

In-situ infrared thermographic analysis during dehydrogenation of cyclohexane over carbon-supported Pt catalysts using spray-pulsed reactor

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Infrared thermography, a tool used for screening of active catalytic materials generally during the exothermic reactions has been used for thermal imaging during strong endothermic reaction of dehydrogenation of cyclohexane on Pt catalyst supported on active carbon cloth (CFF-1500s) sheets. A spray-pulsed mode was used for injection of atomized cyclohexane and to create alternate wet and dry condition on catalyst surface. The simultaneous product gas analysis and recording of the temperature profile of the catalyst surface using an IR camera was carried out. The production rate of hydrogen via endothermic dehydrogenation reaction is greatly dependent on the temperature of the catalyst surface. The observed change in the temperature profile at wet and dry conditions with varying pulse-injection frequency and corresponding product gas analysis reveals that the spray-pulse mode is useful in improving the catalyst activity. Further the reaction conditions were optimized using thermal profile data.

KEY WORDS: IR thermography; cyclohexane dehydrogenation; hydrogen production; Pt catalyst; spray-pulse injection.

1. Introduction

Infrared (IR) thermography has been a successful tool used in time-resolved monitoring of heterogeneous catalytic reactions and for rapid screening of active materials in combinatorial chemistry. The several applications of IR thermography include monitoring of the dynamics of reactions on solid surfaces [1,2], recording of the oscillating behavior during CO oxidation on Rh-SiO₂ [3,4] and over a thin Rh-SiO₂ catalyst layer supported on metallic substrate [5]. Using the fact that the thermal change on the catalysts surface is related to its activity and is dependent on the heat of reaction, the IR thermography was adopted for identifying superior catalyst formulations from the libraries of several candidates in a parallel system by monitoring the temperatures [6–9]. Most of the reactions under consideration for IR thermography were exothermic in nature, it was later reported that the exothermicity is not a requirement in IR thermographic screening of the catalyst and it may also be applicable to endothermic and even thermoneutral reactions. The Ru-catalyzed ring-closing olefin metathesis was used as reaction for this purpose [10]. The more recent reports on application of IR-thermography includes the study of the deactivation of water-pretreated Pt/ γ -Al₂O₃ for low

temperature selective CO oxidation in hydrogen [11], and the study of oscillatory behavior during catalytic oxidation of CO over Pd/glass cloth [12].

Inspired by the applications of IR thermography, we decided to use this approach for explaining spray-pulse mode dehydrogenation of cyclohexane. Although it has been demonstrated earlier that the IR thermography can be used for endothermic reactions such as Ru-catalyzed ring-closing olefin metathesis; the reaction is only slightly endothermic. Indeed the IR thermography has mostly been used for monitoring the dynamics of the exothermic reactions. We have for the first time used strongly endothermic reaction of dehydrogenation of cyclohexane (heat of reaction equal to 205.9 kJ mol⁻¹) over Pt catalyst, to demonstrate the use of IR thermography for optimizing the reaction conditions.

Dehydrogenation of cycloalkanes such as cyclohexane, methylcyclohexane, and decalin to produce hydrogen has been used for storage and transportation of hydrogen fuel at ambient temperature and pressure [13–17]. The advantages of cycloalkane dehydrogenation systems are: (a) The cycloalkanes have high hydrogen contents (6–8 wt%); (b) The reaction is very selective and unlike the other process produces complete dehydrogenated aromatics and hydrogen as the products with no CO, CO₂ and other byproducts; (c) The reactions of dehydrogenation and hydrogenation are reversible with reactants or products

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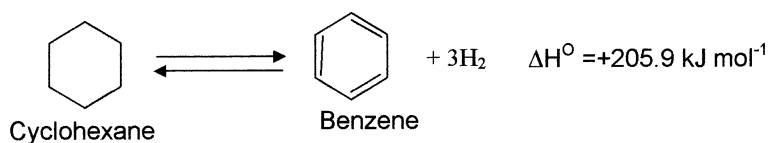
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are recyclable. Due to which the system may be more effectively used as compared to the other hydrogen storage materials; and (d) Boiling point of cycloalkanes is high (80–190 °C), and therefore do not require special infrastructure for its long-distance transportation. As the dehydrogenation of cycloalkanes is endothermic and chemical equilibrium is favorable for dehydrogenation at higher temperature, the reactions were performed at high temperature using steady state operation under gas phase or under liquid-film state [18,19]. We have reported earlier, in case of dehydrogenation of cyclohexane [20,21] and reforming of iso-octane [22] that by feeding of atomized liquid reactant in pulses and alternatively creating wet and dry conditions on heated solid catalyst surface improves the yield of reaction. The alternate wet–dry

and dried at about 110 °C. The Pt loading was ca. 5 wt.% on activated carbon. Further, the catalyst was activated in H₂ flow (99.998%, 50 mL min⁻¹) by following a predetermined heating cycle to reach 300 °C and then keeping temperature at 300 °C for 2 h. The Pt-loaded carbon supports were characterized using X-ray diffraction. The peaks observed in XRD patterns can be attributed to Pt metal. Particle size of Pt based on XRD observations was ca. 2–3 nm.

2.2. Reactions

The dehydrogenation of cyclohexane over Pt catalysts supported on active carbon sheet CFF-1500s has been carried out using pulse-spray injection condition in this study. The reaction is shown in scheme 1.



