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Non-heat assistance chemical vapor deposition of amorphous silicon carbide using monomethylsilane gas under argon plasma

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ABSTRACT

Without any heating assistance, argon plasma and monomethylsilane gas were used for forming silicon carbide film on various substrates, such as aluminum, stainless steel and polyimide. After cleaning the substrate surface by an argon plasma treatment at less than 10 Pa and 0.36 W/cm^2, the monomethylsilane gas at the concentration of 5% was introduced into the argon plasma at 10–15 Pa within 20 min. Based on this process, a dense amorphous silicon carbide film thicker than 200 nm could be produced. The obtained film could perfectly protect the aluminum substrate surface from corrosion by a hydrogen chloride aqueous solution. The same process could form a silicon carbide film on the surfaces of a stainless steel plate and polyimide film.

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1. Introduction

Silicon carbide (SiC) is one of the suitable coating materials for protecting surfaces of various useful and functional materials from a harsh and high-temperature environment [1]. As a typical application, the surface of the susceptor used in the chemical vapor deposition (CVD) reactor is very effectively coated by a silicon carbide film [2]. Such an application is possible, because of the chemical, thermal and mechanical stable nature of the silicon carbide. The silicon carbide coating film is very tolerant to the corrosive environment using hydrogen chloride gas at high concentrations and high temperatures during the CVD reactor cleaning process.

In order to widely apply the suitable nature for coating the surfaces of various materials including low melting point materials, such as aluminum and organic polymers, the silicon carbide deposition temperature should be decreased as much as possible. For achieving the best application of a silicon carbide coating, a room temperature process is desirable.

For this purpose, the authors have studied the room temperature process using the monomethylsilane (MMS) gas without any heating assistance [3,4]. The previous process utilized the soft plasma etching for activating the substrate surface so that the monomethylsilane could be physisorbed after terminating the plasma. However, because the silicon carbide content in the film was unfortunately low, it should be significantly improved. Additionally, the technique should be available for forming films on various material surfaces.

In order to produce various films on various substrates at low temperatures, the plasma enhanced CVD (PECVD) [5–8] technique has been a very popular method. For developing a simple process using the single precursor, the PECVD using monomethylsilane gas has been studied by many researchers [9–13]. Most of them used high power plasma with the help of substrate heating for adjusting and optimizing the film properties, such as the poly-types and film stresses. Based on the chemical bonding strength between silicon, carbon and hydrogen atoms, the soft adjusted plasma may break the weak bonding, such as hydrogen with silicon (less than 299 kJ/mol) and carbon (338 kJ/mol), while maintaining the strong bonding between silicon and carbon (452 kJ/mol) [14]. The silicon–carbon pair is expected to be physisorbed on the activated substrate surface to form the silicon carbide film.

In this study, the chemical vapor deposition process without any heating assistance was developed for producing a silicon carbide film on the substrate surfaces of aluminum, stainless steel and polyimide using the argon plasma and the monomethylsilane gas, without any heating assistance.

2. Experimental procedures

The typical surface chemical process designed in this study is shown in Fig. 1. The substrate surface is usually oxidized and has an airborne contamination, as shown in Fig. 1(a). The argon plasma physically removes the adsorbed atoms on the material surface for producing the dangling bonds, as shown in Fig. 1(b). Fig. 1(c) shows the decomposition of the monomethylsilane molecule introduced into the reactor chamber. The hydrogen atoms are assumed to be removed from the silicon and carbon by means of the argon plasma, while the silicon–carbon bonds

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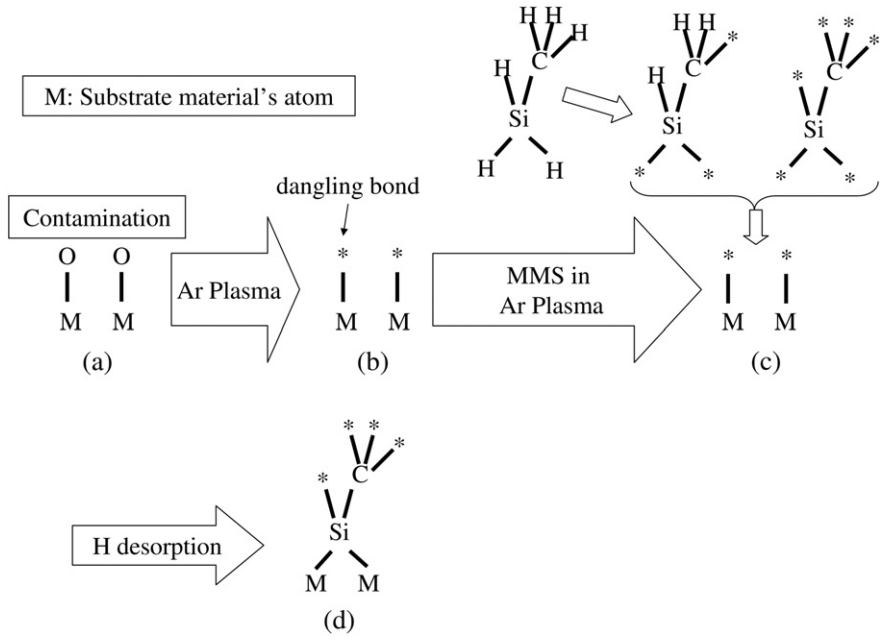

