Experimental study of *n*-heptane ignition delay with carbon dioxide addition in a rapid compression machine under low-temperature conditions

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The ignition delay of *n*-heptane homogeneous charge compression ignition (HCCI) combustion under high levels of carbon dioxide addition was quantitatively measured at elevated pressure from low to intermediate temperatures in a rapid compression machine. The experiments were conducted in the compressed temperature range 613–750 K. Both the compression ratio and fuel/air equivalence ratio were varied to investigate their effects on the ignition delay of *n*-heptane. Carbon dioxide was subsequently added to study the influence of the carbon dioxide level on the ignition delay of *n*-heptane under low-temperature conditions. It was found that carbon dioxide had different effects on the two -stages of ignition delay of *n*-heptane under low-temperature conditions: the concentration of carbon dioxide had little effect on the first-stage ignition time; a certain concentration of carbon dioxide accelerated the first-stage ignition but had a significantly larger impact on the second-stage ignition delay, thus increasing the overall ignition delay time. The results also showed that the first-stage ignition delay of *n*-heptane is only a function of temperature under low-temperature conditions. The mass of *n*-heptane in the combustible mixture, the equivalence ratio, and the pressure at the top dead center had little effect on the first-stage ignition time of *n*-heptane.

rapid compression machine, *n*-heptane, ignition delay, low-temperature condition, carbon dioxide

Environmental pressures and the energy crisis have led to increasing attention being paid to the improvement of combustion efficiency and the reduction of internal combustion (IC) engine emissions. To fundamentally improve emission levels and combustion efficiency, it is necessary to have an understanding of the combustion chemical reaction dynamics and combustion methods of IC engines. A conventional diesel engine has the advantage of a higher combustion efficiency than that of a gasoline engine, but it cannot simultaneously reduce particulate matter (PM) and NO$_x$ because of the partially rich or lean combustion in the combustion chamber. Homogeneous charge compression ignition (HCCI) has become the focus of research because of its excellent efficiency and low emission levels [1–3]. To control HCCI

combustion, it is essential to have sufficient understanding of its ignition time and combustion speed.

Processes such as fuel evaporation, diffusion, combustion, and heat transfer are very complicated in IC engines. In addition, differences exist between each working cycle, making data measurement and control of parameters between combustion cycles very difficult. Moreover, IC engines have complex structures, so it is very difficult to install test equipment or visualization windows. To solve these problems, a variety of experimental devices such as shock tubes [4–6], rapid compression machines (RCMs) [7,8], and jet-stirred flow reactors [9,10] have been developed by researchers to simulate the working processes of real engines. Among these devices, the RCM has advantages that make it more suitable for research into auto-ignition of hydrocarbons in the low-to-intermediate temperature range [11–13];

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for instance, controlling boundary and initial conditions and installing visualization windows are very easy. It also has little variation between cycles and the working process is similar to that of a real engine.

n-Heptane is widely accepted as a surrogate for complex commercial diesel fuel. For this reason, studies of the auto-ignition of *n*-heptane in RCMs have been carried out for many years [14–20]. The results show that *n*-heptane, like other long-chain alkanes, has a typical two-stage ignition process consisting of a low-temperature stage followed by a high-temperature stage. Griffiths et al. [21] studied the influence of compressed gas temperature on the ignition delay and found that at low compressed gas temperatures, two-stage ignition was a clear feature of *n*-heptane. Single-stage ignition was apparent at higher compressed gas temperatures. Recently, Tanaka et al. [22] studied the effects of fuel structure and additives on the HCCI combustion of pure hydrocarbons and mixtures under specified conditions, and found that fuels which contained the structure $-CH_2-CH_2-CH_2-$ showed two-stage ignition, and the ignition delay depended strongly on the first-stage reactivity. The results also showed that the ignition delay, total pressure increase, and burn rate can be independently controlled by changing the *n*-heptane: oxygen molar ratio, additives, the initial temperature, and the octane number, providing new choices for controlling HCCI combustion. Silke et al. [23] studied nine isomers of heptane using an RCM in the temperature range 640–960 K, and the selected compressed pressures were 1.0, 1.5, and 2.0 MPa. The results showed that both reactivity and burn rate decreased as the degree of branching of the heptane isomers increased; this correlated with the research octane number.