Scheme 1

conditions created on catalyst surface helps easy evaporation of liquid reactant on the catalyst surface. The evaporation of the reactant on catalyst surface enriches the catalyst surface with vapor-phase reactants and improves catalyst-reactant contact. The easy removal of unreacted cyclohexane leaves back the cleaned catalyst at the end of the dry step. The unsteady-state results in high frequency factor and show several fold increase in the reaction rate using pulse-spray injection conditions to create alternate wet–dry condition on catalyst surface [20,21]. The controlling factors to maintain the temperature of catalyst surface under spray-pulsed condition are heat transfer to the catalyst surface, pulse-injection frequency and pulse width. We have studied the reaction dynamics and temperature profile of the catalyst surface for Pt/Carbon cloth using IR thermography during cyclohexane dehydrogenation for possible further explanation of spray-pulsed reaction condition.

2. Experimental

2.1. Catalyst preparation

Monometallic Pt catalysts supported on carbon sheets were prepared by immersing active carbon sheet (supplied by Toyobo Co. Ltd, Japan) into 200 mL of aqueous solution of H₂PtCl₆·6H₂O (98% Wako Pure Chemicals) for a period of 24 h. The active carbon sheet (CFF-1500S) used was about 80 cm², weight 0.7–0.8 gm and surface area 1500 m² g⁻¹. After 24 h the solution turns clear and the solution was filtered out. The Pt-impregnated samples were washed by deionized water

The experimental setup used for cyclohexane dehydrogenation to benzene and hydrogen using spray-pulse mode is shown in figure 1. The experiments were targeted to monitor the temperature profile of catalyst surface and simultaneous product analysis under various operating conditions to facilitate understanding of the spray-pulse phenomenon. The main reactor was made of stainless steel equipped with a fine nozzle at the top for atomized injection of liquid reactant (cyclohexane). A pressure of 12 MPa was maintained in the fuel supply line to generate atomized spray. Injection frequency was controlled using a frequency generator and pulse injection width was kept constant at 0.8 s. The

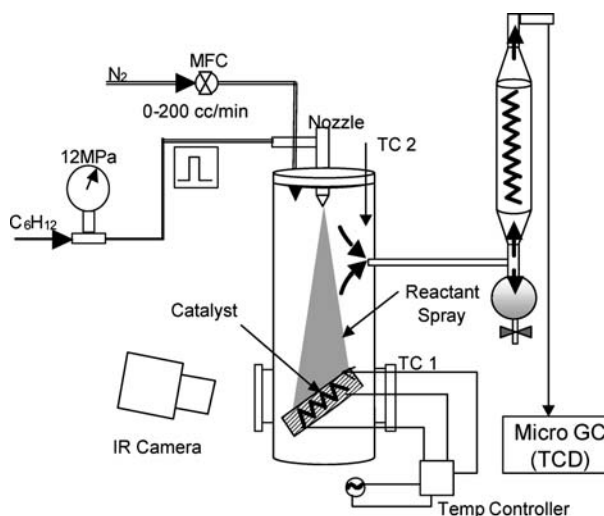


Figure 1. Experimental set-up for pulse-spray injection reactor used to conduct cyclohexane dehydrogenation and IR thermographic analysis.

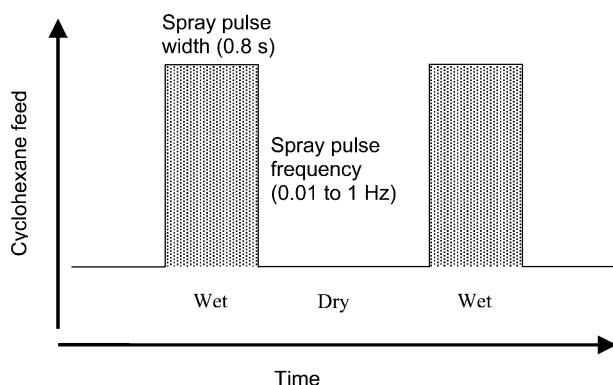


Figure 2. Schematic representation of pulse feeding of cyclohexane to the reactor.

modulation of pulse-injection frequency to create alternate wet and dry condition on catalyst surface is shown in figure 2. The cyclohexane feed rate was varied from 0.268 to 26.8 mmol min⁻¹ by varying the pulse-injection frequency from 0.01 to 1 Hz. A carrier gas N₂ was introduced from the top of the reactor. The catalyst was placed at lower side of the reactor. The catalyst was placed in tilted position with respect to the vertical axis of reactor and facing to a side watch glass window to allow the monitoring of the catalyst surface temperature by a non-contact thermal recorder. An IR camera model Neo-Thermo TVS-8500 (provided by Nippon Avionics Co. Ltd) has been used for this purpose. The frame rate of 60 frames per second has been used for recording temperature profile of the catalyst surface. The reading of temperature by IR camera was calibrated by comparing with temperatures recorded by thermocouple at four different points. Emissivity correction was applied to record the surface temperature of catalyst. The catalyst was heated using a disk type heater and the heating temperature was controlled at 250–300 °C using a temperature controller and thermocouple (thermocouple TC1 in figure 1) arrangement. The thermocouples TC1

and TC2 were used for monitoring apparent heating temperature of catalysts and gas-phase temperature, respectively. The unreacted cyclohexane and condensable products were collected in a reservoir connected to condenser. The product analysis was carried out using FID-GC (Shimadzu GC-14B, J&W-DB-WAX, 30 m, column i.d. 0.25 mm, film 0.25 μm) and the gas products were periodically analyzed using a TCD micro GC (Agilent, molecular sieve 5A, 10 m).

