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The Dissociation of Carbon Monoxide in an Induction-coupled Argon Plasma Jet

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Synopsis. The major products were solid carbon, CO_2 and O_2. As the inside diameter of the probe decreased, the concentration of O_2 in the quenched gas increased and that of CO_2 decreased. The product distribution was independent of the argon-flow rate.

For many high-temperature chemical reactions, the distribution of the final products is determined by the quenching step. One of the techniques used to quench reacting high-temperature gas streams is placing them in contact with a cold wall[1–5] by, for example, passing them through a small-diameter, water-cooled tube. Baddour and Iwasyk have studied the reactions of elemental carbon with hydrogen above 2800 K, using this quenching method.[1] A similar method of quenching has been also applied to the synthesis of nitrogen fluorides[2] and of hydrogen cyanide.[3]

The present work, using small silica tube as a quenching apparatus, was undertaken in order to examine the effects of the operating conditions on the quenching conditions and on the distribution of the final products, when carbon monoxide was decomposed in the induction-coupled argon plasma jet at 1 atm.

Experimental

Materials. The argon used in this work was a high-purity cylinder gas (above 99.99% purity), while the carbon monoxide was commercially obtained from the Seitetsu Kagaku Co. (above 99.0% purity).

Apparatus and Procedures. The apparatus and procedures were similar to those used in previous papers.[6,7] Carbon monoxide was charged into the plasma jet counter-currently. The quenching of the decomposed products was done by means of small water-cooled silica tubes (which we shall hereafter name "probes"). The distance between the probe inlet and the induction coil was 0.5 cm. The mean temperature of the gas at the probe inlet was estimated by a method previously reported.[8] In this estimation, the hot gas was assumed to be composed of argon only, since it was difficult to ascertain precisely the composition of the hot gas at the probe inlet and since argon accounted for more than 93% of the hot gas under the conditions used in this work.

Results and Discussion

Table 1 shows the effects of the inside diameters of probes on the decomposition results and the mean temperatures of gas at the probe inlet. The major products are solid carbon, molecular oxygen, and carbon dioxide. Most of the carbonaceous species formed by the dissociation of carbon monoxide are transformed to solid carbon in the quenching step. As is clear from Table 1, the mean temperature of the gas at the probe inlet reaches 7500—8100 K; it is within the limits of experimental error independent of the inside diameter of the probe with a given outside diameter. Thermodynamic calculations of the carbon-oxygen system with a composition of $C/O=1.0$ at 1 atm have suggested that the partial pressures of molecular oxygen and carbon dioxide are less than 10^{-4} atm above 2500 K.[9] Therefore, it is certain that molecular oxygen and carbon dioxide are formed mainly in the quenching process. It is considered that the cooling rate in the probe with an inside diameter of 1 mm is greater than those in the other two probes.[4,8] Table 1 shows that, as the cooling rate increases, the amount of carbon dioxide formed decreases and that of molecular oxygen increases. Table 1 also shows that the conversion of carbon monoxide decreases with a lowering of cooling rate; this result suggests that the reproduction of carbon monoxide occurs in the quenching process.

Figure 1 shows the effects of the argon-flow rates on the conversions of carbon monoxide and the mean temperatures of gas at the probe inlet. Figures 2 and 3 indicate the variations in the quenched-gas compositions with the argon-flow rates. At a given power

TABLE 1. EFFECT OF PROBE INSIDE DIAMETERS ON DISSOCIATION OF CO
Ar: 1.07±0.01/min, CO: 0.044±0.003 l/min, Power input: 7.60±0.02 kW
Outside diameter of probe: 15 mm

Inside diameter (mm)	Conversion (%)[a]	Solid carbon (%)[a]	Compn. of quenched gas (vol%)			Temp.[b,c] (K)
			CO	O_2	CO_2	
1	37.2±2.8	34.9±2.8	76.9±2.2	20.3±2.3	2.8±0.5	8100
3	31.5±2.1	26.8±2.7	80.8±2.0	12.9±1.9	6.3±0.4	8000
5	32.9±2.1	28.1±2.2	80.3±1.5	14.0±1.4	5.7±0.2	7500

a) Carbon base.
b) The mean temperature of gas at the probe inlet which was estimated from the heat balance based on the temperature of the probe exit gas and the temperature rise of water which cooled the probe.[8]
c) Maximum experimental error: ±700 K.