Because of the constraints associated with the ignition timing control and operating regime of HCCI combustion, coupled with the complexity of making a homogenous mixture of diesel fuel, the development of HCCI combustion with diesel fuel has been severely hampered. Recently, a new combustion model based on diesel-type fuel, which is more efficient and clean, has been proposed internationally –low-temperature combustion (LTC). The basic strategy of this combustion mode is to use a high level of exhaust gas recirculation (higher than 60% or oxygen content less than 10%) to prolong the chemistry timescale, and, at the same time, the physical time for preparation of a homogenous mixture is shortened by using high-pressure and multiple-stage injection and intensive in-cylinder turbulence. Using the above methods, control of ignition and combustion was achieved by modifying the local and overall mixing times as well as the chemistry timescale [24]. Hydrocarbon ignition under low-temperature conditions has attracted much attention because of its potential to reduce NO_x and soot emissions simultaneously [25,26]. Yao et al. [26] have discussed the necessity and feasibility of LTC in detail, and pointed out that LTC has the potential to extend the HCCI operating range. Two-stage ignition (low-temperature and high-tem-

perature reactions) usually occurs during the compression process for diesel-type fuels, and the low-temperature reaction timing and reaction rate play a decisive role in the combustion timing of HCCI combustion [27,28]. However, the majority of the studies mentioned focused on the ignition delay time at a fixed compression ratio (ε) and not enough data have been obtained on comparisons of ignition delays under different compression ratios with different equivalence ratios (φ) in the low-temperature range. Moreover, detailed experiments on *n*-heptane under high exhaust gas recirculation conditions have not been conducted using an RCM and, to the best of the authors' knowledge, experiments conducted with RCMs to study low-temperature ignition characteristics of *n*-heptane achieved using high levels of carbon dioxide have not been published before.

Based on the above background, the main purpose of this paper is to study the influence of compression ratio on the ignition time of *n*-heptane with different equivalence ratios and different concentrations of carbon dioxide in the low-temperature range, and to investigate the factors that affect the ignition characteristics of *n*-heptane under both low- and critical-temperature conditions. The study of ignition characteristics of *n*-heptane in the low-temperature range can provide a reference for HCCI combustion under LTC conditions in conventional engines. This paper first describes the specifications of the RCM used in this study. This is followed by a presentation and discussion of the experimental results.

1 Experimental

1.1 Rapid-compression machine

An RCM is a simulated laboratory device driven by an external force to make a piston move forward rapidly. The combustible mixture in the combustion chamber is compressed by the rapidly moving piston, which is locked once it reaches the top dead center (TDC). This causes the combustible mixture to reach a high pressure and temperature, and then it ignites spontaneously. The experiments in this study were conducted with an RCM which was designed and developed by Shanghai Jiao Tong University. The experimental system is shown in Figure 1.

The RCM is driven by high-pressure gas from an impact pneumatic cylinder (IPC). The IPC has two separate cylinders: a back cylinder (also named the storage cylinder) and a front cylinder. There is a convergent nozzle between the two cylinders. The back cylinder converges into a spout, which leads into the front cylinder. The area ratio of the back cylinder to the end of the spout is 9∶1. Hence, when high-pressure gas is stored in the back cylinder, the gas in the front cylinder has a much lower pressure, maintaining the IPC piston at its initial position. When the RCM starts running, gas in the front cylinder can be released rapidly because of its low pressure and small volume. This results
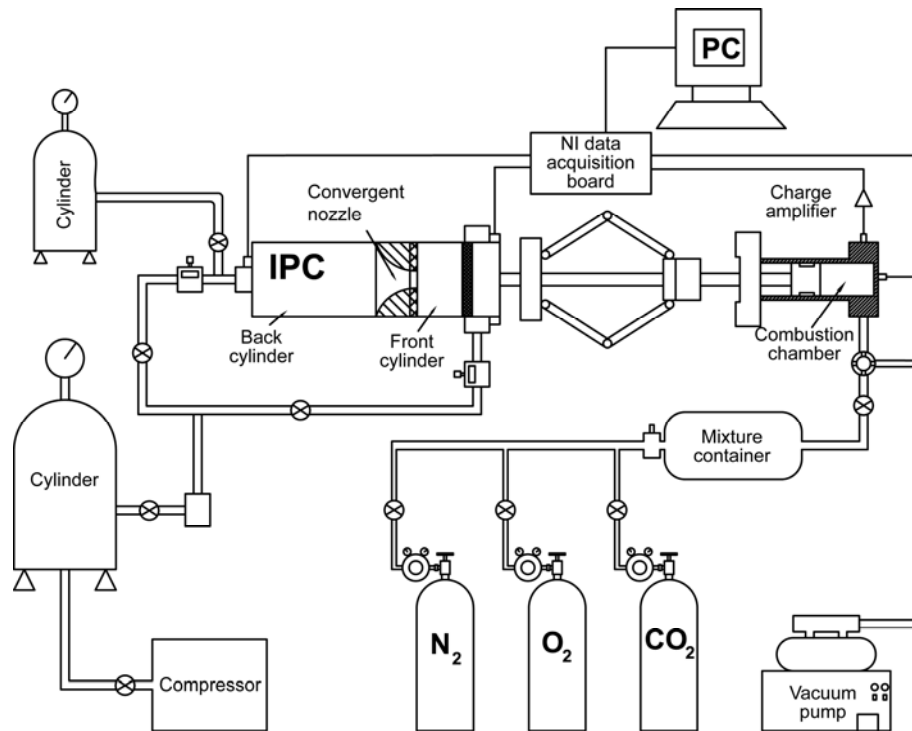
Figure 1   Schematic diagram of experimental system.