3. Results and discussion

3.1. Effect of the temperature

The reaction of dehydrogenation of cyclohexane to benzene and hydrogen has been carried out at different heating temperatures from 250 to 300 °C in step change of 10 °C each. The injection frequency of cyclohexane was kept constant at 0.1 Hz (corresponding to 2.68 mmol min⁻¹). Figure 3 shows the time series data of hydrogen evolution rates over 5 wt.% Pt supported on CFF-1500 S active carbon sheet at varying temperatures. The rate of hydrogen evolution was observed to be increasing marginally from 260 to 300 °C as compared to the large difference between 250 and 260 °C. The maximum rate of hydrogen evolution was 2 mmol min⁻¹, with corresponding efficiency of 30% for conversion of cyclohexane. The reaction rate for lower temperatures apparently remains in unsteady state, while at the higher temperatures of 280–300 °C the reaction reaches to equilibrium at about 20 min after the start. The lower rate of hydrogen evolution and later attainment of equilibrium may be attributed to the slow evaporation of the reactant on catalyst surface and forming the wet area to reduce the temperature of catalyst. Since the dehydrogenation of cyclohexane is endothermic, the higher reaction temperature is more favorable. The higher reaction temperature also promotes rapid elimination of aromatic products and

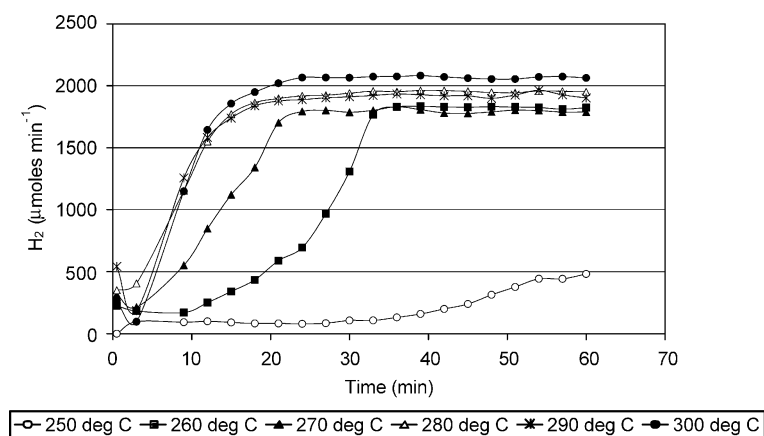


Figure 3. Hydrogen production rates via dehydrogenation of cyclohexane over Pt/AC catalyst at various heating temperatures, the pulse-injection frequency 0.1 Hz.

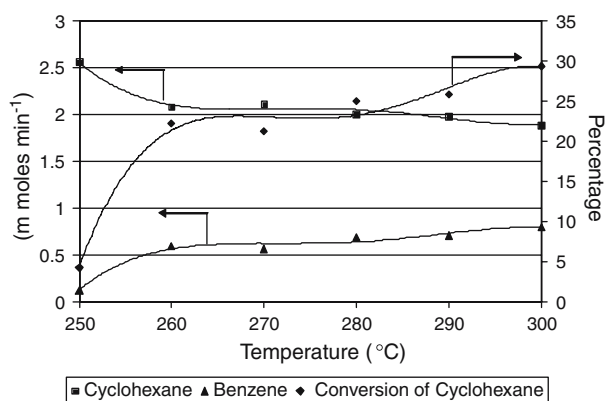


Figure 4. Concentrations of cyclohexane and benzene in product and cyclohexane conversion at different heating temperatures.

hydrogen from catalyst surface resulting in suppression of the reverse reaction. The theoretical equilibrium conversion of cyclohexane over Pt catalyst may be about 30% and 92% at 250 °C and 300 °C, respectively. The 30% conversion obtained in this study at heating temperature (may be designated as T_w) of 300 °C suggests that the average temperature of catalyst surface may be in the range of 250 °C. This is further supported by the thermal profile of the catalyst surface in the data reported in later section.

Conversion of cyclohexane to benzene was estimated using the FID-GC analysis of the condensed products after completion of the reaction. The conversion efficiency generally observed to be increasing with increase in catalyst heating temperature with a marginal increase from 260 to 300 °C as shown in figure 4. The condensed products contain cyclohexane and benzene. No other products such as partially dehydrogenated products (cyclohexene, methylcyclohexene) were observed. Also no hydrocarbons were observed in the product gas. The selectivity of the catalyst for dehydrogenation of cyclohexane to produce hydrogen and benzene was observed to be as high as 100%.

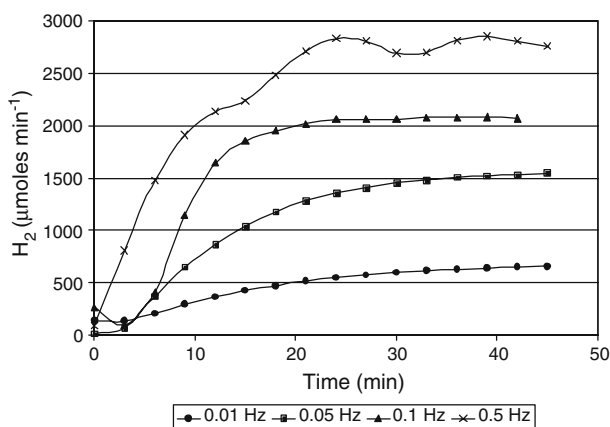


Figure 5. Effect of different feed rates of cyclohexane on rate of hydrogen production, catalyst heating temperature 300 °C.

