Coal Liquefaction Mechanism Using a Tritium Tracer Method

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To determine the behavior of hydrogen in tetralin, the reaction of tetralin with tritiated gaseous hydrogen was studied in a flow reactor at 400–450°C, 2.5–9.8 MPa for various residence times. The amount of hydrogen exchange between tetralin and tritiated hydrogen was estimated from the balance of hydrogen and tritium. Although yields of methylindan and naphthalene, and the hydrogen-exchange ratio (HER) of tetralin increased monotonously with residence time, these values were scarcely influenced by the reaction pressure at every temperature. It was thought that the formation of tetralyl radicals in this system would be the rate-determining step for both the conversions of tetralin into methylindan and naphthalene, and the hydrogen exchange of tetralin. Conversions of tetralin into methylindan and naphthalene, and the hydrogen-exchange reaction using the autoclave were very close to those using the flow reactor.

Introduction

Since liquefaction includes hydrogenation and hydrocracking of coal with hydrogen in the gas phase and solvent, it is important to elucidate the hydrogen-transfer mechanism. A number of attempts have been made to accomplish this (Billmers et al., 1986; Malhotra and McMillen, 1990, 1993; Oviawe et al., 1995; Autrey, 1995). We have already reported that tritium and carbon-14 tracer techniques were effective in tracing quantitatively the behavior of hydrogen during coal liquefaction (Kabe et al., 1986, 1987a,b, 1989, 1990a,b, 1991a,b; Ishihara et al., 1993, 1995; Godo et al., 1997a,b). These works show that the quantitative analysis of hydrogen mobility of coal and coal-related compounds could be given through the hydrogen-exchange reactions among coal, gas phase, and solvent as well as the hydrogen addition.

In previous works, we assumed that the hydrogen addition and exchange reactions between tetralin and gaseous hydrogen, and the formations of naphthalene and 1-methylindan from tetralin, proceeded with a radical reaction mechanism (Kabe et al., 1990b; Ishihara et al., 1995; Godo et al., 1997a), where a tetralyl radical was an intermediate in the hydrogen exchange and the conversion of tetralin. Therefore, the formation of the tetralyl radical in this system was assumed to be the rate-determining step for both the hydrogen exchange and the conversion of tetralin.

In the system of tetralin and gaseous hydrogen, tetralin may collide with not only itself but also tritiated hydrogen. It is possible that tritiated hydrogen affects the formation step of a tetralyl radical. If tritiated hydrogen affects the formation of the tetralyl radical, the hydrogen exchange ratio (HER) and conversion of tetralin will change by the partial pressure of gaseous hydrogen. In our previous reports, we conducted batch reactions using an autoclave. However, it was difficult to control the reaction pressure in the reactions using the autoclave because the effective reaction pressure changes depending on reaction temperature. Further, an autoclave experiment takes longer than about 30 min to heat up the system to 400°C, which is the effective temperature for coal liquefaction. The hydrogen transfer and conversion of tetralin would occur while the autoclave is heated up to the set temperature. This may not be a major problem for comparative experiments, but would be unsatisfactory for kinetic studies, especially in the short reaction time. In contrast, a flow-type reactor can control the reaction pressure strictly, and is suitable for the short reaction time.

In this article, the hydrogen-exchange reactions between tetralin and gaseous hydrogen were investigated in a flow-type reactor to estimate the effect of reaction pressure and the kinetics in the short reaction time.

Experimental Studies

Materials

A commercial guaranteed reagent tetralin was used without further purification. Gaseous hydrogen was supplied by