in a forward force from the high-pressure gas in the back cylinder, which pushes the IPC piston forward. Once the IPC piston moves forward, the force acting on it increases eight-fold, driving the IPC piston forward rapidly without any resistance from the front cylinder gas. The RCM piston speed can be easily changed by adjusting the pressure of the IPC back cylinder. A brake arm and a block are used to stop the RCM piston when it reaches the TDC. The reactor cylinder has a bore of 50 mm and the stroke is 218.2 mm. The compression ratio can be changed in the range 6.5–25 by adjusting the length of the connecting rod between the RCM piston and the IPC. Typical pressure-time histories with and without ignition are shown in Figure 2.

A creviced piston is used to maintain a homogeneous temperature field for as long as possible after the piston

reaches the TDC. The creviced piston leaves gaps between the piston head and the cylinder, so the boundary vortex caused by the movement of the piston can be squeezed into the clearance volume around the piston skirt, thus reducing the influence of the boundary vortex on the internal flow field. The piston is shown in Figure 3. The design of this piston is based on studies conducted by researchers at Massachusetts Institute of Technology, Cambridge, MA, USA [29,30]. In their studies, they found that the size of the crevice along the cylinder periphery was very important. A creviced piston head of an appropriate size can make the temperature distribution significantly more homogeneous and can increase the extent of the core region [31,32]. The "adiabatic core" hypothesis can be used more accurately when the temperature field is homogeneous inside the
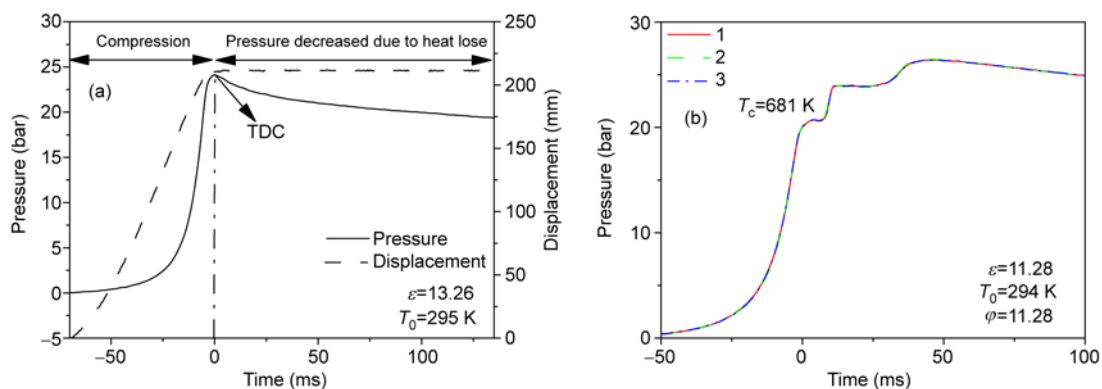

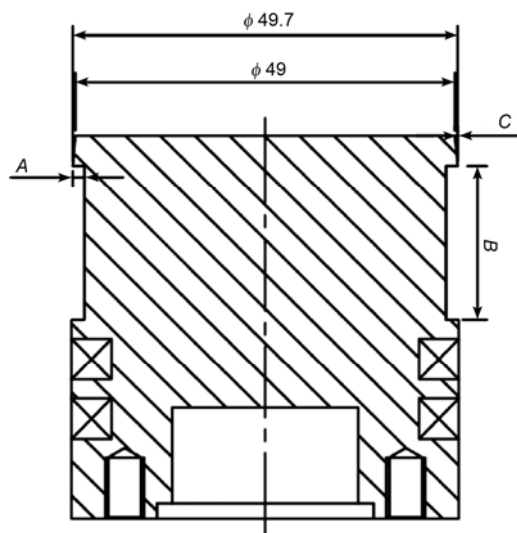**Figure 2**   Typical pressure curves of the RCM.