3.2. Effect of cyclohexane feed rate

The rates of hydrogen evolution were observed to be dependent on cyclohexane feed rates. The feed rate of reactant was varied on catalyst by changing the pulse-injection frequency as 0.01, 0.05, 0.1, 0.5 Hz. With increase in the feed rate, the hydrogen evolution observed to be increasing at the heating temperature of 300 °C. However, the increase in hydrogen evolution is not linear with feed rate. For example, change in feed rate by 50 folds the hydrogen evolution rate increased by about 5 times as shown in figure 5. The conversion efficiency decreases with increase in feed rate of cyclohexane e.g. at the injection frequency of 0.01 Hz conversion efficiency was 39% and at 0.5 Hz the conversion was 12.7%. However, at the heating temperature of 300 °C the sufficient energy is available for evaporation of cyclohexane and dehydrogenation reaction. The energy required for evaporation and the dehydrogenation reaction was calculated at heating temperature of 300 °C and at pulse-injection frequency of 0.5 Hz. Considering the conversion efficiency of about 12.7% at feed rate of 0.5 Hz, the energy requirement is 21.25 J/s. At the same feed rate the energy required for complete conversion of cyclohexane injected is 61.39 J/s. In a separate experiment using same reactor and heater arrangement, the transient heat flux from plate type heater to the catalyst was estimated to be about 1.5×10^4 W/m². This indicates that the heat provided to the catalyst was about 61.04 J/s. This energy is sufficient to meet the total energy requirement for evaporation and dehydrogenation reaction for cyclohexane dose equivalent to 13.4 mmol min⁻¹ corresponding to 0.5 Hz. Therefore, increasing the pulse-injection frequency till 0.5 Hz at heating temperature of 300 °C resulted in increase in hydrogen production rate. Further, increase in injection frequency forms a liquid pool on the catalyst surface. The increase in hydrogen evolution rate was not observed at heating temperature (T_w) of 250 °C when cyclohexane feed rate was increased. Figure 6 reveals that at the heating temperature of 250 °C the hydrogen evolution rate decreased when

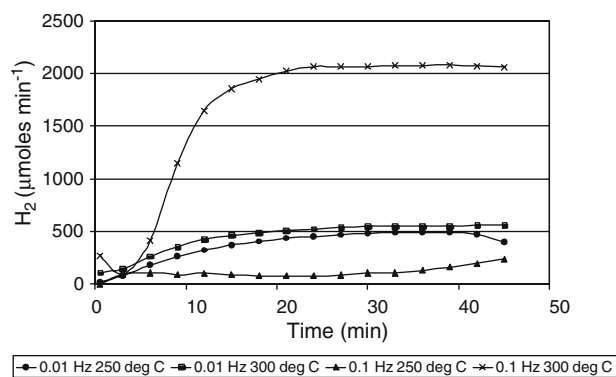


Figure 6. Effect of change in feed rates of cyclohexane on the rate of hydrogen production at different heating temperatures.

injection frequency was increased from 0.01 to 0.1 Hz. This may be explained as at heating temperature of 250 °C the actual catalyst temperature may be still lower and the heat flux to catalyst surface may not be sufficient to provide required energy for evaporation of reactants and for endothermic dehydrogenation reaction. Nearly a liquid pool might have formed on the catalyst surface, and thus resulted in decrease in hydrogen production rate. The IR thermography results shows the average temperature of catalyst surface was much lower as compared to set heating temperature.

3.3. Temperature profile during pulse-spray injection

Alternate wet and dry conditions were created on the catalyst surface for formation of vapor phase reactant in the vicinity of catalyst surface and also to facilitate the easy removal of products or evaporation of unreacted cyclohexane. We have monitored the thermal profile of the catalyst surface during unsteady state and equilibrium part of the reaction at various operating conditions with respect to pulse-injection frequency and bulk cat-

alyst heating temperature. The temperature read by thermocouple TC1 (as shown in figure 1) was used for monitoring and control of catalyst heating temperature, designated as T_w . Temperature in void space designated as T_i in reactor was monitored by thermocouple TC2. The temperature profile of catalyst surface was recorded using an IR camera with recording speed of 60 frames per second. The thermographs were generated at interval of 10 frames each. Figure 7 shows the thermographs for the pulse-injection frequency of 0.1 Hz, $T_w = 300$ °C and T_i observed was 114 °C. The circular portion at the central part of the thermograph is catalyst surface and is the area of interest in the following discussions, whereas other portion at the periphery is of the reactor body, which is at lower temperatures. Since the catalyst support used was carbon cloth, during the reaction flow of vapor of cyclohexane and product hydrogen and carrier gas the contact between catalyst and heater plate is not uniform as revealed by temperature variations on catalysts surface observed in thermographic images. Also due to introduction of cyclohexane on the catalyst surface there may be some

