GENERATION OF MICROWAVE COLD PLASMA AT ATMOSPHERIC PRESSURE

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Abstract

A steady state atmospheric cold plasma discharge could be generated when the microwave output power of several ten watts was induced to the perovskite type oxide powders such as $La_{0.8}Sr_{0.2}CoO_3$ in a stream of argon. Apparent value of the plasma temperature was approximately 120-250℃. A high decomposition rate of nitrogen monoxide was obtained under a condition of considerable high feed rate.

1. Introduction

Microwaves are closely linked with today's information society and our daily life: various communication equipment and radar utilize their property of rectilinear propagation, while they are used as a heat source in domestic microwave ovens. In addition, microwaves and high-frequency waves, which induced cold plasmas in the vacuum chambers, have played an important role in the manufacture of various electronic components such as semiconductors.

In industrial plasma processing, a steady state atmospheric glow discharge would allow many surface modification and other plasma processing operations to be carried out under atmospheric conditions, rather than in expensive vacuum systems which enforce batch processing. The atmospheric glow plasma process was recently developed, and many application experiments such as the surface treatment of organic polymer sheets carried out by using that process[1-3].

By the way, we found out that an atmospheric argon cold plasma discharge generates on the perovskite type oxide powders such as $La_{0.8}Sr_{0.2}CoO_3$ with an irradiation of microwave to the sample[4]. In this symposium, we report on the results of the material research relating to a steady state atmospheric cold plasma genefation and the examining of nitrogen monoxide(NO) decomposition in the plasma.

2. Experimental

Figure 1 illustrates a schematic drawing of the microwave plasma generator. The microwave generator consisted of a microwave-generating unit(MR-500, Ewig), an isolator, a power monitor, a three-stub tuner and an applicator. The microwave frequency was 2.45 GHz and the output power could be varied within the range of 0-500 W. The power monitor was provided with output meters for incident and reflected waves. The difference between the readings of these meters was considered to be the microwave output power at the time of plasma generation.

In the applicator, which is the plasma generation unit, a quartz tube (20 mm O.D.) was used as the generator. One gram of metal oxide powder sample was placed in the quartz tube generator and then argon gas was introduced to the generator. The microwave output power of several ten watts was supplied from magnetron through a wave guide. The impedance was adjusted at this time in order to increase the efficiency of plasma generation. The three-stub tuner was adjusted to minimize the power of the deflected wave, and concurrently the surface of the oxide sample attained the plasma state.

The compounds $Ln_{1-x}Sr_xCoO_3$ were synthesized by ordinary ceramic techniques. As starting materials, metal acetates were used. The appropriate amounts of the reagents were weighed and then dissolved in pure water to give a solution was evaporated in a rotating flask between at 70℃ under an aspirator vacuum (ca. 20 mmHg). The residue powder was a fine mixture of the component acetates. The powder placed in a quartz boat was decomposed at temperatures at 300℃. The decomposition processes took place after melting of the mixture. The decomposed and oxidized sample had a caramel-like surface that was black or dark brown in color. The sample was pelletized into 250-840 μ m diameter and then heated at 850℃ for 5 hours.

The activity measurements for NO decomposition were performed in a conventional flow reactor system. The NO conversion was determined by gas chromatography.

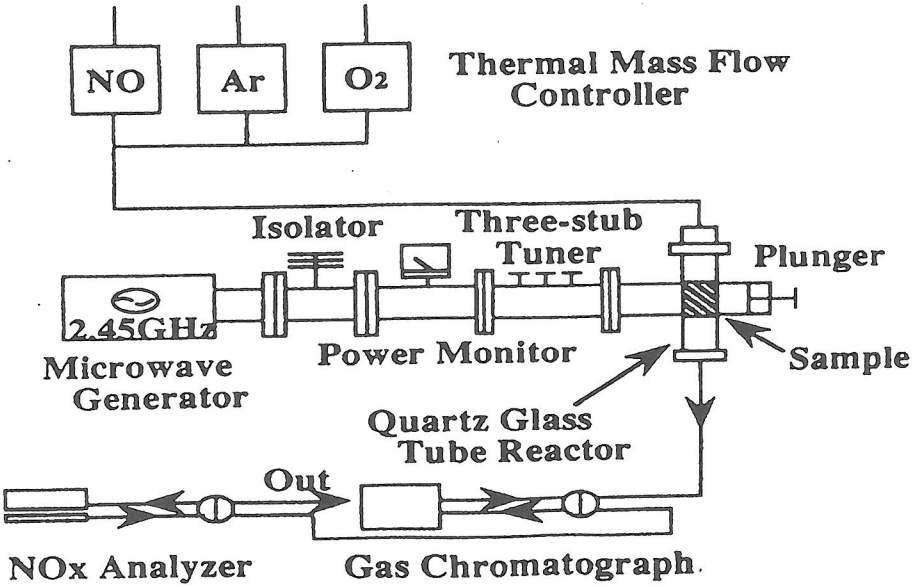

Fig.1 Apparatus

3. Results and Discussion

A pale violet color of stable plasma discharge was seen around the perovskite oxide powder of $La_{0.8}Sr_{0.2}CoO_3$ sample when microwave output power of 30W was induced to the sample in a stream of argon. It was difficult to directly measure the plasma gas temperature and so the temperature of outside wall of the quartz-glass generator was measured by the infrared irradiation thermometer. The apparent temperature was 120℃.

Figure 2 shows the emission spectrum of the plasma discharge. Several peaks appeared in the range of 700-850 nm of emission fields, especially the emission peak of 811 nm was high. These all peaks could be assigned to 4s-4p transition state of argon atom[5].