Figure 3   Creviced piston.

Figure 4   Definitions of ignition delays.

combustion chamber. Thus, the temperature at the TDC can be calculated using the following equation:

$$\int_{T_0}^{T_c} \frac{\gamma(T)}{\gamma(T)-1} \frac{\mathrm{d}T}{T} = \ln\left(\frac{P_c}{P_0}\right), \tag{1}$$

where T_0 and P_0 are the initial temperature and pressure, T_c and P_c are the compressed gas temperature and pressure at the TDC, and $\gamma(T)$ is the ratio of the specific heats of mixture, which is a function of temperature.

1.2   Definitions of ignition delay and burn rate

Ignition delay is an important parameter in research on ignition of hydrocarbons. The definitions of ignition delay used in this study are illustrated in Figure 4. $P'(t)$ is the numerical differentiation of pressure and time data ($\mathrm{d}P/\mathrm{d}t$), and $\mathrm{d}P_{max}$ is the peak value of $P'(t)$. Ignition delay is defined as the time between the end of compression and achieving the maximum rate of pressure rise during ignition. Specifically, τ_1 is the first-stage ignition delay, which is the time interval between the end of compression and the onset of first-stage ignition, and τ_2 is the time between the first-stage ignition and the second-stage ignition. The overall ignition delay, τ, is the sum of τ_1 and τ_2; t_1 and t_2 represent the time of onset of the first-stage and second-stage ignition, respectively. In this study, the burn rate is defined as the maximum rate of pressure rise, $\mathrm{d}P_{max}$.

1.3   Preparation of mixture

The combustible mixture was prepared in a stainless-steel cylinder. The purities of the gases used were as follows: oxygen, 99.9%; nitrogen, 99.9%; and carbon dioxide, 99.9%. The purity of *n*-heptane was >97.3% (research grade). Before the mixture preparation, the stainless-steel cylinder was
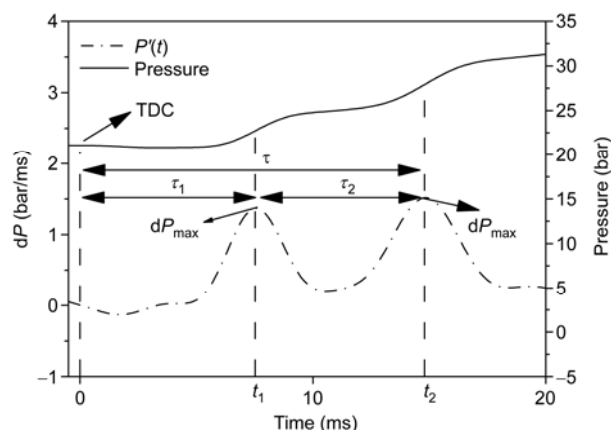
evacuated using a vacuum pump, and the initial pressure was less than 40 Pa. The proportions of various gaseous components were determined manometrically under ambient temperature and atmospheric pressure conditions, and allowed to homogenize for an appropriate time.

1.4   Experimental conditions and uncertainties

The compression ratio was changed by adjusting the length of the connecting rod, and the compression ratios chosen in this study were 9.52, 10.48, 11.28, 12.93, 14.45, and 16.13. The initial mixture temperature was 295 K and was measured using a K-type thermocouple, and the initial mixture pressure was ambient atmospheric pressure. Pressure–time data were measured using a Kistler 6125B transducer with a 5015 charge amplifier. The pressure and displacement data were recorded synchronously using an NI PCI-6132 data-acquisition board. The experiments were repeated three to five times under each set of conditions, and one representative pressure curve close to the mean was chosen to determine the ignition delay times. Figure 2(b) shows the results from three experiments conducted under the same condition. It can be seen that the reproducibility of this RCM is very good and the error in the maximum pressure under identical conditions was less than 1%.