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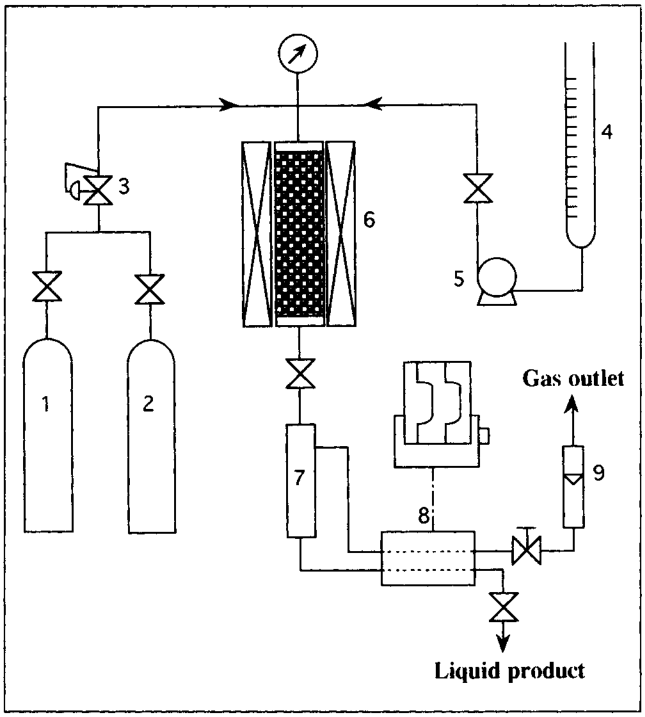


Figure 1. Experimental apparatus.

1. Hydrogen gas cylinder; 2. tritiated hydrogen-gas cylinder; 3. pressure regulator; 4. tetralin supply buret; 5. high-pressure micropump; 6. reactor; 7. gas-liquid separator; 8. radio analyzer; 9. flowmeter.

Tohei Chemical. Tritiated molecular hydrogen was obtained by the electrolysis of tritiated water using an HG-225 Hydrogen Generator supplied by Gasukuro Kogyo Inc.

Apparatus and procedure

Reaction with a Flow-Type reactor. As shown in Figure 1, the experiments were performed in a tubular flow reactor with a stainless-steel tube, 8 mm in dia. and 300 mm in length, packed with quartz sand. The reaction zone was heated by an external electrical heater whose temperature was controlled. The reactor was pressurized by tritiated hydrogen (radioactivity, about 3,000,000 dpm/mol). Then, tetralin was supplied by a high-pressure micropump (Kyowa Seimitsu KHD-16). The reaction was carried out under these conditions: temperature, 400–450°C; pressure, 2.5–9.8 MPa; flow rate of tritiated gaseous hydrogen: 20–80 mL/min; flow rate of tetralin, 0.1–0.4 g/min; H_2/tetralin, 1.2 mol/mol. In every reaction condition, the ratio of gaseous hydrogen and tetralin solvent was constant. Residence times were calculated by the volume of reaction zone and the flow rates of reactants. Conversion and hydrogen-exchange reaction reached a steady state within about 3 h. Then, samples of products, gas phase, and liquid phase were collected from a gas–liquid separator. Gas and liquid products were analyzed by gas chromatography with FID to get more precise compositions. For the measurement of radioactivities of the products, a Radio Analyzer (Aloka RLC-701) that was a flow detector of radioisotopes was used. The Radio Analyzer was able to measure the radioactivities of the gas phase and the liquid phase instantly. For the measurement of the radioactivity of the liquid products, liquid scintillation counting was also applied (Kobayashi, 1974; Horocks, 1974; Crook, 1978). Liquid products were directly dissolved into 14 mL of a scintillator reagent (Instaflour for an organic sample. Packard Japan), and the radioactivity of the obtained solution was measured with a Liquid Scintillation Counter (Aloka LSC-1050).

Calculation of Hydrogen Exchange Ratio. All tritium data appear in Tables 1–3. All the experimental points related to hydrogen exchange were obtained and calculated on the basis of such data. The HER described in this article means the ratio of hydrogen exchanged in tetralin to total hydrogen in tetralin. The HER between the solvent and gaseous hydrogen was calculated on the basis of Eq. 1. In Eq. 1, $H_{ex(G \Leftrightarrow S)}$ is the amount of hydrogen exchanged in tetralin in the reaction of tetralin with gaseous hydrogen. $H_{ex(G \Leftrightarrow S)}$ was calculated on the basis of Eq. 2. In these calculations, it was assumed that the hydrogen exchange between tetralin and gaseous hydrogen reached equilibrium at the end of reaction. The tritium concentration in exchanged hydrogen of tetralin was equal to that in gaseous hydrogen. When hydrogen addition from the gas phase to tetralin occurred, the amount of added hydrogen from the gas phase to solvent (H_{adt}), which was equal to the decrease in molecular hydrogen, was subtracted from $H_{ex(G \Leftrightarrow S)}$. The decrease in molecular hydrogen was determined by the change in the partial pressure. The partial pressure of gaseous products determined by gas chromatographic analysis was subtracted from the final pressure

