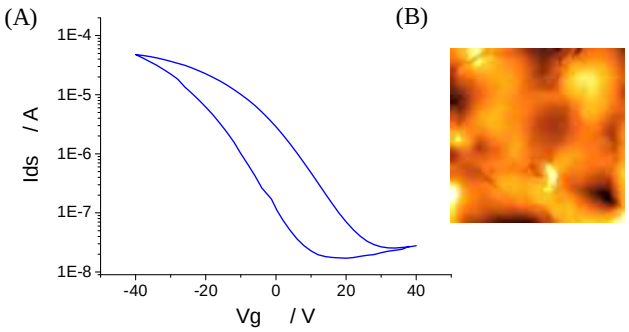

Fig. 3 Transfer characteristics of OTFT (A) fabricated with PBLG containing 20wt.% PMMA as a gate dielectric and AFM images of the surface of PBLG (B).

In order to confirm the assumption, the effect of the blending PMMA with PBLG was investigated. Fig.3 shows transfer characteristic of OTFT fabricated with PBLG containing 20wt. % PMMA [Fig.3(A)] and the AFM image of the surface of gate dielectric layer [Fig.3 (B)]. In the electric property, the large hysteresis was observed and fibrous crystal structure was disappeared. From these results, it reveals that the ferroelectric hysteresis behavior of poly (glutamate) film is observation when the interaction between main chains is decreased.

4. Conclusions

An OTFT memory fabricated with PBLG film as a gate dielectric layer dose not show the hysteresis property in the transfer characteristic due to aggregation of PBLG. In the OTFT memory device fabricated with poly (glutamate), it is important to prevent the interaction between main chains. As a method, it is effective to add an insulator material such as PMMA.

Acknowledgements

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