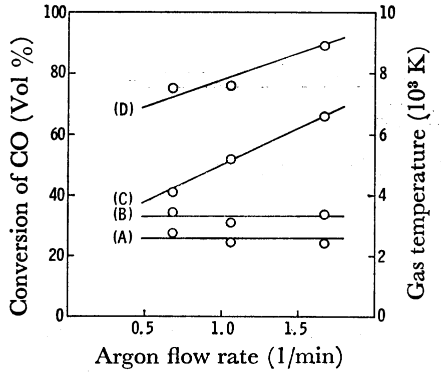

Fig. 1. Effect of argon flow rate on conversion of CO and mean temperature of gas at the probe inlet (I.D. of probe: 3 mm, O.D. of probe: 15 mm, CO: $0.044+0.003$ l/min).

(A) and (B): Conversion of CO at 4.62 ± 0.02 kW and 8.65 ± 0.02 kW, respectively.

(C) and (D): Mean temperature of gas at the probe inlet at 4.62 ± 0.02 kW and 8.65 ± 0.02 kW, respectively.

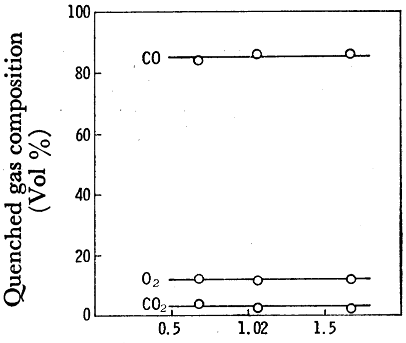

Fig. 2. Variation of quenched gas composition with argon flow rate at 4.62 ± 0.02 kW (other conditions are the same as that of Fig. 1).

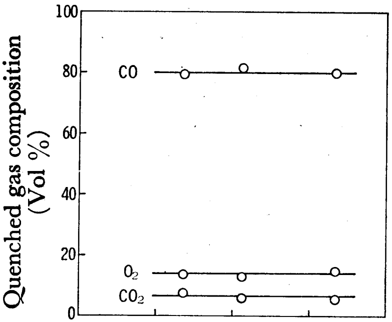

Fig. 3. Variation of quenched gas composition with argon flow rate at 8.65 ± 0.02 kW (other conditions are the same as that of Fig. 1).

input, the mean temperature of the gas at the probe inlet becomes higher as the argon-flow rate increases, but the conversion of carbon monoxide and the composition of the quenched gas are almost independent of the argon-flow rate. It has been reported by Sundstrom and DeMichiell that the rate of quenching is greater at a higher mass flow rate in the case of the same initial gas temperature.[4] However, when the initial gas temperature is low at a given mass flow rate, it has been assumed that the decomposed products are cooled to the quenching temperature in a time shorter than when the initial gas temperature is high.[4,8] That is, when the argon-flow rate is increased, the increase in the rate of quenching is cancelled out by the rise of the initial gas temperature; apparently, then, the quenching temperature does not depend on the argon-flow rate. Therefore, it may be considered that the conversion of carbon monoxide and the composition of the quenched gas are almost independent of the argon-flow rate.

At a given argon-flow rate, as is clear from Figs. 2 and 3, both the conversion of carbon monoxide and the concentration of carbon dioxide in the quenched gas increase slightly with an increase in the power input. The mean temperature of gas at the probe inlet at 8.65 kW is higher than that at 4.62 kW. Thus, the situation in the quenching step is similar to that described above. However, one may say that, at a given argon flow rate, the dissociation of carbon monoxide at a high power input gives a quenched gas in which the concentration of carbon dioxide is higher than does the dissociation of carbon monoxide at a low power input.

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