2   Results and discussion

2.1   Two-stage ignition of *n*-heptane

Figure 5 shows the pressure traces and burn rate of *n*-heptane at $\varepsilon=9.52$, $P_c\sim17.5$ bar, and the T_c was 644 K. Figure 5(a) shows that the first-stage ignition delay, τ_1, increased, but the second-stage ignition delay, τ_2, decreased as φ increased. This was because the overall heat capacity of the fuel and "air" mixture changed when φ increased, thus prolonging the first-stage induction time and increasing the ignition delay. The second-stage ignition delay decreased because the number of radicals created during the low-temperature reaction
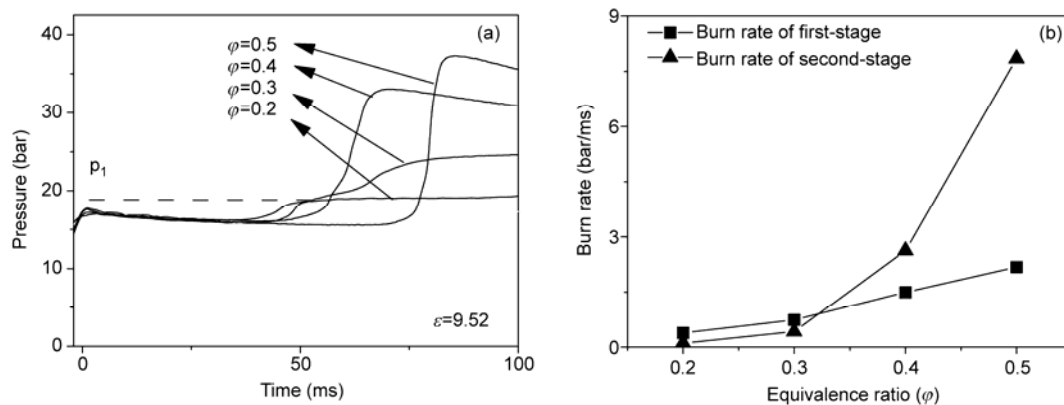
Figure 5   Effect of equivalence ratio on ignition delay and burn rate.

increased with φ and accelerated the second-stage reaction rate. In this temperature range, low-temperature kinetics dominated the oxidation process, and fuel concentration had a large influence on chain branching. When the low-temperature reaction was completed, the pressures in the combustion chamber under different φ conditions were almost the same. Figure 5(b) shows that the burn rates of both stages increased with φ. The influence of φ on the second-stage burn rate was more significant. The burn rate of the second-stage reaction was related to the number of radicals created in the first-stage. Increasing numbers of radicals were created during the first-stage reaction as φ increased, making the second-stage reaction more rapid and the ignition delay shorter.

Figure 6 shows the variations in ignition starting time with T_c. The φ values were 0.2 and 0.4. When the compression ratio increased, both the temperature and pressure increased, resulting in the first-stage ignition beginning before the TDC under certain conditions. The ignition times of both stages advanced with increasing T_c under the different φ conditions. For a given gas temperature, the number of radicals created during the low-temperature reaction increased
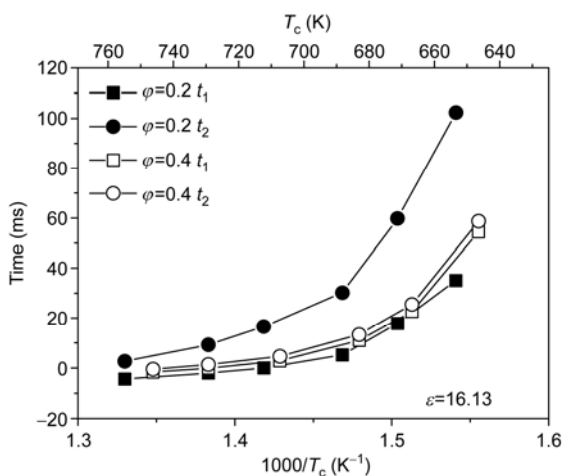
with increasing φ and advanced the second-stage ignition time, thus shortening the second-stage ignition delay, τ_2. However, the influence of φ on the first-stage ignition time under the same temperature conditions was not significant.

2.2   Influence of carbon dioxide on two-stage ignition

In this part, the amount of nitrogen in the "air" was kept constant, while the amount of oxygen was reduced to increase the amount of carbon dioxide. Two sets of experiments were conducted. In each set of experiments, the mass of n-heptane in the mixture was kept constant as the volume ratio of carbon dioxide in the "air" was progressively increased to 3%, 6%, 9%, and 12%. The masses of n-heptane in the stainless-steel cylinder in each set of experiments were 7.19 and 12.23 g, respectively, which was the same as in the mixtures whose φ values were 0.3 and 0.5, respectively, in the earlier experiments. Figure 7 shows the influence of carbon dioxide on ignition time with a fixed mass of n-heptane in the mixture. Carbon dioxide had a big influence on the second-stage ignition time. Carbon dioxide did not change the generation time of the major products of the low-temperature reaction, such as CH_2O and H_2O_2, but it prolonged the induction time before the high-temperature reaction. The consumption rate of activated radicals was reduced [28]. Adding carbon dioxide changed the heat capacity of the combustible mixture, and the compressed gas temperature reached 621 K with 12% carbon dioxide in the mixture at ε =9.52. Under these conditions, only a single ignition stage was observed. However, when the amount of n-heptane was increased, two-stage ignition occurred, even when T_c was reduced to 613 K.