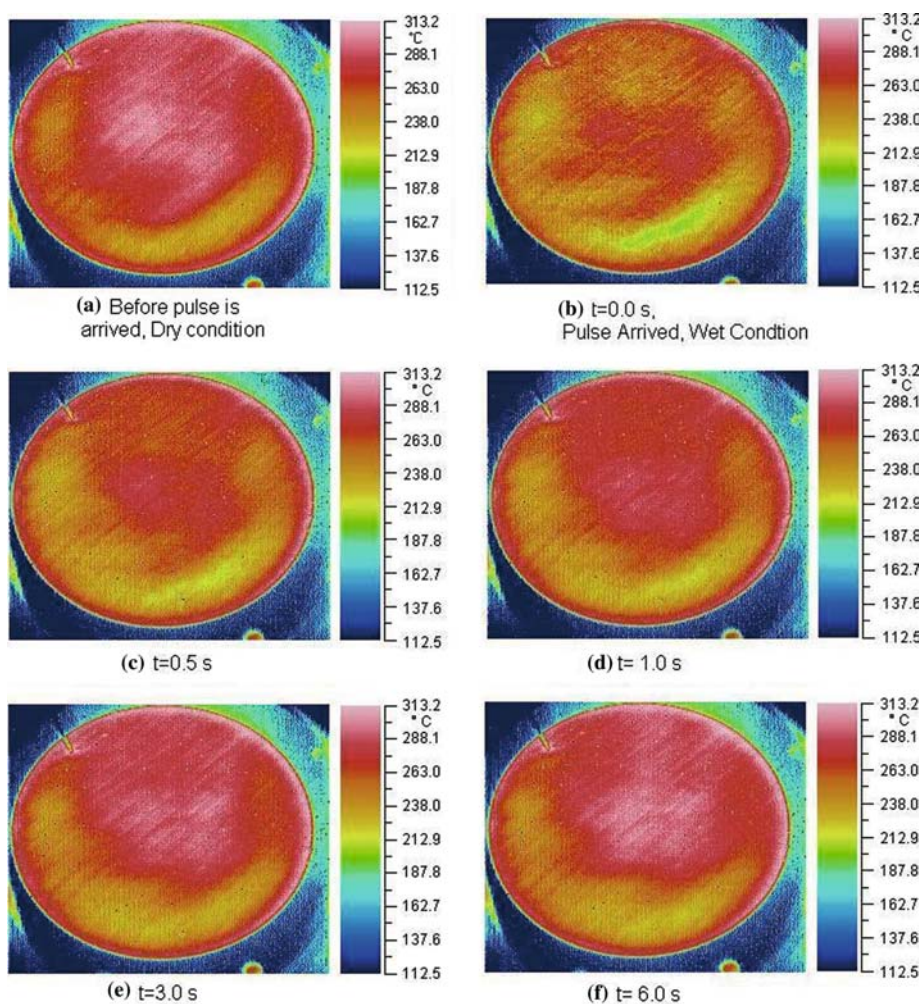


Figure 7. Thermographic image showing spread of wet area on catalyst surface at set heating temperature $T_w = 300$ °C, pulse-feed frequency 0.1 Hz, unsteady state.

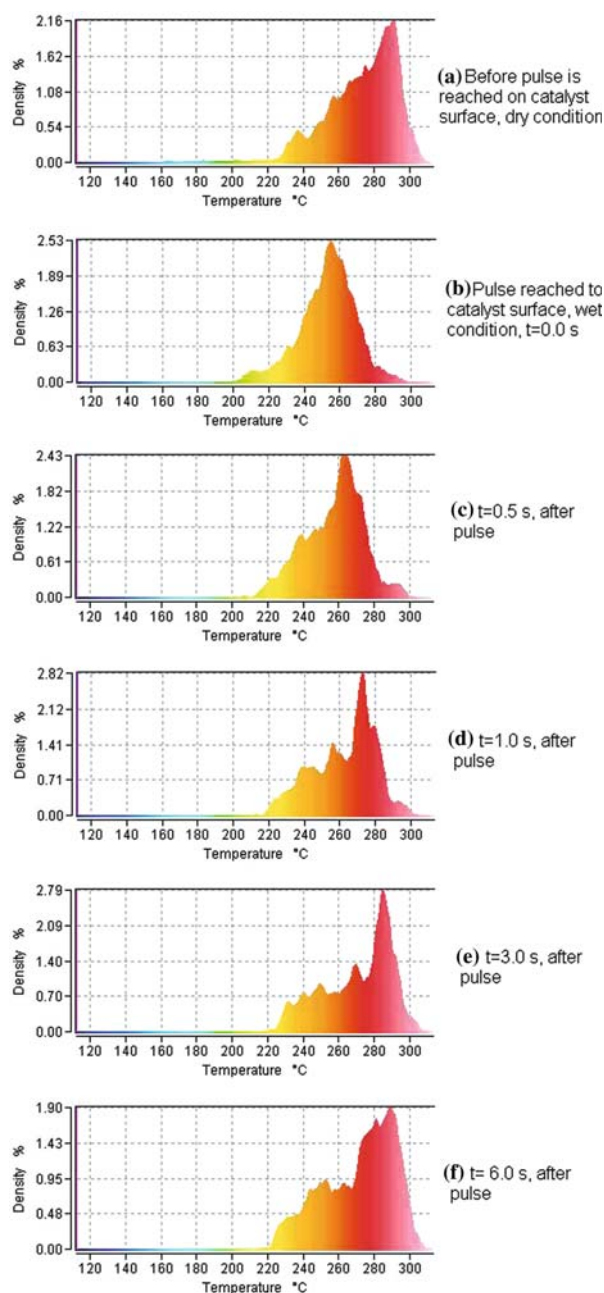


Figure 8. Histograms derived from IR thermal images showing chronology of temperature profile change at 0.1 Hz, $T_w = 300$ °C, unsteady state.

variation in the emissivity depending on the contact between cyclohexane and catalyst. The corresponding histograms derived from the IR images in figure 7 at 0.1 Hz and heating temperature of 300 °C are shown in figure 8 and may be referred for further explanation of the behavior of the spray-pulsed injection mode. At the initial time when reactant spray pulse was not arrived on the catalyst surface, the catalyst was under dry condition with average surface temperature of 276 °C. When the first pulse was injected on catalyst surface ($t = 0.0$ s) the most of the catalyst was covered with liquid

cyclohexane and temperature was lowered with major area of catalyst in the range of about 220–280 °C. A part of the catalyst surface was at above 280 °C. During the dry step (interval between two pulse injections) as time progresses wet area gets reduced and temperature of the most of the part of the catalyst increases and at $t = 6.0$ s temperature reaches between 270 and 300 °C with a small portion at lower temperatures of about 240 °C. The average temperature of catalyst surface was increased from 253 °C at wet condition to 272 °C at dry condition at unsteady state. The overall and average temperature of the catalyst surface was observed to be increased at dry condition at all frequencies. This explains the advantage of using spray-pulsed injection mode for improving the catalyst activity. Since during the dry condition catalyst surface is free from the liquid reactant, its temperature is increased. The increased temperatures facilitate to keep the higher catalytic activity.