Table 1. Tritium Distribution and Hydrogen Exchange after Reaction of Tetralin with Tritiated Gaseous Hydrogen at 25 atm*

Reaction Condition				Tritium Distribution		
Temp. (°C)	Flow Rate of H_2 (mL/min)	Flow Rate of Tetralin (mL/min)	Residence Time (s)	R_{gas} (dpm/h)	$R_{solvent}$ (dpm/h)	Hydrogen Exchange (mg/g Tetralin)
400	80	0.4	25	999,696	304	5.27×10^{-3}
400	40	0.2	49	999,273	723	1.23×10^{-2}
400	20	0.1	98	998,872	1,128	1.91×10^{-2}
425	80	0.4	24	999,353	647	1.10×10^{-2}
425	40	0.2	47	998,820	1,180	1.89×10^{-2}
425	20	0.1	94	998,415	1,585	2.68×10^{-2}
450	80	0.4	23	998,911	1,088	1.84×10^{-2}
450	40	0.2	46	998,162	1,838	3.14×10^{-2}
450	20	0.1	91	996,881	3,119	5.29×10^{-2}

*Total radioactivities were normalized to 10^6 dpm.

Table 2. Tritium Distribution and Hydrogen Exchange After Reaction of Tetralin with Tritiated Gaseous Hydrogen at 50 atm*

Reaction Condition				Tritium Distribution		
Temp. (°C)	Flow Rate of H_2 (mL/min)	Flow Rate of Tetralin (mL/min)	Residence Time (s)	R_{gas} (dpm/h)	$R_{solvent}$ (dpm/h)	Hydrogen Exchange (mg/g Tetralin)
400	80	0.4	48	999,312	687	1.18×10^{-2}
400	40	0.2	96	999,043	957	1.64×10^{-2}
400	20	0.1	192	998,754	1,246	2.13×10^{-2}
425	80	0.4	46	998,781	1,219	2.06×10^{-2}
425	40	0.2	93	998,185	1,815	3.06×10^{-2}
425	20	0.1	186	997,657	2,343	3.97×10^{-2}
450	80	0.4	45	998,384	1,616	2.74×10^{-2}
450	40	0.2	90	997,811	2,189	3.69×10^{-2}
450	20	0.1	179	995,739	4,261	7.23×10^{-2}

*Total radioactivities were normalized to 10^6 dpm.

in the gas phase. The isotope effect was regarded as small and was ignored in these calculations because most of the reactions were performed at comparatively high temperature. Recently, it was reported that the isotope effect was negligible under coal liquefaction conditions (Kamo et al., 1993):

$$\mathrm{HER} = H_{ex(G \Leftrightarrow S)}/H_{\mathrm{solvent}} \tag{1}$$

$$H_{ex(G \Leftrightarrow S)} = H_{\mathrm{gas}} \times R_{\mathrm{solvent}}/R_{\mathrm{gas}} - H_{adt} \tag{2}$$

where

HER = hydrogen exchange ratio

$H_{ex(G \Leftrightarrow S)}$ = amount of exchanged hydrogen in solvent with gaseous hydrogen