Figure 8 shows the relationship between temperature and ignition delay for n-heptane. Figure 8(b)–(d) are Arrhenius-type plots. The equivalence ratio changed as the concentration of carbon dioxide changed with a fixed mass of n-heptane in the mixture. From Figure 8(a) and (b), it can be seen that before the negative temperature coefficient region, the first-stage ignition delay for n-heptane was only a function of temperature. Factors such as the mass of n-heptane

Figure 6   Influence of temperature on ignition time under different equivalence ratio conditions.
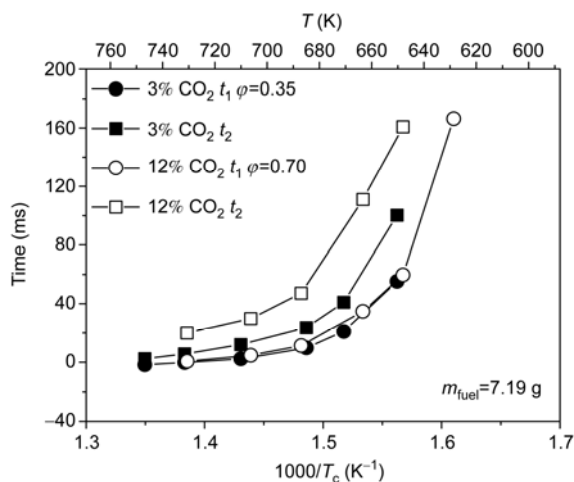
Figure 7   Influence of carbon dioxide on ignition time.

in the combustible mixtures, the equivalence ratio, the pressure at the TDC, and the concentration of carbon dioxide had little effect on the first-stage ignition time, and the first-stage ignition delay complied with the linear relationship under Arrhenius-type plot (the reciprocal of temperature and ignition delay have a linear relationship). The

compressed temperature is the dominant factor influencing the starting time of low-temperature reactions [33]. Figure 8(c) and (d) show the relationship between temperature and second-stage ignition delay and overall ignition delay, respectively. Carbon dioxide had a bigger influence on the second-stage ignition delay of mixtures containing less *n*-heptane. The second-stage and overall ignition delays increased as the concentration of carbon dioxide increased, with the same mass of *n*-heptane in the mixtures. When the level of carbon dioxide was the same, the second-stage and overall ignition delays became longer as the amount of *n*-heptane in the mixtures decreased. This meant that a lower equivalence ratio would lead to a longer ignition delay at the same carbon dioxide level, which is consistent with the results for mixtures with no carbon dioxide. Although the carbon dioxide had little influence on the first-stage ignition delay, it had a greater impact on the second-stage ignition delay. The second-stage and overall ignition delays were no longer a single function of temperature, but for a given experimental condition, they have a linear relationship with the reciprocal of temperature under Arrhenius-type plot.