The void space temperature T_i (112–114 °C) was well above the b.p. of cyclohexane, and thus there may be evaporation of considerable amount of cyclohexane from atomized spray before reaching to the catalyst surface. These vapors may be carried out by carrier gas as unreacted cyclohexane. The higher is the value of T_i the percentage of unreacted cyclohexane in products may increase.

The temperature profile of the catalyst surface at feed pulse-injection frequency of 0.1 Hz and at steady state of the reaction after about 33 min (corresponding to more than 198 pulse injections) is shown in histogram in figure 9. When the reaction was continued and catalyst was exposed to several injection pulses of the reactant, the temperature of the almost all area of the catalyst was lowered to the average temperature of about 257 °C with temperatures ranging between 220 and 280 °C during the dry condition i.e. when pulse was not arrived. When pulse was arrived ($t = 0.0$ s) and catalyst surface was wet, the average temperature was lowered to 228 °C. Whereas during dry condition on catalyst surface average temperature increases and was nearing to 290 °C (at $t = 0.0$ s) with average temperature increasing to 252 °C. The temperature of catalyst surface was observed to be on lower side during steady state (about after 33 min) as compared to unsteady state (first few pulses). The lower temperatures of catalysts (still well above the b.p. of reactant) in this condition may be forming a vapor phase of reactant at very close vicinity of the catalyst and most of the time the reactant–catalyst contact is maintained which results in the higher catalytic activity at steady state and maintaining the production rate of hydrogen. This is also clear from distinct doublet observed in figure 9. A portion of catalyst surface is still at lower temperature at $t = 1.0$ s and a part is already being dried. The spatial variation in the catalyst surface temperature may be the reason for doublet occurring at 0.1 Hz. After about 3 s almost all

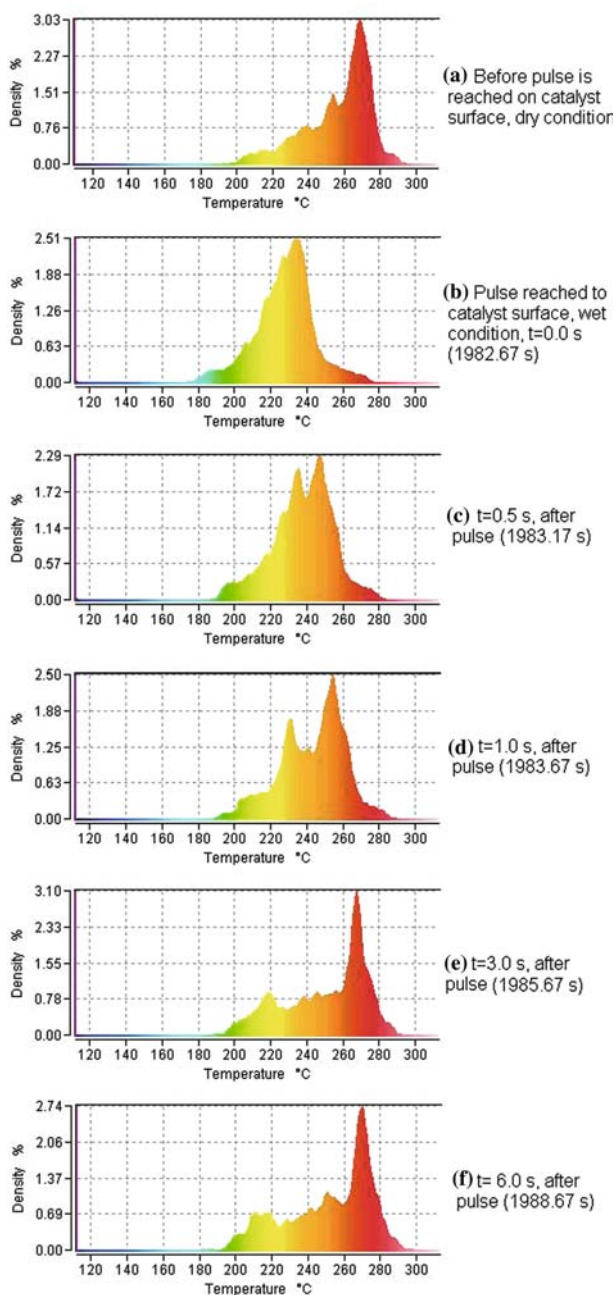


Figure 9. Histograms showing the chronology of temperature profile at 0.1 Hz, $T_w = 300\text{ }^{\circ}\text{C}$, steady state, time in brackets indicate accumulated time from the start of the reaction.

surface of the catalysts becomes dry and the temperature is more uniform. At more uniform temperature profile the doublet disappears.

