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Deposition of silicon dioxide films on amorphous carbon films by plasma enhanced chemical vapor deposition for low dielectric constant interlayer dielectrics

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Deposition of silicon dioxide films on fluorinated amorphous carbon films (*a*-C:F) for low dielectric constant interlayer dielectrics was investigated. Both SiO_2 and *a*-C:F films were deposited by helicon wave plasma enhanced chemical vapor deposition with C_4F_8 for *a*-C:F and SiH_4+O_2 mixtures for SiO_2. The SiO_2 films on the *a*-C:F films peeled off soon after deposition. However, the peeling was suppressed by inserting a thin *a*-C:H buffer layer grown from CH_4 between them. The adhesion between the films was increased by making the stoichiometry of SiO_2 Si-rich. It was found that the Si–C bonds formed at the interface increased the adhesion. © *1997 American Institute of Physics.* [S0003-6951(97)03509-2]

For the reduction of the delay time of ultralarge scale integrated (ULSI) circuits caused by the parasitic capacitance of multilevel interconnections, it is attractive to use low dielectric constant (low-k) materials as interlayer dielectrics (ILD). It is known that the incorporation of fluorine into the SiO_2 film decreases its dielectric constant. Many groups are now investigating SiOF films with a dielectric constant around 3.5.[1,2] However, for the further shrinkage of the design rule of interconnections, it is required to use a low-k ILD with a dielectric constant below 3. Low dielectric constant polymers such as polytetrafluoroethylene (PTFE), fluorinated polyimide, and benzocyclobutene (BCB) are now promising because their dielectric constants are lower than those of inorganic materials.[3–5]

Recently, we have developed fluorinated amorphous carbon thin films (*a*-C:F), which have both a cross-linking structure, as in a polyimide, and C–F bonds, as in PTFE, for new low dielectric constant interlayer dielectrics of ULSI interconnections,[6,7] The *a*-C:F films can be deposited with a high deposition rate (around 300 nm/min) using a helicon wave plasma enhanced chemical vapor deposition (CVD) method from 100% C_4F_8 and CH_4 gases. The dielectric constant of the *a*-C:F films was 2.3, which was much lower than that of the conventional SiO_2 dielectrics ($\varepsilon_r=4.0$). In a previous report, it was revealed that the gaps between aluminum wiring (width 0.5 μm, height 0.6 μm, gap 0.35 μm) were completely filled with *a*-C:F films by the application of bias power to the substrate.[8]

For further integration of low-k ILD, the deposition of ILD will be followed by the patterning, etching, and chemical mechanical polishing (CMP) process for global planarization. However, incorporation of the organic film into the conventional interlayer process seems to be difficult because of its low resistance to oxygen plasma, and weak mechanical strength. Thus, an SiO_2 film which has adequate mechanical strength and which can prevent irradiation of oxygen plasma to organic low-k ILD, will be required on the organic ILD. However, it is not trivial to deposit the SiO_2 film on the *a*-C:F films because of low adhesion due to fluorine atoms. In this letter, we investigated the SiO_2 deposition on the *a*-C:F film by controlling the interface between the *a*-C:F and the SiO_2 films.

The plasma enhanced CVD reactor used in this study was helicon wave plasma reactor (ANELVA I-4100-HBiC). The source gases were C_4F_8 for the deposition of *a*-C:F films, and SiH_4, O_2, and Ar for SiO_2 deposition. The details of the *a*-C:F deposition in the helicon wave plasma reactor have been described in a previous letter.[7] The 13.56-MHz, 3-kW rf source was coupled to a helical coil wound around the quartz tube. The 400-kHz bias power was applied to the substrate. 6-in. Si wafers with AlSiCu line and space patterns (width 0.5 μm, height 0.6 μm, gap 0.35 μm) were used as substrates. The helicon source power of *a*-C:F film deposition was 2 kW. To fill the *a*-C:F films between aluminum wiring, 30-W bias power was applied to the substrate.[8] The helicon source power of the SiO_2 deposition was 2.5 kW, and the flow ratio of O_2/SiH_4 was varied. 13.56-MHz, 800-W bias power was applied to the substrate in the case of SiO_2 deposition. The chemical compositions of *a*-C:F and SiO_2 films were measured by x-ray photoelectron spectroscopy (XPS).

When the *a*-C:F films were deposited with C_4F_8 directly onto the SiO_2 substrate, the *a*-C:F films peeled off from the substrate as reported previously.[6] However, the peeling was suppressed by inserting a thin hydrogenated amorphous carbon (*a*-C:H) buffer layer (30 nm) between the substrates and the *a*-C:F films.[6] Next, we investigated the direct deposition of SiO_2 films on *a*-C:F films. The SiO_2 film was grown from SiH_4+O_2 mixtures. In this case, O_2 and SiH_4 flow rates are 100 sccm and 50 sccm, respectively, so the O_2/SiH_4 mixing ratio was 200%. The SiO_2 film on *a*-C:F peeled off soon after its deposition because of low adhesion, but this peeling was suppressed by inserting a thin *a*-C:H buffer layer between the *a*-C:F and SiO_2 films. Thus, SiO_2/*a*-C:H/ *a*-C:F/ *a*-C:H/SiO_2 multi-layered structures were required to use *a*-C:F and SiO_2 films as a multi-ILD.