Figure 9 shows the influence of carbon dioxide on the two-stage burn rate of *n*-heptane for a fixed mass of *n*-heptane in
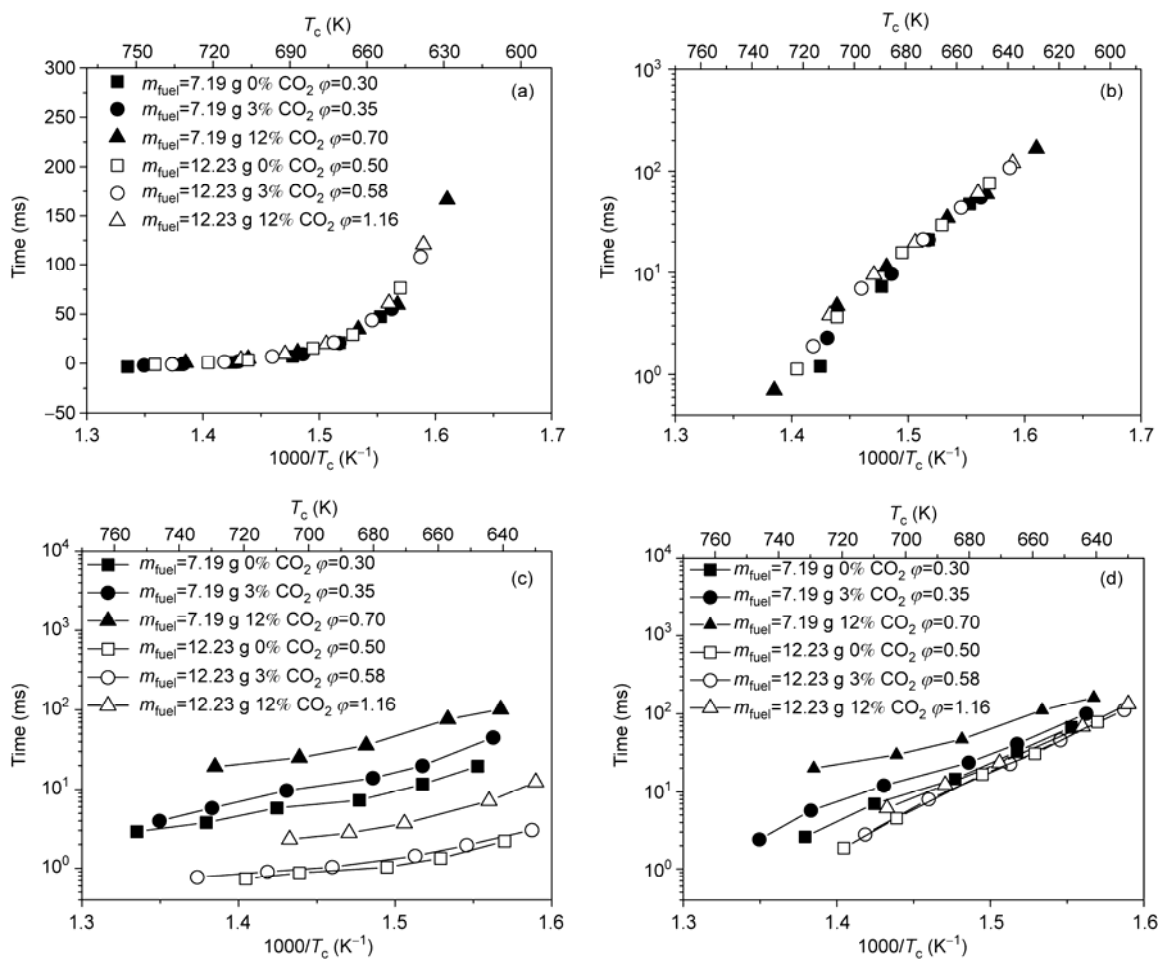

Figure 8   Ignition delay versus compressed gas temperature with different carbon dioxide concentrations.
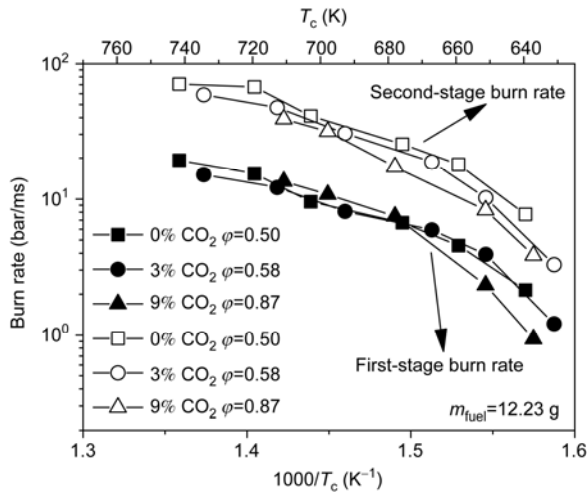
Figure 9 Influence of carbon dioxide on two-stage burn rate. Solid symbol indicates first-stage burn rates, and hollow symbol indicates second-stage burn rates.

the mixture. It can be seen that the burn rates of both stages decreased as the temperature decreased. The influence of carbon dioxide on the first-stage burn rate was not very significant, but the second-stage burn rate decreased as the amount of carbon dioxide increased. The influence of carbon dioxide on the burn rate was consistent with the effects on ignition delay; as the concentration of carbon dioxide increased, the amount of oxygen in the mixture decreased, resulting in a reduction in the number of radicals created in the low-temperature reaction, thereby prolonging the second-stage ignition delay and reducing the burn rate. The burn rates of both stages decreased with decreasing amounts of *n*-heptane in the mixtures.