The variations in temperature profile at steady state at the pulse-injection frequency of 0.5 Hz and the heating temperature T_w at $300\text{ }^{\circ}\text{C}$ is shown in histograms in figure 10. The average dry condition temperature of catalyst was about $235\text{ }^{\circ}\text{C}$. The average temperature was about $215\text{ }^{\circ}\text{C}$ during wet condition ($t = 0.0\text{ s}$) and was recovered to about $235\text{ }^{\circ}\text{C}$ during relatively dry condition ($t = 1.5\text{ s}$). The next pulse and wet condition appears at $t = 2.0\text{ s}$, corresponding to

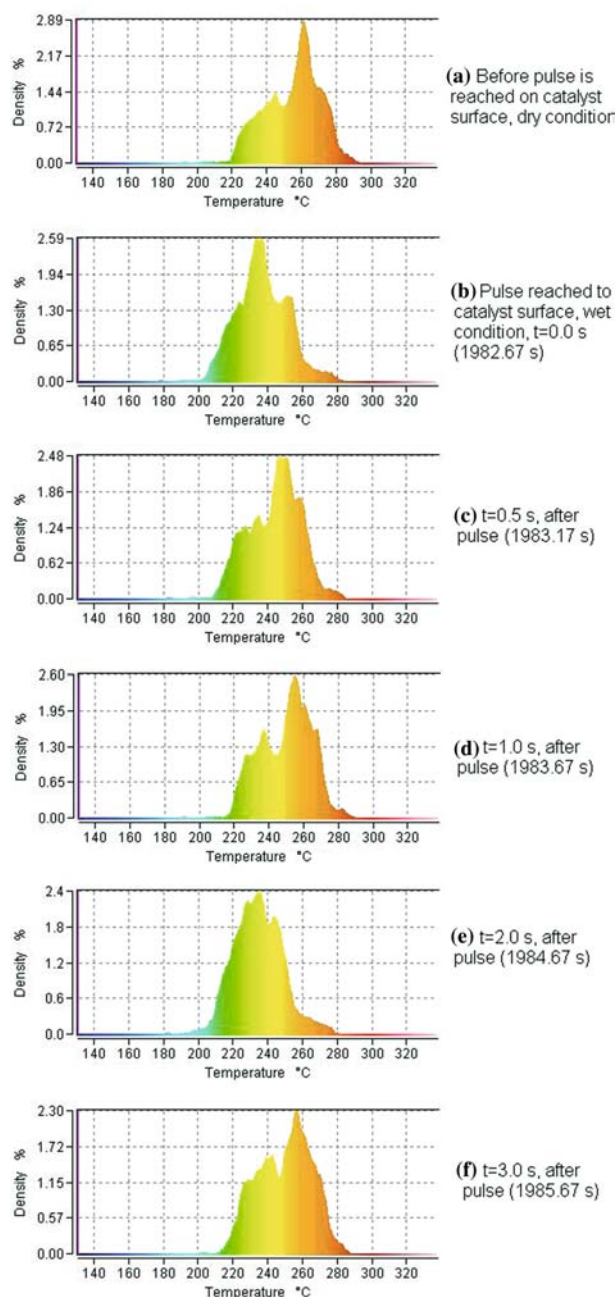


Figure 10. Histograms showing the chronology of temperature profile at 0.5 Hz, $T_w = 300\text{ }^{\circ}\text{C}$, steady state, time in brackets indicate accumulated time from the start of the reaction.

cyclic injection of reactant spray on catalyst surface. As at injection frequency of 0.5 Hz, the reactant may have been always present on the catalyst surface, increasing the pulse injection frequency to 0.5 Hz resulted in increase in hydrogen production rate. The hydrogen evolution rate observed at 0.5 Hz appears to be slightly fluctuating (figure 5) which may be because of possible enrichment of the catalyst surface by the reactant posing the resistance to easy evolution of hydrogen after the reaction. At the frequency of 1 Hz there is a liquid pool formation on the catalyst surface and accumulation of liquid reactant within reactor, therefore no reaction was