The materials having a generative ability of the atmospheric cold plasma discharge were researched from many oxides, nitrides and carbides. Figure 3 illustrates the results of the research. The most effective material for the plasma generation at atmospheric pressure was the perovskite type oxide of $La_{0.8}Sr_{0.2}CoO_3$. This oxide is well known as an excellent catalyst of oxidation and NO decomposition at a high temperature[6].

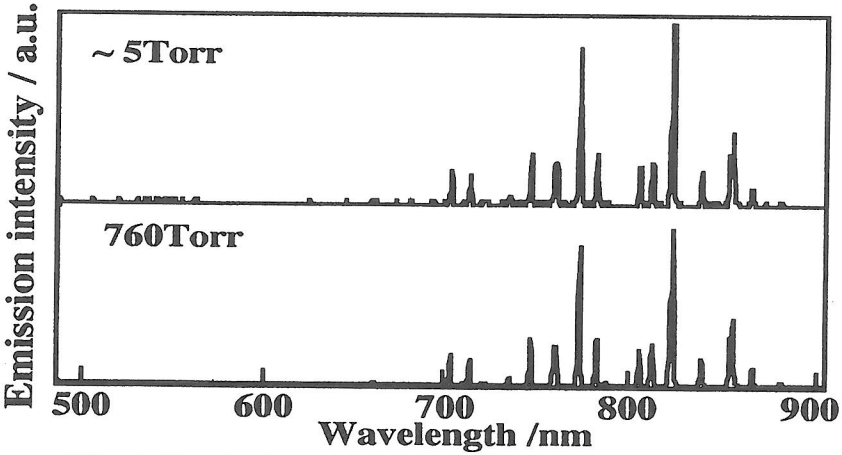

Fig.2 Emission spectrum of activated argon

Active samples

$La_{1-x}Sr_xCoO_3$ (X=0~0.8)	$Nb_{1-x}Sr_xCoO_3$ (X=0~0.2)
$LaNiO_3$	$LaMnO_3$
$YBa_2Cu_3O_7$	Fe_3O_4
active carbon	graphite

Inactive samples

γ - Fe_2O_3	α - Fe_2O_3	SiO_2	Al_2O_3
MgO	TiO_2	CaO	SnO_2
ZnO	ZrO_2	Nb_2O_5	MnO_2
Mn_3O_4	Co_3O_4	CuO	NiO
MoO_3	PbO	WO_3	
TiN	AlN	SiC	
$BaTiO_3$	$SrTiO_3$	$NaNbO_3$	$SrCoO_3$

Fig.3 Active and inactive materials

The decomposition of NO was examined in the plasma generator by supplying a mixed gas of 2000 ppm NO/argon at a rate of 600 $ml \cdot min^{-1}$ to the powder sample of 1g. As shown in Figure 4, a high conversion value of 63% from NO to N_2 and O_2 was obtained at the microwave output power of 30 W. While in the case of the thermal catalytic decomposition of NO, the oxide sample of $La_{0.8}Sr_{0.2}CoO_3$ gave only 3% of NO conversion at 800℃. Figure 5 shows the relation between an NO conversion and gas feed rate. A high decomposition rate of NO was obtained even at a considerable high feed rate such as 1200 $ml \cdot min^{-1}$. It is known that an atomic nitrogen accelerates the decomposition of NO rather than the oxidation of NO at low temperature[7], and hence it seems that a high conversion value of NO decomposition obtained in this experiment is attributable to the radical decomposition of NO in the atmospheric cold plasma.

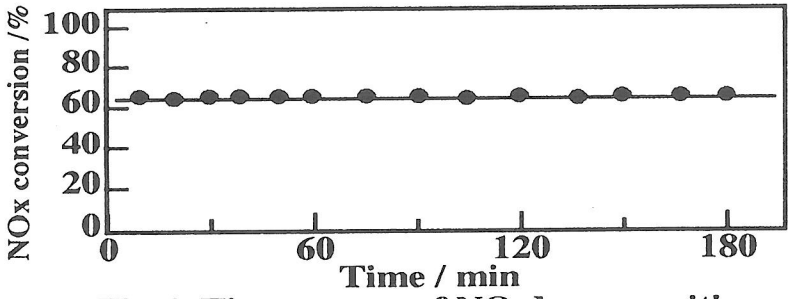

Fig.4 Time course of NO decomposition

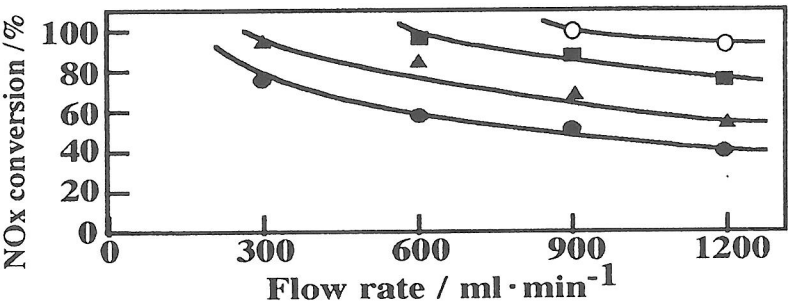

Fig.5 Effect of flow rate on NO decomposition
Output power : ● : 30W, ▲ : 60W
: ■ : 90W, ○ : 120W

4. Conclusion

An atmospheric cold plasma process was developed. A microwave irradiation to the perovskite type oxide powders in an atmosphere of argon resulted in a cold plasma generation. This plasma process was effectively to the high-speed decomposition of NO to N_2 and O_2.

References

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