Figure 1 shows the results from an adhesion yield test of SiO_2 films grown on the *a*-C:H films as a function of O_2/

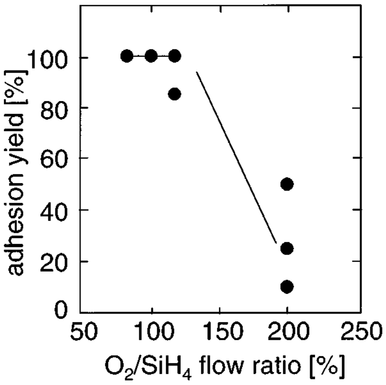

FIG. 1. Adhesion yield of SiO_2 films deposited on the a-C:H film by scotch tape peeling test.

SiH_4 flow ratio. In this test, scotch tape was used to remove the film that adhered to it, and the adhesion yield indicates the area of the film that remained after the peeling test. The SiO_2 film with the 200% O_2/SiH_4 mixing ratio resulted in poor adhesion by the peeling test. However, as the mixing ratio of O_2/SiH_4 decreased, the adhesion yield increased. Figure 2 shows the XPS depth profile of the interface between SiO_2 and a-C:H films. The Si $2p$ peaks of SiO_2 film with an O_2/SiH_4 flow ratio of 200% appeared at their normal SiO_2 position. With an O_2/SiH_4 flow ratio of 100%, the Si $2p$ peaks shifted towards a lower binding energy, and Si sub-oxide peaks at around 100 eV were detected. This indicates that the stoichiometry of the SiO_2 film was changed to Si-rich allowing the Si–Si bonds to form. Thus, it was revealed that the adhesion of the SiO_2 and a-C:H film was dramatically increased by making the stoichiometry of the SiO_2 film Si-rich.

On the other hand, the C $1s$ peaks of the a-C:H film at the interface between the SiO_2 film with the mixing ratio of 200% appears at the same energy as the C $1s$ peaks from the inner a-C:H film. However, the spectrum of the film with the mixing ratio of 100% changed to a lower binding energy and approached the energy of the Si–C bond at 282.5 eV reported by Smith *et al.*[9] from α-SiC. This indicates that the SiO_2 bonds were formed at the a-C:H/SiO_2 interface by making the stoichiometry of the SiO_2 Si-rich. Thus, the improved adhesion between the SiO_2 and a-C:F films may be due to the Si–C bonds formed at the interface between the Si-rich SiO_2 and a-C:H film.

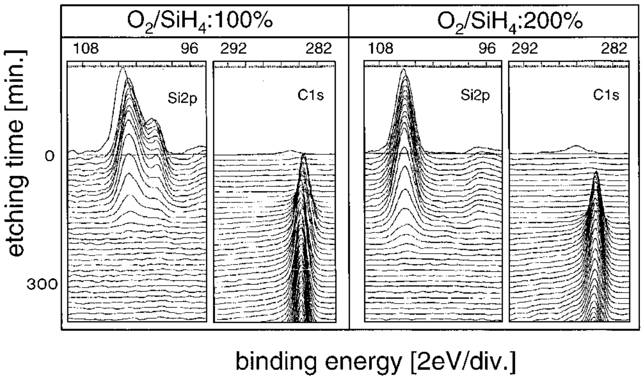

FIG. 2. XPS depth profile of the adhesion layer consisting of Si-rich SiO_2 and a-C:H film.

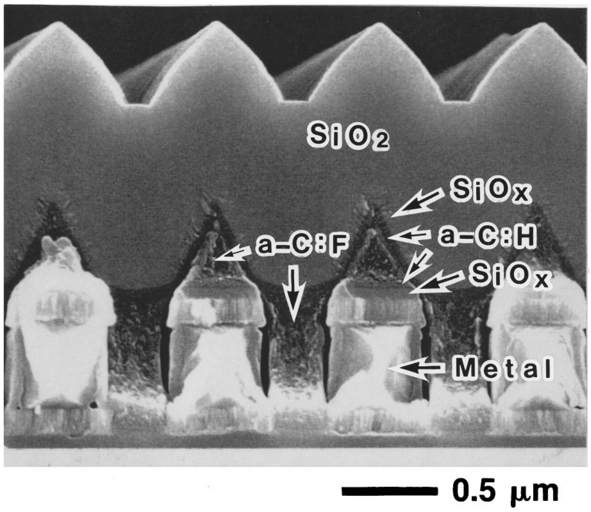

FIG. 3. SEM image of a multi-layer ILD that consists of a-C:H, a-C:F, Si-rich SiO_2 and normal SiO_2 films.

Figure 3 shows the scanning electron microscope (SEM) image of a multi-interlayer dielectric film that consists of a a-C:F film, an adhesion layer (a-C:H and Si-rich SiO_2), and a normal SiO_2. By applying a 30-W bias power to the substrate, the wiring gaps could be completely filled with the a-C:F film, and a triangular shaped a-C:F film was formed on top of the aluminum wiring. By inserting the adhesion layer, a SiO_2 film could be attached to the a-C:F without peeling. The top of the SiO_2 film also had a peaked shape. This rugged surface is suitable for the CMP process, because the convex SiO_2 will be completely polished by the CMP process. The deposition of the ILD and the CMP process can be followed by a conventional interlayer process (resist coating, etching, and resist removal), because the resistance to oxygen plasma and the mechanical strength of the SiO_2 film will fully protect the a-C:F film.

In summary, a SiO_2 film can be deposited on a a-C:F film by applying a a-C:H adhesion layer. The adhesion of the SiO_2 films on a-C:H films was increased by making the stoichiometry of the SiO_2 film Si-rich. Evidence was found that Si–C bonds were formed at the interface between the aC:H and Si-rich SiO_2 films.

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