H_{solvent} = amount of hydrogen in solvent

H_{gas} = amount of hydrogen in gas phase

R_{solvent} = radioactivity in solvent after reaction

R_{gas} = radioactivity in gas phase after reaction

H_{adt} = amount of hydrogen added from gas phase to solvent after reaction

Results and Discussion

Reaction of tetralin with tritiated hydrogen in a flow reactor

Reactions of tetralin with tritiated hydrogen were performed under the conditions 400–450°C and 2.5–9.8 MPa for 25–420 s. Yields of the products at 425°C are plotted against the reaction time in Figure 2. In this temperature range, the reaction products from tetralin were 1-methylindan by isomerization (Figure 2a), naphthalene by dehydrogenation (Figure 2b), and *n*-butylbenzene by hydrocracking (Figure 2c); the main product was 1-methylindan. Decalin by disproportionation was not formed under these conditions. It is consistent with the previous result in the reaction using the autoclave (Godo 1997a), in which the disproportionation of tetralin to decalin does not occur. The products increased monotonously with residence time, and the yields of methylindan, naphthalene and butylbenzene at 425°C for 450 s reached 0.13, 0.05 and 0.06%, respectively. As shown in Figure 2a, the plots of methylindan at 2.5, 4.9 and 9.8 MPa showed approximately the same straight line and were not influenced by the reaction pressure. A similar result was also obtained for naphthalene (Figure 2b). The results in the reaction using the autoclave were also plotted in Figure 2, which were conducted under the conditions at initial pressure 5.9 MPa for 0–300 min. In these figures, the ratios between the reaction time and the yields of products in the reactions using the autoclave are the same as those in the reactions using the flow reactor. So the relationship between the reaction time and the product yield can be compared within the same figure. The yields of methylindan and naphthalene in the reaction using the autoclave were a little less than those of the flow reactor; however, these values were not affected significantly by the type of reactor. In the reaction using the autoclave, although the initial pressure was 5.9 MPa, the effective reaction pressure was about 14.7 MPa at 425°C by thermal

Table 3. Tritium Distribution and Hydrogen Exchange after Reaction of Tetralin with Tritiated Gaseous Hydrogen at 100 atm*

Reaction Condition				Tritium Distribution		
Temp. (°C)	Flow Rate of H_2 (mL/min)	Flow Rate of Tetralin (mL/min)	Residence Time (s)	R_{gas} (dpm/h)	$R_{solvent}$ (dpm/h)	Hydrogen Exchange (g)
400	80	0.4	130	999,417	583	9.92×10^{-3}
400	40	0.2	260	998,632	1,368	2.33×10^{-2}
400	20	0.1	520	997,977	2,023	3.49×10^{-2}
425	80	0.4	112	996,943	3,057	5.20×10^{-2}
425	40	0.2	224	995,430	4,570	7.73×10^{-2}
425	20	0.1	448	994,099	5,901	1.01×10^{-1}
450	80	0.4	105	996,694	3,306	5.54×10^{-2}
450	40	0.2	210	990,802	9,198	1.56×10^{-1}
450	20	0.1	420	984,120	15,880	2.71×10^{-1}

*Total radioactivities were normalized to 10^6 dpm.

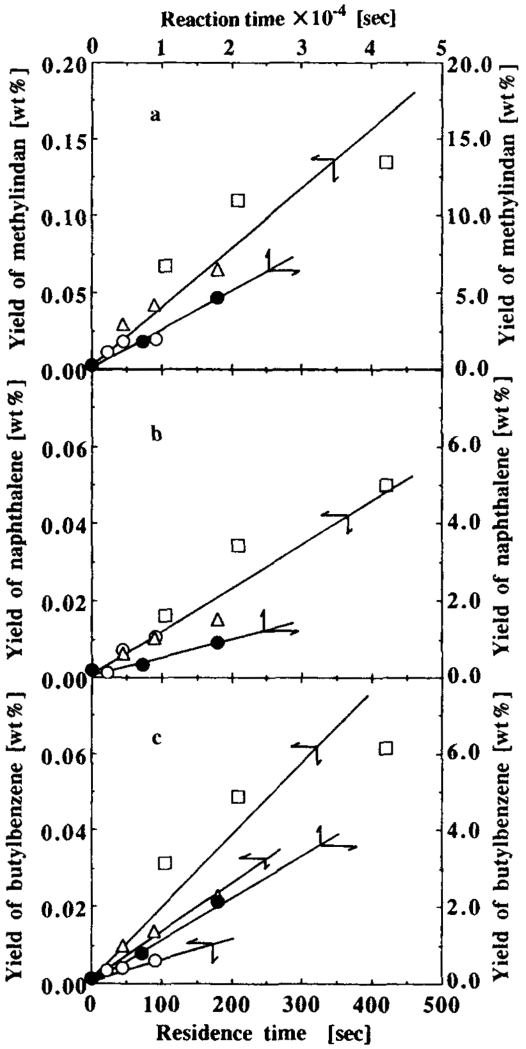


Figure 2. Effect of residence time on the yield of methylindan.

