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Investigation on Interaction between Cold Plasma with Catalysts

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Abstract: The characteristic parameters were measured with floating double probe method when cold plasma was interacting with catalysts, such as MoO_3/Al_2O_3, NiY, Pd/Al_2O_3, which were used in the conversion of natural gas to C_2 hydrocarbons through electrical field enhanced plasma catalysis. These parameters were compared in different input voltage, different atmosphere, before and after reaction in plasma field. The interaction between catalysts and cold plasma was also investigated. This confirm that cold plasma can enhanced catalysis effect.

Keywords: Cold plasma, floating double probe, catalyst.

Plasma chemical processing is a promising route for synthesis of chemicals that have high activation energies, because very high energy can supply to the plasma and many kinds of reactive particles, electrons, free radicals, ions metastable species and photons. The combination of free radicals, neutral particles, ions and electrons offer a rich 'soup' that provide opportunities for innovation and manufacturing beyond the traditional methods[1]. The reactor chamber is a quartz tube with an inside diameter of 18 mm. Using two position circles with two grounded sealers the reactor can be disassembled and two electrodes were kept in the right positions respect to the reactor and a small quartz boat can be put in or taken out. There are two mouthes on the reactor, one is connected with vacuum system and another is connected with argon or methane line. The discharge is created by an AC high voltage power. The power is connected with electrodes through a resistance in order to measure the current across the circuit. A Tektronix TDS 210 digital real-time oscilloscope with a Tektronix P6015A high voltage probe monitors is used to measure the discharge voltages.

The comparative methods are used for investigation of the interaction between plasma and catalysts. First, a small quartz empty boat in plasma zone is measured with the floating double probe in argon or methane, respectively. Then 1 g catalyst power (40~60 mesh) was loaded in the small quartz boat in position as mentioned above in argon or methane, again. The measurements are carried out. The conditions of all experiments mentioned above, including the position of electrodes and the small boat, are held in the same place respect to the reactor.

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Figure 1 Electron temperature *versus* time

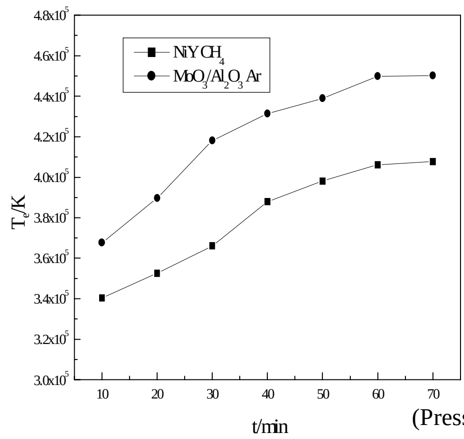
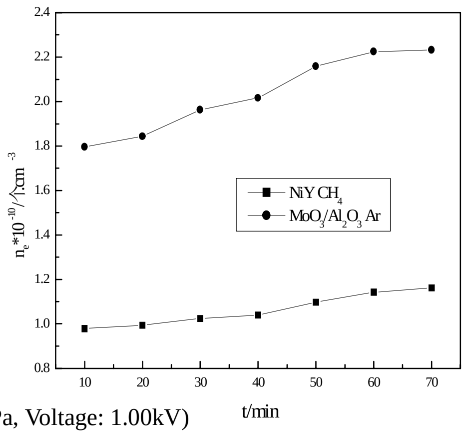


Figure 2 Electron density *versus* time



(Pressure: 30Pa, Voltage: 1.00kV)

Figure 1 shows that the electron temperature over NiY catalysts in methane and over MoO_3/Al_2O_3 catalysts in argon increases with prolonging the cold plasma treatment time. **Figure 2** indicates that the electron density also increases with prolonging the cold plasma treatment time.

Table 1 Comparison of T_e and n_e over Pd/Al_2O_3 catalysts before and after reaction

		before reaction			after reaction		
		0.73kV	1.00kV	1.03kV	0.73kV	1.00kV	1.03kV
Ar	$T_e \times 10^5$/K	3.688	3.932	4.681	3.591	3.760	4.477
	$n_e \times 10^{10}$/个.cm^{-3}	1.514	2.492	3.210	1.422	2.186	2.951
CH_4	$T_e \times 10^5$/K	3.732	3.929	4.087	3.595	3.741	3.968
	$n_e \times 10^{10}$/个.cm^{-3}	1.112	1.808	2.118	1.006	1.528	1.973

Table 1 shows that the electron temperature and the electron density over Pd/Al_2O_3 catalysts increase with increasing the discharge voltage in methane and in argon. This is consistent with what the conversion of methane increase when discharge voltage is increasing[2, 3]. The electron temperature and the electron density are higher before reaction than those after reaction both in methane and in argon because part of surface is covered with carbon deposit in the reaction. The ionization energy of methane is more than the dissociate energy of methane into CH_3*and H*[4], therefore, the electron density is less in methane than that in argon.

Most electrical energy in gas discharge is put into the production of energetic electrons, rather than into gas heating. When the gas discharge is introduced into the catalyst layer, it changes the electronic state of the gas species. The charge on catalyst surface, together with other effects of excited species in the gas discharge lead to variations of the potential and work function of catalyst surface. The sorption and desorption performances of the catalyst can be modified. The cold plasma was perturbed with catalyst, thus, the catalytic action can be improved in the chemical reactivity of the system.

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References

1. M. Kenward, *Physics word*, **1995**,31.
2. C. J. Liu, A. Marafee, B. Hill, *et al. Ind. Eng. Chem. Res.*, **1996,***35* (10), 3295.
3. B. W. Wang, G.H Xu, *Science in China*, **2002**, *45* (3), 299.
4. A. A Radzig, B. M Smirnov. *Reference data on atoms, molecules, and ions*, Berlin, Springer-Verlag Berlin Heidelberg, **1985**,pp321-327.

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