Oxidized diamond-supported catalyst for simultaneous synthesis of nanocarbons and hydrogen

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High yields of hydrogen and nanocarbons could be obtained by the decomposition of methane using oxidized diamond-supported Ni and Pd catalysts at 873 K. This is the report to show that oxidized diamond can serve as an active catalytic support material. The simultaneous synthesis of nanocarbons and hydrogen is especially interesting. Oxidized diamond is proposed as a novel catalytic means for the synthesis of hydrogen and nanocarbons, and it is suggested that oxidized diamond may catalyze unique reactions of importance in nanomaterial synthesis.

The production and utilization of nano-carbon materials such as fullerene and carbon nanotubes have attracted much recent attention. Diamond is also one of the most important, interesting, unique and stable forms of carbonaceous materials.

Recently, fuel cell expected as one solution in the development of zero-emission moving vehicles. H_2 is a very attractive clean fuel for reducing atmospheric pollution. However, significant problems are involved in the economic production in use of large quantity, without co-producing CO_2. If H_2 could be efficiently produced, it could provide clean universal fuel. CH_4 is an attractive source of H_2 production. Decomposition of CH_4 is an effective H_2 production process, and can feed a fuel cell. H_2 production process without CO are required, because CO strongly poisons the electrode of fuel cell.

We focused on the oxidized diamond which has the weak interaction with loaded active metal species. We have found that Ni and Pd-loaded oxidized diamond catalysts markedly promoted the decomposition of CH4 and simultaneously produced novel carbon composite material made of a combination of diamond (sp^3 carbon) and nanocarbons (sp^2 carbon).

Table 1 shows effect of temperature on the H_2 and carbon yield over the oxidized diamond-supported Ni (5 wt%) catalyst. H_2 and carbon yield increased with increasing a reaction temperature, and the highest H_2 and carbon yields were obtained at 873 K. To produce H_2, metallic Ni is required. In the case of the oxidized diamond supported system, the surface oxygen species of oxidized diamond surface may retain Ni on the oxidized diamond active. Such a synergistic promotion effect on the active phase formation is believed to be the reason for the observed apparent support effect. We may consider that the high level of activity of the oxidized diamond-supported catalyst originated from the chemical interaction between the loaded Ni and the oxidized diamond support surface.

Table 1. Effect of temperature of Ni(5wt%)/Oxidized diamond catalyst on the decomposition of CH_4 at 873 K

Temperature	Reacted	Yield	
	CH_4	H_2	Carbon
(K)	(mmol)	(mmol)	
723	1.3	2.3	1.2
773	1.9	3.4	2.1
823	4.0	6.9	3.8
873	5.7	11.3	5.4

Catalyst, 0.06 g; Flow rate of CH_4, 10 mL/min (24.8 mmol/h); reaction time = 1 h, Space velocity = 10000 h-1mL/g-cat.

Figures 1a and b show scanning electron microscope (SEM) images of nanocarbons over oxidized diamond-supported Ni and Pd catalysts, respectively. The nanocarbons were prepared by the decomposition of methane at 873 K. Both nanocarbons over oxidized diamond-supported Ni and Pd catalysts showed zigzag structures, however, diameters and lengths of nanocarbons were respectively different from each other. Although it is premature to discuss the role of metal particles or species, these observations might suggest that the formation mechanisms and characteristics of nanocarbons which were formed over oxidized diamond supported Ni and Pd strongly depended upon the metal species employed.

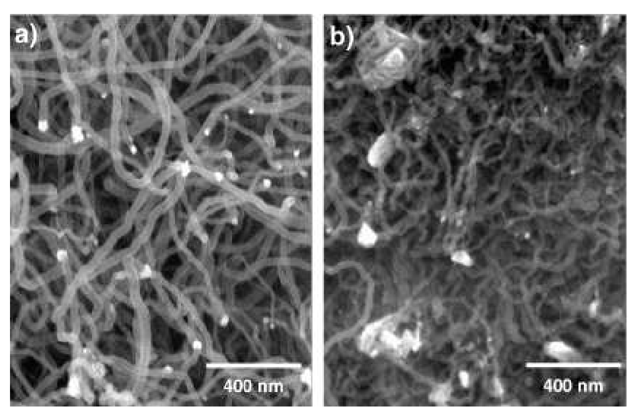

Figure 1. SEM images of carbon nanomaterials obtained by decomposition of CH_4
a) Ni/Oxidized diamond, b) Pd/Oxidized diamond

The novel composite carbon material of diamond (sp^3 carbon) and carbon nanotube or filament (sp^2 carbon) can be regarded as a unique material which expresses new physical properties such as high thermal and electrical conductivities for use in nanometer-sized semiconductor devices and interconnects, and the electrode for the fuel cell.

Acknowledgments

Some parts of this work were financially supported by Foundation for Promotion of Material Science and Technology of Japan (MST Foundation), Japan Science and Technology Corporation (JST), Toppan Printing Co. Ltd., and Novel Carbon Project (NIIMS #166509).