Reaction temperature: 425°C. ○: 2.5 MPa; △: 4.9 MPa; □: 9.8 MPa; ●: autoclave.

expansion of the gas phase. The result shows that yields of methylindan and naphthalene are not affected greatly by the reaction pressure in the range from 2.5 to 14.7 MPa. In contrast, yields of butylbenzene of the reaction in a flow reactor increased with the rise in reaction pressure, and the formation of butylbenzene depended on the reaction pressure. The yield of butylbenzene in the reaction using the autoclave also increased steadily with reaction time because the reaction pressure of the autoclave was nearly equal when the reaction temperatures were the same and was close to that in the reaction of a flow reactor at 4.9 MPa.

The amount of tritium introduced into tetralin from the gas phase was represented by the HER (see the Experimental Section). Figure 3 shows the change in HER with reaction time. The HER of tetralin increased gradually for a period of time, and reached 0.11% at 425°C for 450 s. However, at 400°C, the hydrogen-exchange ratio was only about 0.04% for 520 s. Further, the plots of HER could be shown with the same line in the pressure range from 2.5 MPa to 9.8 MPa. In addition, using the autoclave, HER in the reaction was almost the same as that in a flow reactor. The result is similar to that of the conversions of tetralin into methylindan and naphthalene.

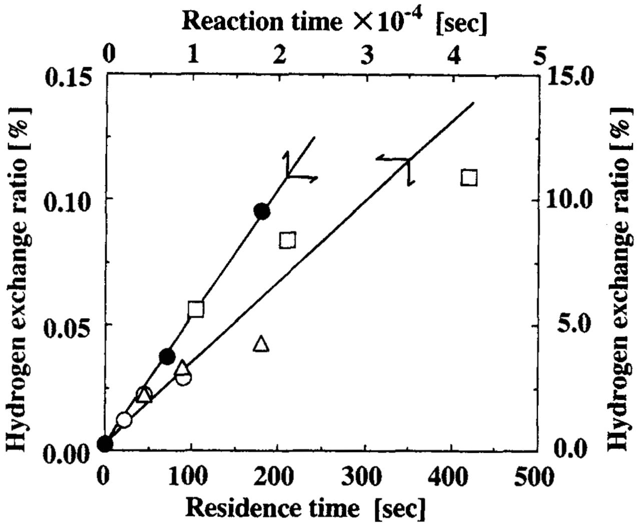


Figure 3. Effect of residence time on the hydrogen exchange ratio of tetralin.

Reaction temperature: 425°C. ○: 2.5 MPa; △: 4.9 MPa; □: 9.8 MPa; ●: autoclave.

First-order plots of these data for the formation of methylindan and naphthalene, and hydrogen exchange are shown in Figure 4. All plots were approximately the same as the linear relationships, indicating that these reactions could be treated as first-order reactions. The formation rate constants of methylindan from tetralin could be determined from the slopes of first-order plots as shown in Eq. 3:

$$k = -\frac{1}{t}\ln(1-X) \tag{3}$$

where X is the conversion of tetralin into methylindan; k is the rate constant for formation of methylindan; and t is the residence time, which is calculated by the volume of reaction zone and the flow rate of the reactant. These constants are shown in Table 4. In the same way, the formation rate constants of naphthalene, and the rate constants of hydrogen exchange were obtained and are also listed in Table 4. Figure 5 shows Arrhenius plots of the rate constants of tetralin conversions into methylindan and naphthalene, and hydrogen exchange between tetralin and tritiated hydrogen. Activation energies of the tetralin conversions into methylindan and naphthalene, and hydrogen exchange reaction were 32 ± 2, 33 ± 2, and 33 ± 2 kcal/mol, respectively, as shown in Table 5. Activation energies in the reaction using the autoclave are also in Table 5. These values were very close to each other. This suggested that the conversion of tetralin into methylindan and naphthalene, and the hydrogen-exchange reaction of tetralin depended on the same reaction mechanism.

