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Processing of hydrocarbons in an AC discharge nonthermal plasma reactor: An approach to generate reducing agents for on-board automotive exhaust gas cleaning

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Abstract

Light hydrocarbons and H_2 can be used to enhance NO_x reduction efficiency and regenerate sulfur-poisoned NO_x storage catalysts, and therefore are valuable for automotive exhaust gas cleaning. The processing of hydrocarbons in an alternating current (AC) discharge nonthermal plasma reactor was studied for the instant generation of light hydrocarbons and H_2 at room temperature and atmospheric pressure. *n*-Octane and *n*-hexane were used as model hydrocarbons. Effects of hydrocarbon feedstock, electrode diameter, applied voltage, flow rate of carrier gas, gap size, and residence time of hydrocarbon molecules, were investigated systematically. Cracking is the only detected reaction during *n*-octane conversion (which might be very attractive for the cracking of heavy oil), and is the dominant reaction during *n*-hexane conversion. Catalytic dehydrogenation, catalytic addition, and noncatalytic cracking reactions, were discussed. The cleavage mode of single carbon–carbon bonds is revealed to be relevant to the carbon number of hydrocarbon molecules. Conversions, yields, power consumption, energy efficiencies, generation of hydrogen, etc, were determined and discussed. This study is of importance to novel processing of hydrocarbons at room temperature and atmospheric pressure, instant generation of hydrogen, cleaning of automotive exhaust gas, and chemistry in nonthermal plasma reactors.
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1. Introduction

NO_x storage-reduction (NSR) catalysis technology was developed for the emission control of lean-burn gasoline and diesel engines, due to the difficulties for conventional three-way catalysts to remove NO_x from oxygen-rich exhaust gases [1,2]. The regenerating rich pulse is typically accomplished by engine control or by directly injecting diesel fuel into the exhaust stream. However, the long chain hydrocarbon molecules found in automotive fuel typically have very low reactivity. Much higher NO_x reduction efficiency could be achieved if the hydrocarbon fuel could first be converted on-board to lighter hydrocarbons such as ethylene (C_2H_4), propylene (C_3H_6), or H_2. In addition, these reducing agents, light hydrocarbons and H_2, are also more reactive for the regeneration of sulfur poisoned NO_x storage catalysts [1,3,4].

Conventional hydrocarbon cracking technologies [5] are not suitable for on-board automotive systems because of the need for catalyst warm-up time, a hydrogen source, or coke removal, etc. A possible alternative strategy is to use nonthermal plasma technology which has received considerable attention in the fields of chemistry and material processing [6]. In addition, catalysis technology has been successfully com-

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bined with nonthermal plasma technology in various applications [7–13].

We reported the preliminary research on conversion of *n*-hexane over different electrode compositions in a PACT (plasma and catalysis integrated technologies) reactor, a type of nonthermal plasma reactor [7]. Herein we report studies on the processing of hydrocarbons in an alternating current (AC) discharge PACT nonthermal plasma reactor, where hydrogen or/and light alkanes/alkenes were produced instantly at room temperature and atmospheric pressure. *n*-Octane (n-C8) and *n*-hexane (n-C6) were used as model hydrocarbons. Effects of hydrocarbon feedstock, electrode diameter, applied voltage, flow rate of carrier gas, gap size, and residence time of hydrocarbon molecules, were studied systematically.

2. Experimental

2.1. PACT tubular reactor setup

The reactions were carried out in a tubular PACT reactor, as depicted in Fig. 1. The reactor consists of an exchangeable, cylindrical, nonporous inner metal electrode of 10 cm length and 8.0 (or 9.5) mm diameter, screwed onto a supporting metal rod, and a quartz tube (17.9 cm long, 9.85 mm i.d., and 12.55 mm o.d.) acting as dielectric separating outer and inner electrodes. Aluminum foil (10 cm long) is wrapped around this quartz tube and acts as the outer electrode. Nickel was electroplated onto copper rods at a thickness of 100 μm and these

