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論　　文　　要　　旨

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主論文題名

Application of Nanocarbon Materials for Long-Term Reliability Improvement of Copper Metallization

内容の要旨

Semiconductor memory (SM) devices, which can be used as storage media in various applications, have been extensively developed. Because of the rapid increase of digital recording in the world, SM devices are also expected to use for long-term storage of valuable digital data over 100 years. Regarding advanced SM technologies, copper (Cu) has been widely used as interconnect and bond pad material in place of Al to enhance the electrical conductivity and current endurance. Therefore, the long-term reliability of Cu used in SMs is a serious concern for the long-term storage application (>100 years). A potential cause for the failure of long-term data storage is Cu oxidation due to moisture. To avoid Cu oxidation, an impermeable thin film coat can be used. Nanocarbon materials, such as graphene and amorphous carbon (a-C) composites, are excellent candidates due to their small atomic radius of carbon. In the beginning, high-quality large-grained graphene was studied its potential and issues in preventing the Cu surface from oxidation in principle because its forming temperature is still high and not compatible with the current LSIs fabrication. Then, the properties of room-temperature nitrogen-doped a-C (a-C:N) were investigated and expected as a practical method to maintain the Cu surface from oxidation for more than 100 years. Before improving the long-term resistance of Cu against oxidation due to moisture, the effects of Cu oxidation on the sheet resistance were studied under accelerated conditions using a temperature humidity storage (THS) test. In addition, the dependency of Cu oxidation on the temperature and humidity was studied, then, the prediction model of Cu lifetime and its important parameters were derived. The Cu lifetime against humidity was estimated based on the proposed prediction model.

Large-grain single-layer graphene (SLG) was deposited by high-temperature chemical vapor deposition (CVD) for coating on the Cu surface. SLG was demonstrated the potential as an atomic-layer barrier against moisture. The THS test shows that large-grain SLG protects large areas of the Cu surface and only areas with grain boundaries and defects were oxidized.

To avoid the Cu oxidation in areas with grain boundaries and defects, stacked SLG layers, that is, double-layer graphene (DLG) and triple-layer graphene (TLG), were fabricated. The DLG- and TLG-coated Cu samples were tested and compared with bare Cu without a barrier and SLG-coated Cu samples. The results of the Raman and X-ray photoelectron spectroscopy (XPS) measurements indicate that the SLG grain boundaries and defects enhance the Cu oxidation due to the formation of galvanic cells during long-term THS tests. However, DLG can be used to solve this issue. It can be used to cover the areas of SLG grain boundaries and prevent galvanic corrosion during the THS test. In addition, TLG can be used to protect the Cu surface, ensuring the efficiency of graphene in protecting the sample surface from oxidation due to moisture.

However, it is still difficult to apply graphene as moisture barrier of Cu metallization in current LSIs fabrication because of the high-temperature deposition of graphene. Therefore, a-C:N was studied as an alternative nanocarbon material that can be simply deposited by sputtering at room temperature. The THS test results show that a-C:N with an Ar:N_2 ratio of 90:10 can preserve the sheet resistance of an a-C:N/Cu sample for ~400 years (at 27℃/60% RH) based on the proposed prediction model. The XPS depth profiles obtained after the THS test confirm the excellent efficiency of a-C:N in preventing Cu oxidation due to moisture.

This work provides promising guidelines for the improvement of the long-term reliability of Cu metallization used in advanced SM devices and thus increase their lifetime for the storage of valuable digital data over 100 years.

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