Reaction mechanism of tetralin with tritiated hydrogen

Our previous reports assumed that formations of methylindan and naphthalene, and the hydrogen transfer from the tetralin, proceeded by a tetralyl radical, which acted as an

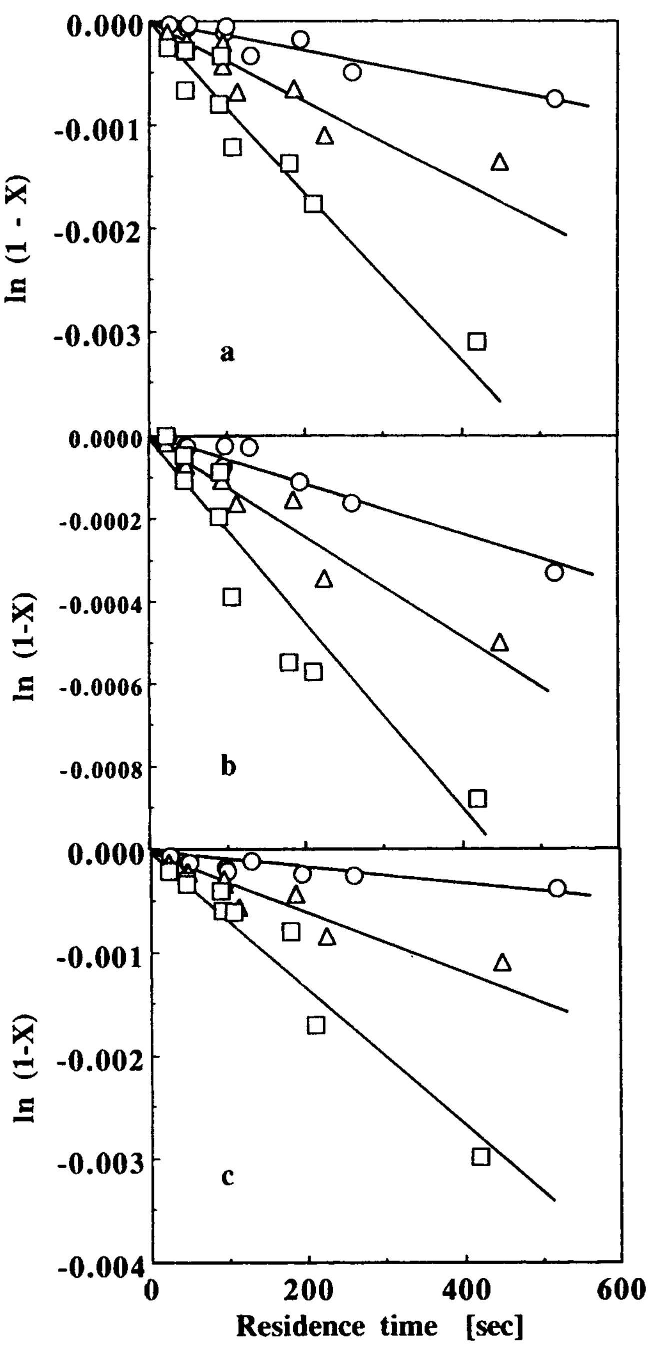


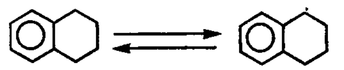
Figure 4. First-order plot of conversion of tetralin and HER.

(a) Conversion of tetralin into methylindan; (b) conversion of tetralin into naphthalene; (c) HER. ○: 400°C; △: 425°C; □: 450°C.

intermediate in the conversion and the hydrogen exchange of tetralin (Kabe et al., 1990b; Ishihara et al., 1995; Godo et al., 1997a). In the system of tetralin and gaseous hydrogen, the conversions into methylindan and naphthalene, and the hydrogen-exchange reaction of tetralin, were not changed by the reaction pressure. The result shows that gaseous hydrogen does not affect the formation of a tetralyl radical, at least in the present reaction system using a flow reactor. Tetralin may be activated by collisions and become a tetralyl radical and a hydrogen atom by the means of Eq. 4.

Table 4. Rate Constants in the Reaction of Tetralin with Tritiated Hydrogen

Temp. (°C)	Conversion of Tetralin: Into Methylindan ($\times 10^{-6}$ s^{-1})	Conversion of Tetralin: Into Naphthalene ($\times 10^{-6} s^{-1}$)	Hydrogen Exchange of Tetralin ($\times 10^{-6} s^{-1}$)
400	1.6±0.3	3.7±0.7	8.3±1.7
425	0.5±0.1	1.0±0.2	2.8±0.6
450	1.1±0.2	2.7±0.5	6.5±1.3

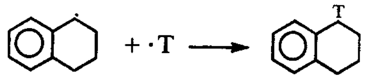


(4)

A part of the hydrogen atoms produced from tetralin would react with the tritiated hydrogen molecule, leading to the formation of a tritium atom, as shown in Eq. 5.