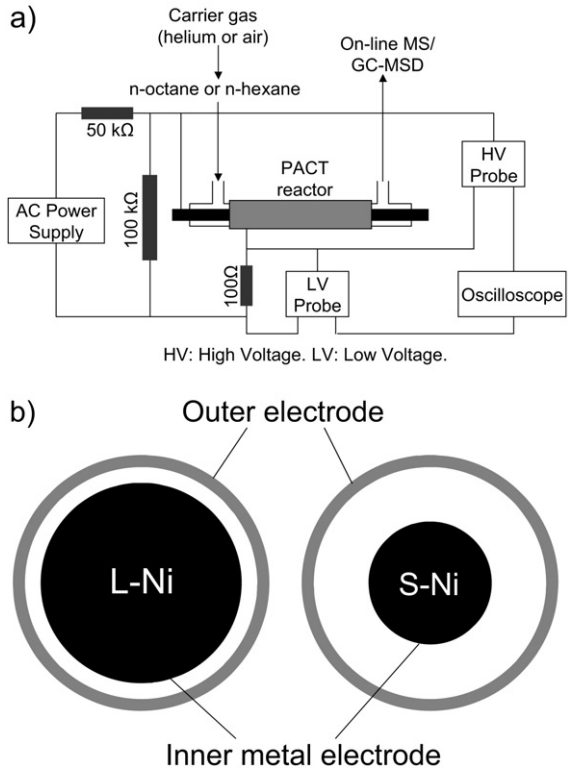

Fig. 1. Diagram of the PACT reactor system. (a) The circuit setup and PACT reactor setup. (b) Cross section of the PACT reactors. L-Ni: large-diameter nickel electrode. S-Ni: small-diameter nickel electrode.

Table 1
Plasma zone data for electrodes with different diameters

Inner electrode	Diam.[a] (cm)	Length[b] (cm)	SA[c] (cm^2)	Gap size[d] (mm)	Plasma zone vol.[e] (cm^3)
L-Ni[f]	0.950	10.000	29.845	0.175	0.532
S-Ni[g]	0.800	10.000	25.130	0.925	2.594

[a] Diameter of inner electrode.
[b] Length of inner electrode.
[c] Side surface area in plasma zone.
[d] The distance between the inner electrode and the tubular quartz barrier. The diameter of inner wall of quartz tube barrier is constantly 9.85 mm.
[e] The volume of the cavity between the inner electrode and the tubular quartz barrier, where a nonthermal plasma be generated by AC discharge and exists.
[f] Large-diameter nickel electrode.
[g] Small-diameter nickel electrode.

products were used as electrodes. The metal electrodes were obtained from Mitsubishi Materials Corporation. Table 1 shows detailed data on the types of the electrodes used in this research and the sizes of plasma zones.

2.2. Circuit setup

The circuit setup is also shown in Fig. 1. The high-voltage supply was 120 V AC (60 Hz). The high voltage was generated by an UHV-10 AC high-voltage power supply. A digital DL-1540 Yokogawa oscilloscope with a high-voltage probe (Tektronix P6025) and a low-voltage probe (Yokogawa 70996) was used to measure input voltage and input current of the PACT reactor. The voltage across a 100 Ω standard resistor in series with the reactor was used to determine the input current.

2.3. Experimental parameters and product analysis

In a typical experiment, a carrier gas (helium) was passed through a bubbler containing about 500 ml *n*-hexane liquid, at a flow rate of 44 ml/min, and the gaseous mixture was introduced into the PACT reactor at room temperature and atmospheric pressure. *n*-Hexane and *n*-octane feeds were purchased from J.T. BAKER and ALFA AESAR, respectively. *n*-Hexane feed contained 95% *n*-hexane and 5% hexanes including methyl cyclopentane and 3-methyl pentane. The reactor's inner metal electrode was connected to the high-voltage line, while the outer aluminum/quartz electrode was connected to the ground line.

During the experiments, the PACT reactor was not heated, and no significant temperature change was observed. For the purpose of simplification, the reactor was assumed to be an ideal plug flow reactor. The reaction system was vent to atmosphere, which maintained a constant reactor pressure at atmospheric pressure. The set carrier gas flow rate under these experimental conditions was 15, 44, 87, 145, or 218 ml/min. Due to the use of the dial of the UHV-10 HV supply and the consistency of tests, the set value of applied voltage under these experimental conditions was 5.4, 6.8, 8.2, 9.5, 10.6, or 12.0 kV. Unless specified otherwise, the reactor was run for 20 min to allow the system to come to equilibrium before a sample of the

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