Abstract

Flexible electronics has emerged as an important technology due to its versatility and potential applications in various on-body wearables, electronic skin, smart prosthetics, and various devices for health monitoring. In recent years, resistive random-access memory (RRAM) has been extensively explored as a suitable non-volatile memory device for flexible electronics owing to its simple structure, ease of fabrication, and the ability to incorporate various engineered and solution processed switching layers which are suitable for low cost, low temperature and large area processing. Though there have been continuous efforts to enhance the performance of the solution processed flexible RRAMs, there have been multiple issues such as poor endurance and cycle to cycle variability which need to be addressed. This work is an effort towards applying strategies to address the performance issues of solution processed flexible RRAM devices. More specifically, it involves comprehensive exploration of composites of solution processed polymers and 2D materials aiming to improve the performance parameters as well as shelf-life stability. Moreover, multilevel resistive switching in flexible RRAM devices is also demonstrated.

To start with, resistive switching behavior of bilayer of poly(4-vinylphenol) (PVP): molybdenum disulfide (MoS_2) composite and TiO_2 in RRAM devices were explored on both rigid and flexible substrates. Flexible devices fabricated with this PVP:MoS_2/TiO_2 bilayer on polyethylene naphthalate (PEN) substrate operated on low average SET and RESET voltages (V_{SET} & V_{RESET}) of 1.5 V and −0.7 V with decent endurance of 200 cycles and high I_{ON}/I_{OFF} of more than 10^3 at reading voltage of 0.2 V. These devices maintained decent switching upon bending up to a radius of 7 mm.

The next engineered combination was poly(N-vinylcarbazole) (PVK):MoS_2/TiO_2. Flexible RRAM devices with this hybrid bilayer exhibited excellent switching with low V_{SET} and V_{RESET} of 1.2 and -1.5 V respectively, high endurance of 1000 cycles, and high stability upon bending with radii up to 7 mm for 100 cycles. A comprehensive investigation of long-term environmental stability and performance was also performed by systematically characterizing the devices for a duration of 20 months and it was found that devices were able to maintain their switching behavior with excellent retention of 10^4 s even after being in ambient environment for this long duration, indicating high shelf life. Moreover, effect of exposure to humidity and heating were studied confirming good stability against these. These investigations indicated suitability of PVK:MoS_2/TiO_2 as a switching layer candidate for environmentally stable RRAM devices for flexible electronics.

Finally, composite of a rarely explored 2D material ($MoSe_2$) with PVP (PVP:$MoSe_2$) was demonstrated as a potential resistive switching layer for flexible RRAM devices. Fabricated flexible RRAM devices exhibited forming-free and excellent resistive switching with low V_{SET} & V_{RESET} (0.7 V / ~ -1 V), high DC endurance of more than 1000 cycles, and excellent retention time of 10^4 s with decent I_{ON}/I_{OFF} of ~10^3. These devices exhibit multilevel resistive switching for 2-bit storage (four levels), by tuning the compliance current values, which can be the simplest way to achieve this functionality. This study also opens up a direction for exploration of other unique material combinations for switching layers towards application in multibit storage, as it may contribute to low cost, high density, and nonvolatile flexible RRAM devices.