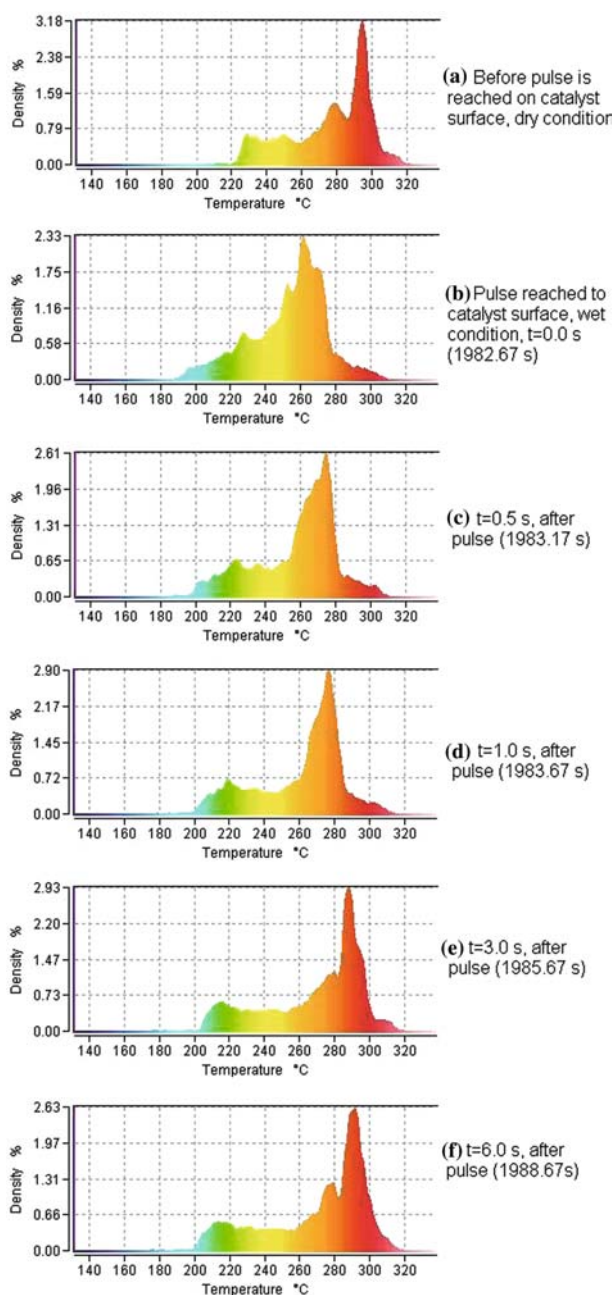


Figure 11. Histograms showing the chronology of temperature profile at 0.01 Hz, $T_w = 300\text{ }^{\circ}\text{C}$, steady state, time in brackets indicate accumulated time from the start of the reaction.

carried out at the frequency higher than 0.5 Hz. The trend observed at lower frequency of 0.01 Hz is similar to the 0.1 Hz with prolonged dry as shown in figure 11.

3.4. Selection of optimum operating conditions

The hydrogen production rate and the temperature difference between the dry and the wet conditions at varying operating frequencies were compared as shown in table 1. The temperature at dry conditions was decreased with increase in injection frequency. Whereas the hydrogen production rate both in terms of $\text{mmol g}_{\text{Pt}}^{-1} \text{ min}^{-1}$ and $\mu\text{mol min}^{-1}$ was observed to be increasing with injection frequency. The conversion efficiency was strongly dependent on average temperature of the catalyst surface. At injection frequency of 0.1 Hz, the difference between dry and wet condition was about $29\text{ }^{\circ}\text{C}$ at steady state, which was maximum as compared to other pulse-injection frequencies. Since the dehydrogenation of cyclohexane over Pt catalyst was observed to be highly selective, the energy consumption, which results in temperature decrease of catalyst, may be attributed to energy required for evaporation of cyclohexane and dehydrogenation only. The differences in temperature compared (in table 1) are after introduction of only one pulse at different frequencies. As the pulse width and fuel injection pressure is constant, the amount of cyclohexane delivered was the same in one pulse at different frequencies. Therefore, it may be considered that the more is the decrease in temperature more is the dehydrogenation occurring at that particular condition. This indicates that at the pulse-injection frequency of 0.1 Hz there may be the optimum conditions for cyclohexane dehydrogenation. Also the conversion efficiency was only marginally lowered as compare to 0.01 and 0.05 Hz. Although the hydrogen production rate was higher at 0.5 Hz it was not proportionate to the increase in feed rate, and also the conversion efficiency was dropped considerably. Therefore, based on the reaction data and thermal profile obtained by IR thermography indicates that the pulse injection frequency of 0.1 Hz is the optimum operating frequency. Increase in heating temperature may be required for increase in catalyst activity. A conductive support may be more

Table 1

Comparison of dehydrogenation of cyclohexane over 5 wt.% Pt/AC at heating temperature of $300\text{ }^{\circ}\text{C}$ at different operating conditions

Cyclohexane feed frequency (Hz)	Dry condition average temperature of catalyst surface ^a ($^{\circ}\text{C}$)	Difference in average temperature of catalyst surface at dry and wet condition ^b ($^{\circ}\text{C}$)	Hydrogen production ($\text{mmol g}_{\text{Pt}}^{-1} \text{ min}^{-1}$)	Hydrogen evolution rate ($\mu\text{mol min}^{-1}$)	Conversion efficiency (%)
0.01	276	23	21.57	613	38.8
0.05	277	25	51.91	1457	32.7
0.1	257	29	72.76	2070	29.5
0.5	235	20	97.87	2784	12.7

^aTemperature at steady state at about 33 min after start of reaction.

^bAverage difference occur by cooling due to introduction of one pulse-spray injection.

useful for obtaining uniform temperature profile and minimum difference between average temperature of catalyst and heating temperature.

4. Conclusions

The dehydrogenation of cyclohexane was carried out on Pt/CFF-1500s catalyst using pulse-spray reactor. The rate of hydrogen evolution observed to be increasing with increase in reaction temperature (250–300 °C) or increase in cyclohexane feed rate frequency (0.01–0.5 Hz) particularly at heating temperature of 300 °C. Therefore, it may be concluded that it is possible to optimize the spray-pulsed reactor operating conditions for heating temperature and cyclohexane dose to obtain higher hydrogen production rates.

The temperature profiles of catalyst surface studied using IR camera during the pulse -spray injection of cyclohexane reveals the actual temperature of catalyst surface is much lower than that of the set heating temperature. It is clear from the decreased surface temperature observed that under the liquid pool conditions the catalyst efficiency may be lowered. Whereas when dry condition is generated by varying the injection frequency the catalyst temperature is increased, this may help improving catalyst activity. Hence use of alternate wet and dry conditions by spray-pulse injection helps in achieving better catalyst activity. The lowering of the catalyst temperature in case of endothermic reactions may be monitored by IR thermography and it may indicate the activity of catalyst. Therefore, the use of IR thermography has been demonstrated for strongly endothermic reactions, this may be useful in timely research in the field of IR thermography for screening the catalysts for even strong endothermic reactions. Better heat flux and the uniform temperatures on catalyst surface may be obtained using conductive support such as metallic support.

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