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Deposition of uniform and well adhesive diamond layers on planar tungsten copper substrates for heat spreading applications

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Abstract

For a more effective heat spreading we deposited thick diamond layers on tungsten copper substrates, which are used as heat sinks in the microelectronic industry. Disadvantages of high growth rate CVD techniques like microwave plasma assisted CVD (MWPACVD) or DC plasma jet CVD are the need for flat and preferable discoidal substrates, the limitation of the total substrate surface and the uniformity of the deposited layers. Thus, the uniform deposition of diamond on a large number of substrates with a rectangular or more complex geometry (like a 3D geometry) is limited. By using hot filament CVD (HFCVD) the reactor size is not limited and it is possible to increase the batch size. Even at typically lower growth rates in HFCVD the total coated surface can over-compensate this disadvantage and renders HFCVD economically more viable. We were able to scale up the number of substrates per HFCVD batch up to 240 pieces which is equal to a coating surface of 480 cm². The substrate temperature was kept between 820 and 870 ◦C to avoid copper diffusion to the surface and evaporation of copper. An average diamond growth rate of 0.23m/h allows the deposition of diamond layers in less than half of the time necessary by using, e.g. microwave plasma assisted CVD at typical process conditions for the same surface (1 μ m/h growth rate; substrate temperature of about 850 °C; 45.5 cm² coating surface per batch). In spite of the significantly increased coating surface it was possible to deposit uniform diamond layers with a high diamond quality. A further challenge is the bending of the substrates during cooling down caused by the different coefficients of thermal expansion for diamond and WCu. Cooling devices require a planar geometry. It was possible to prevent the substrates from bending after cooling down by using a special pretreatment. © 2005 Elsevier B.V. All rights reserved.

Keywords: Diamond; Thermal properties; Tungsten; Adhesion; Surface and interface states; Surface stress

1. Introduction

The application of CVD diamond for thermal management is still limited by the high price of the material. Because of its outstanding properties like very high thermal conductivity combined with the possibility of heat spreading CVD diamond is the most promising material for thermal management applications. The deposition of diamond on tungsten copper (WCu) substrates, which are used as heat sinks in microelectronic industry, combines the good thermal conductivity of WCu $(200-240 \text{ W/mK}$ [\[1\]\)](#page--1-0) with the heat spreading properties of diamond. Moreover, the coefficient of thermal expansion is similar to that of common semiconductor materials and therefore ensures a long lifetime of the whole package.

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As a film thickness between 50 and 100 μ m with a high quality of the deposited diamond film is necessary for effective heat spreading [\[2\],](#page--1-0) one main driver for cost is the slow growth rate of CVD diamond.

Microwave assisted plasma CVD (MWPACVD) or DC plasma jet systems allow the deposition of diamond layers with high growth rates combined with a high purity of the deposited layers $[2-8]$. Up to $40 \mu m/h$ or more are possible by using DC plasma jet [\[8\],](#page--1-0) up to $30 \mu m/h$ by using microwave plasma assisted CVD [\[3\]. D](#page--1-0)isadvantages of these systems are the need for flat and preferable discoidal substrates, the limitation of the total substrate surface and the uniformity of the deposited layers. Thus, the uniform deposition of diamond on a large number of substrates especially with a rectangular or more complex geometry (like a 3D geometry) is limited. Furthermore, high substrate temperatures are necessary to achieve high growth rates [\[3,6–8\]](#page--1-0) and it is not possible to vary the substrate positions in all three dimensions. In several works scaling to large area substrates was

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reported [\[8–11\].](#page--1-0) Substrate diameters of up to 15 cm were used [\[8–10\].](#page--1-0)

The growth rates by using hot filament CVD (HFCVD) are much lower $(0.1-0.2 \text{ mm/h})$ [\[5\],](#page--1-0) but as the substrates can be placed more flexible inside the reactor it is possible to increase the batch size significantly. If the number of pieces in one process is large enough, this CVD technique is more economical in many cases.

The need for very thick diamond layers causes the substrates to bend which is also unwanted in heat spreading applications. Tensile and compressive residual stresses are imposed on the metal and the diamond layer during cooling. These residual stresses are caused by the different coefficients of thermal expansion and can be expressed by the following equation assuming elastic deformation:

$$
\sigma_{\text{Film}} = \int_{T_0}^{T} \frac{E_{\text{Dia}}}{1 - \nu_{\text{Dia}}} \Delta \alpha \, \text{d}T \tag{1}
$$

where E_{Dia} and v_{Dia} are the Young's modulus and the Poisson constant of the diamond film, $\Delta \alpha$ is the difference in thermal expansion coefficients of the film and the substrate, *T* the deposition temperature and T_0 the room temperature. Bending stresses develop to balance the bending moments induced by the compressive residual stresses. It is important to differ between elastic, partially plastic and fully plastic deformation in the substrate [\[12\].](#page--1-0) Cooling devices require a planar geometry which can be achieved by preventing the substrates from bending and by the deposition of planar diamond layers.

2. Experimental

Diamond deposition took place in an industrial hot filament reactor (CemeCon CC800Dia; see Fig. 1) at total flow rates between 1000 and 3000 sccm containing hydrogen and 0.3–1.3% of methane. The process pressure was between 10 and 20 mbar. All the results were obtained by using tungsten or tantalum filaments. The diamond layers were deposited on WCu substrates with a dimension of $20 \text{ mm} \times 10 \text{ mm} \times 1 \text{ mm}$. Copper was removed from the substrate surface via etching.

The substrate temperature was kept between 820 and 870 ◦C to avoid copper diffusion to the surface and evaporation of copper. The use of different creep and temperature resistant substrate holders and an effective 3D arrangement of the substrate holders following the temperature distribution inside the deposition chamber ensured the uniform diamond deposition on up to 240 substrates simultaneously. The filaments were arranged vertical in rows and two substrates were placed in front of each filament, respectively. A schematic drawing of the used set-up is given in Fig. 1.

After deposition the film thickness was measured by infrared interferometry assuming the refractive index of diamond is 2.4 over the analyzed spectral range $(700–4000 \text{ cm}^{-1})$. Moreover, an influence of the deposition parameters was neglected. In addition, cross sectional SEM investigations were carried out to verify these measurements. All results concerning the growth rate are displayed in carat per hour taking the average growth rate of all substrates of each process into account. The phase boundary diamond/substrate was determined by X-ray-diffraction (XRD). Measurements were carried out at an angle between 30◦ and 130◦.

The quality and the residual stresses of the diamond coatings were calculated using the effective Raman spectra (Ar⁺-Laser, λ = 514 nm). The spectra were fitted by Lorentzian type peaks for the diamond peak and by Gaussian type peaks for the different graphitic sites. Background was subtracted. To quantify the quality of the diamond coatings *q*, we used the relation of the area below the diamond peak I (with a factor of 75) to the total area of the peaks, less underground [\[4\]:](#page--1-0)

$$
q = \frac{75I_{\rm d}}{75I_{\rm d} + \sum_{\rm nd} I_{\rm nd}} 100\% \tag{2}
$$

The index nd marks the non-diamond phase and the factor 75 takes the more effective Raman scattering on sp^2 -structures for the wavelength of the used laser into account [\[13,14\]. T](#page--1-0)he essential coating stress can be determined by using the shift of the diamond peak in the Raman spectrum. The Raman shift is proportional to biaxial stresses in the diamond films which can be

Fig. 1. The industrial hot filament reactor (CemeCon CC800Dia) used for this work and a schematic drawing of the used set-up. The filaments were arranged in rows and two substrates were placed in front of each filament, respectively.

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