Fig. 1. Gas phase and surface process for activating substrate surface and forming silicon carbide film.

are expected to be maintained. The reactive fragment, such as the silicon–carbon pair, is chemisorbed by the dangling bonds at the substrate surface. Finally, the silicon carbide film is formed as shown in Fig. 1(d). Because the intermediate species remains on the film surface at the termination of the film deposition, the surface is oxidized to produce silicon oxide in the ambient air.

Fig. 2 shows the reactor and process developed in this study. The metal substrates used were the plates made of aluminum and stainless steel (SUS430), which are a low cost materials manufactured for handicraft use. Additionally, a polyimide film (Kapton 100H, Toray Industries, Inc., Japan) was used. Because the metallic materials have been exposed to air for a sufficiently long period, their surfaces were covered with a native oxide film. The substrates having the dimensions of 10 mm wide and 10 mm long were cut from these plates and were cleaned by ethanol without any additional wet cleaning. The surface morphology of the substrate materials was observed over the area of 30 cm × 25 cm, for the aluminum, stainless steel and polyimide, in order to confirm that there was the uniform surface morphology. The substrate pieces were cut from the same sheet. The typical morphology of the substrate and that of the obtained film at each center position were compared.

The reactive substrate surface was prepared by argon plasma etching, following Fig. 2 (A). Step (A) is the plasma etching using a parallel plate plasma reactor (Soft Plasma Etcher-SE, Meiwafosis Co., Ltd., Tokyo, Japan). This reactor consisted of two electrodes, two fluorocarbon resin parts and quartz vessel. The substrate was placed on the bottom electrode surface. Typically, the plasma power is less than 0.36 W/cm^2. Step (A) prepared the reactive surface by etching for 10–20 min in the argon plasma at 8 Pa and room temperature. Next, in Step (B), the monomethylsilane gas at 5% in argon gas was introduced for 10–20 min into the chamber containing the argon plasma. The partial pressure in

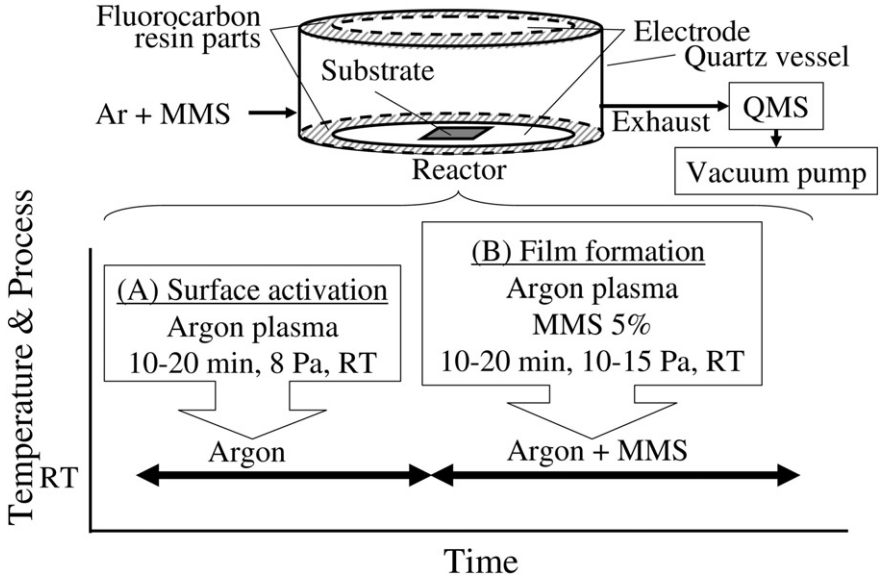

Fig. 2. Reactor and process developed in this study. (A) is the surface activation process using argon plasma, and (B) is the silicon carbide deposition process using monomethylsilane gas with argon plasma.

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