Title:	Electroactive Organic Ferroelectric Thin Film: Advancing Memory Window and Retention Behavior.
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Abstract:	Organic memory has emerged as a promising technology in future electronic devices such as flexible smart labels, electronic paper, contactless identification transponders, RFID tags, and other consumer electronics. In this context, non-volatile memory is one of the essential components of an electronic circuit that can store and retrieve data whenever it is required. Among various types of memory, non-volatile memory comprised of organic ferroelectric polymers is witnessing a surge in demand due to their low temperature processability, chemical resistiveness, superior flexibility, and lightweight. Ferroelectric polymers, such as polyvinylidene fluoride (PVDF) and its copolymer with trifluoroethylene (TrFE) (i.e., P(VDF-TrFE)) are significant materials for memory devices, on the account of prospective advantage, such as their easy solution processing fabrication, wide range of selective solvents and ability to integrate with silicon-based technology. Noteworthy to mention that P(VDF-TrFE) copolymer has been extensively studied in non- volatile memory applications because of its thermodynamically stable ferroelectric β- phase and low-temperature solution processability. However, large-scale integration is hindered by the limited thermal stability, due to its low Curie transition temperature of P(VDF-TrFE). Besides this, higher cost (almost 10 times higher than PVDF) is one of the prime bottlenecks for industrial bath fabrications. On the other hand, PVDF has the highest thermal stability due to the physical absence of Curie transition temperature, however, limited attempts have been carried out for the ferroelectric memory applications since the nucleation of electroactive crystalline phases is the main hurdle. Therefore, the present thesis is mainly focused on the nucleation of the electroactive phases in PVDF thin films and then ferroelectric memory-related prime parameters, such as memory window, switching voltage and retention behavior are studied. In particular, the ice quenching technique is introduced to nucleate the electroactive δ- phase, in thin PVDF film that rules out the requirement of the higher electric field. The requirement of high electric field (~MV/m) is not suitable for thin film-based memory devices since it often causes electrical breakdown. To study the memory properties, we have employed a metal-ferroelectric-insulator–semiconductor (MFIS) diode with δ-PVDF thin film as the ferroelectric layer. It displayed a capacitance–voltage (C–V) hysteresis with a notable memory window of 7.5 V up to a temperature of 140 °C, overcoming lower fatigue temperature which is limited by Curie transition in the case of P(VDF-TrFE) vcopolymer. In addition, ferroelectric (i.e., “write”, “erase” and “read” pulses) and retention responses of δ-PVDF thin film have been demonstrated through piezoresponse force microscopy (PFM). Here, we observed a very stable polarization state of 10 h, where only a 5 % loss of its initial capacitance value is noticed. So, we affirmed that δ-PVDF thin film have higher thermal stability, better retention, and wider operating temperature range compared to P(VDF-TrFE) copolymer, which could make it an excellent alternative low- cost ferroelectric memory device. Furthermore, we have attempted electroactive on β- phase in PVDF thin film where preferential edge-on orientation is achieved by the incorporation of external filler of copper chloride dihydrate (Cu salt). Here we have employed orientation-controlled spin coating (OCSC) technique. Briefly, we tried to present a clear understanding between the interfacial interactions of the Cu salt with PVDF, which favours the edge-on β-crystallites formation through hydrogen bonding. Formation of the edge-on β-crystallites in Cu salt meditated PVDF (Cu/PVDF) thin film, promise the low-voltage ferroelectric switching compared to neat PVDF counterpart. The capacitance−voltage (C−V) characteristic of the MFIS diode (with Cu/PVDF thin film as a ferroelectric layer) exhibits a memory window of 12 V, which is more than twice then the value obtained in neat PVDF. In addition, we have also observed stable retention of more than 7 days. In continuation with this work, we have also attempted the electroactive β-phase nucleation with iron, cobalt and nickel containing hydrated salts mediated PVDF thin films. The X-ray photoelectron spectroscopy is conducted to understand the underlying mechanism of β-phase nucleation and their respective edge-on or face-on orientation. Thus, we affirmed the preferential formation of β-phase with edge-on orientation in metal-hydrated salt-mediated PVDF thin films that exhibit low switching voltage, good retention and memory window. It indicates that additional electrical poling or external stretching is completely possible to be ruled out, thus offering a new prospect for the evolution of devices with long-lasting nonvolatile ferroelectric memories. Apart from the study of metal hydrated salt mediated PVDF thin films, we have also attempted to fabricate the edge-on oriented β-phase in PVDF and montmorillonite (MMT) clay nanocomposite thin film. It is worth noting that the addition of MMT into the PVDF matrix, not only improves the ferroelectric property (i.e., the ferroelectric switching at a lower voltage and improved retention) but also it controls the leakage current due to intercalation properties. Furthermore, we found that ferroelectric retention in MMT- embedded PVDF thin film is stable for longer than 20 days (> 480 h) with no degradation viin the switched domains, as probed by the PFM. It also exhibits a memory window of 7 V with temperature stability up to 150 °C. In summary, we have nucleated electroactive δ and β phases in PVDF thin film to obtain lower switching voltage, large memory window, long retention, and controlled leakage current that could offer the futuristic application for non-volatile organic ferroelectric memories.
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