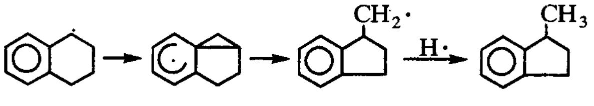
$$HT + H\cdot \longrightarrow H_2 + T\cdot \qquad (5)$$

If the tetralyl radical is quenched by tetralin to form the original tetralin, the conversion of tetralin and hydrogen exchange would be inhibited. The reaction of the tetralyl radical with tritium atom leads to hydrogen exchange in the reproduction of tetralin, as shown in Eq. 6.



(6)

The hydrogen exchange between tetralyl radicals and tritiated hydrogen can proceed by the radical hydrogen-transfer reaction depending on the concentration of tetralyl radicals, which also control the formation of methylindan and naphthalene in Eqs. 7 and 8.



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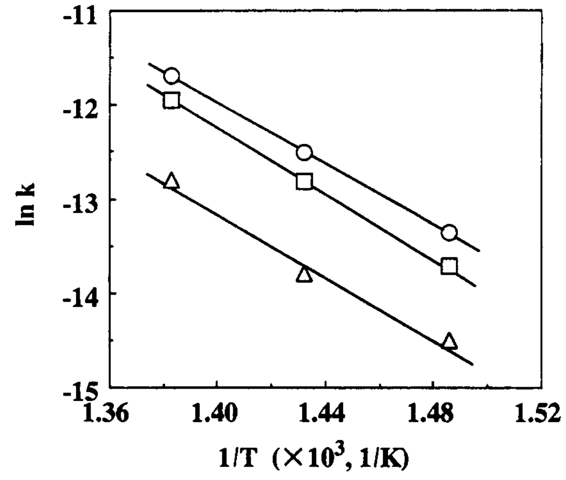
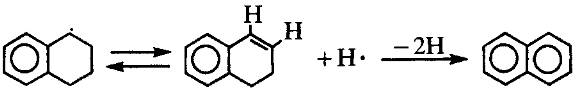


Figure 5. Arrhenius plots of rate constants of tetralin conversion into methylindan and naphthalene, and hydrogen exchange.

○: methylindan; △: naphthalene; □: hydrogen exchange.

Table 5. Activation Energies in the Reaction of Tetralin with Tritiated Hydrogen

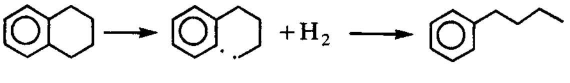
Reactor	Conversion of Tetralin Into Methylindan (kcal/mol)	Conversion of Tetralin Into Naphthalene (kcal/mol)	Hydrogen Exchange of Tetralin (kcal/mol)
Flow reactor	32±2	33±2	33±2
Autoclave	32±2	33±2	30±2



(8)

Therefore, the formation of the tetralyl radical by unimolecular scission in this system may be the rate-determining step for both the conversion of tetralin into methylindan and naphthalene, and the hydrogen exchange. At 400°C, the conversion and hydrogen exchange ratio of tetralin were very low. If coal is included in the system, a tetralyl radical may be formed easily. However, if coal that forms a radical is not included, a tetralyl radical is difficult to form. Then, the unimolecular scission of tetralin hardly occurs at 400°C.

Hooper et al. (1979) reported that butylbenzene was formed by the thermal dissociation of tetralin, as shown in Eq. 9:



(9)

While the plot of the yields of butylbenzene showed some scatter, the effect of the reaction pressure was seen at every temperature. It is possible that molecular hydrogen affected the hydrogenation of butylbenzyl radical.

Conclusion

The hydrogen-exchange mechanism between tetralin and gaseous hydrogen was investigated in a flow-type reactor to clarify the effect of the reaction pressure. The yields of 1-methylindan and naphthalene, and the HER of tetralin were not affected by the pressure. The tetralyl radical that was considered to be an intermediate of the thermal and hydrogen exchange reactions was produced by the unimolecular thermal dissociation of tetralin.

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