

# DIAMOND SYNTHESIS BY THE MICROWAVE PLASMA CVD METHOD USING A MIXTURE OF CARBON MONOXIDE AND HYDROGEN GAS

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We have succeeded in diamond synthesis from carbon monoxide (CO) and hydrogen (H<sub>2</sub>) mixed gas, which has been thought to be difficult to achieve, by a microwave plasma chemical vapor deposition (CVD) method. The growth rate when using CO as the source gas is several times faster than that when using CH<sub>4</sub>. Also, it is found that diamond could be synthesized up to a CO concentration of 80 vol%.

## 1. Introduction

Diamond is synthesized conventionally by plasma-enhanced chemical vapor deposition, using hydrocarbon and hydrogen mixed gas<sup>1</sup>, a mixture of oxygen-containing organic compounds such as alcohols or acetones and hydrogen gas, or a mixture of nitrogen-containing organic compounds such as amines and hydrogen gas<sup>2</sup>. It is presumed that these reactions proceed via methyl radicals<sup>3,4</sup>.

We have attempted diamond synthesis using CO as the source gas, which is generally thought to be

difficult to use for diamond synthesis, and were successful. It is assumed from these findings that this reaction may proceed by a mechanism not explained by the conventional methyl radical theory alone.

## 2. Experimental

The vapor deposition method assisted by microwave-induced plasma was employed to prepare diamond. Figure 1 shows a schematic drawing of our diamond synthesis apparatus<sup>5</sup>. The apparatus

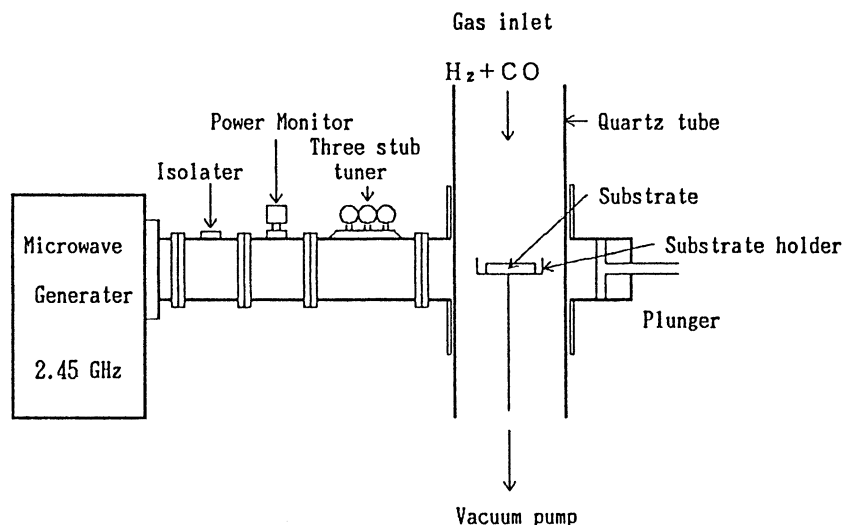


Fig. 1. Schematic drawing of microwave plasma enhanced chemical vapor deposition system.

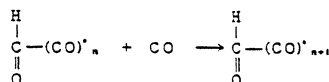
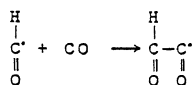
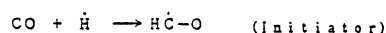


proach to resolve the formation mechanism of diamond caused by microwave-induced plasma. We assumed that the formation mechanisms of diamond were divided into two steps: an initiation process for forming reactive species and a propagation process for forming diamond. We tried to analyze the initiation process by using *ab initio* methods and to analyze the propagation process by using the Hückel method. From these analyses, it was estimated that the cluster played an important role in diamond formation. This scheme showed the reaction between CO and H<sub>2</sub> (Table 2).

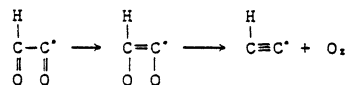
Table 2. Reaction scheme of diamond synthesis from H<sub>2</sub>-CO system

(1) CO + H<sub>2</sub> system (reduction mechanism)

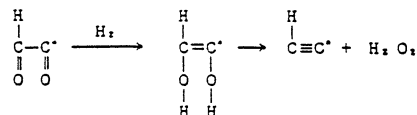
① growth scheme of cluster



② de-oxidation scheme (I)



③ de-oxidation scheme (II)



We now consider a reductive reaction to be important for diamond synthesis using a CO and H<sub>2</sub> mixed gas. In addition, we would like to propose the cluster-growing model, in which CO molecules are successively linked to a formyl radical as an initiator, followed by a deoxidation reaction in the propagation process. In plasma CVD using CH<sub>4</sub> and H<sub>2</sub> mixed gas, a hydrogen extraction reaction is an important process for diamond formation; i.e., an oxidation reaction. Also, both formation mechanisms of diamond are supposed to be radical reactions rather than ionic reactions.

#### 4. Conclusions

(a) Diamond can be synthesized from CO and H<sub>2</sub> mixed gas by microwave plasma enhanced chemical vapor deposition.

(b) The growth rate when using CO and H<sub>2</sub> mixed gas is several times faster than that when using CH<sub>4</sub> and H<sub>2</sub> mixed gas.

(c) Diamonds can be synthesized up to a CO concentration of 80 vol%.

(d) Diamond is formed by a reductive mechanism when using CO as the source gas. If CH<sub>4</sub> is used as the source gas, the process should be an oxidation mechanism.

#### REFERENCES

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