Abstract

The demand for high-performance flexible non-volatile memories has escalated to new highs with applications extending to flexible and wearable electronics, healthcare, computation, radio frequency identification (RFID) tags, foldable displays, sensors, and data storage. The resistive random access memory (RRAM) devices, a class of non-volatile memories, have emerged as the game-changer in the field of flexible electronics with tremendous capabilities such as high mechanical strength, flexibility, transparency, low power consumption, fast switching speed, miniaturization capabilities, and compatibility with CMOS technology. Operation of RRAMs is based on the physical phenomenon of resistive switching (RS), which is the capability of selective organic, metal oxides, perovskites, and low-dimensional dielectric materials to demonstrate multiple electrical resistance states namely high and low resistance states (HRS and LRS respectively), regulated under the effect of an external bias. In this work, a hybrid bilayer of HfO_x and PVP composite with 2D materials has been presented for a larger memory window, lower power consumption, and greater flexibility.

During the initial work, the single-layer high-k metal oxide dielectrics were explored where the switching behavior of the device was studied with optimized thickness and deposition temperature of the active layer. Based on this study, a novel multi-temperature deposition scheme using atomic layer deposition was reported with AlO_x as the switching layer and Ti as the bottom and top electrode. Using this technique, an artificial concentration gradient was created within the switching layer by varying the deposition temperature. Due to different deposition temperatures, oxygen vacancy concentration was different in each layer of the MTD film, which led to the formation of a conductive filament with nonhomogeneous strength and dimensions across the film. This technique has resulted in improved performance in devices with high reliability, higher I_{on}/I_{off}, comparatively low voltage switching operation, repeatability of 75 DC cycles, and retention time of over 10^3 s. However, these devices exhibited the poor memory window, which needs to be higher for proper readout operation.

This issue was later resolved by using an ultra-thin 5 nm HfO_x layer with poly(4-vinylphenol) (PVP) as the active layer with Ag as top electrode, that demonstrated decent switching behavior with switching voltages less the 1 V, an endurance of over 2000 cycles, and ~80 of I_{on}/I_{off}. This improvement in device performance has been attributed to the pinhole assisted electromigration of Ag ions through the switching layer that eventually guided the filament growth and dissolution process. The pinholes formation over the PVP surface was confirmed using the atomic force microscopy. Furthermore, the confined growth of conductive filament resulted in improved reliability and the memory window. Also, the pinholes in the PVP film don't affect the device-to-device variability, and the sample yield was measured to be >75%. The only concern with these devices was that the devices couldn't produce the memory window up to the standard of the state-of-the-art technology. This is possibly due to the pinholes on PVP surface, causing a rise in the leakage current and hence needs to be eliminated.

The gradually improving performance of PVP/HfO_x hybrid switching layer based RRAMs was the motivation for further exploration for low-voltages switching operation with enhanced memory window and electro-mechanical stability. To achieve this objective, the graphene oxide (GO) was mixed with PVP to prepare composite, which was later explored as the secondary layer in flexible RRAMs. The Ag/PVP:GO/HfO_x/ITO demonstrated excellent resistive switching behavior with switching voltages 0.6 V and -1.46 V with an excellent memory window larger than 10^6 for more than 10^4 s, and no performance degradation till bending radius till 5 mm. The enhanced memory window is attributed to the removal of pinhole in the composite film and hence lesser leakage current. Also, the incorporation of GO in the PVP solution drives better control over the filament growth and dissolution process that further enhanced the memory window, repeatability, and reliability. Moreover, the flexible devices exhibited a memory window of $>10^3$ even after undergoing large mechanical strain (corresponding to a 5-mm bending radius). In addition, after 150 cumulative cycles of consecutive tensile and

compressive strain at a 5-mm bending radius, flexible RRAMs demonstrated a clear memory window of 4×10^3 for 10^4 s.

As a final point, the GO was replaced with molybdenum disulfide (MoS_2) that enhanced the device performance by reducing the switching current to the nano ampere range and hence pulling down the power consumption to sub nanowatt range. Furthermore, the flexibility study of these devices demonstrated excellent mechanical strength up to a bending radius of 2.5 mm and 100 cycles of continuous cycles of compressive and tensile strain at a ±5 mm radius. The ultra-low ON current in these devices has been attributed to the formation and dissolution of multiple weak nanosized conducting filaments created due to the electromigration of Ag ions through the switching layer.

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