Figure 10 shows the influence of carbon dioxide on the two-stage ignition delay with $\varphi=0.35$ and 0.7. When φ was kept constant, adding carbon dioxide increased the overall ignition delay, and the influence of carbon dioxide on the second-stage ignition delay was significant. From Figure 10(a) it can be seen that the first-stage ignition delay of a combus-

tible mixture which contained 3% carbon dioxide was shorter than that of the mixture without carbon dioxide, indicating that when a mixture contained a certain amount of carbon dioxide, the low-temperature reaction was accelerated. The low-temperature reaction starting time was related to the thermal properties of the mixture. Heat transfer during and after compression was affected when carbon dioxide was added to the mixture, leading to the above- mentioned results. Würmel et al. [34] explained how a diluent gas affected the ignition delay in an RCM. In this study, it can be seen that although carbon dioxide could decelerate the overall ignition of *n*-heptane, it could accelerate the first-stage ignition under certain experimental conditions. From Figure 10(b), it can be seen that the first-stage ignition delay of the mixture which contained 12% carbon dioxide was longer than that of the mixture which contained 6% carbon dioxide, with $\varphi=0.7$. This was contrary to the results shown in Figure 10(a), and meant that with a fixed equivalence ratio, an increase in the carbon dioxide level would increase the first-stage ignition delay. Further experiments are required to verify the influence of carbon dioxide on the first-stage ignition delay under certain equivalence ratios and carbon dioxide levels.

3   Conclusions

This paper has introduced the characteristics of a RCM based on a high-pressure gas from an IPC. A series of experiments were conducted to investigate the two-stage ignition characteristics of *n*-heptane with high levels of carbon dioxide in the low-temperature region. The following conclusions can be drawn from the above analysis:

(1) The starting time of the low-temperature reaction of *n*-heptane does not change much for different carbon dioxide levels and is only a function of temperature. Factors such as the mass of *n*-heptane in the combustible mixture, the equivalence ratio, and the pressure at the TDC also had little effect on the starting time. However, these factors have
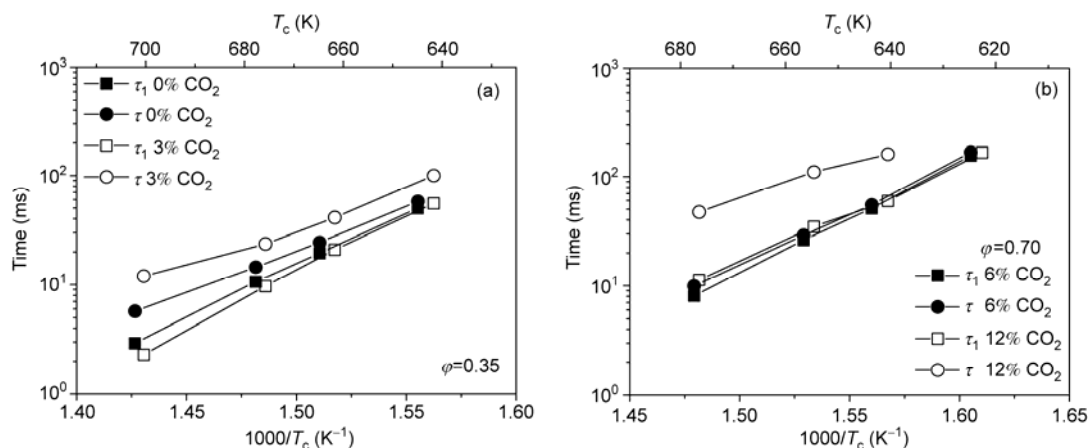

Figure 10 Influence of carbon dioxide on ignition delay under fixed equivalence ratio conditions.

different influences on the second-stage ignition delay. For a given experimental condition, both first-stage and second-stage ignition delays have a linear relationship with the reciprocal of temperature under Arrhenius-type plot.

(2) When the compressed gas temperature was reduced to a critical value under conditions with high levels of carbon dioxide, *n*-heptane only showed one-stage ignition. This critical value was reduced even further when the mass of *n*-heptane in the mixture was increased.

(3) The influence of carbon dioxide concentration on the first-stage burn rate is not significant, but an increase in the carbon dioxide concentration causes a decrease in the second-stage burn rate under the same temperature conditions. Carbon dioxide has the same effects on the burn rate and ignition delay.

(4) The first-stage reaction of *n*-heptane is promoted when the mixture contains a certain amount of carbon dioxide. Dilute gases have different effects on the ignition delay. Due attention should therefore be paid when using an RCM to study the ignition characteristics of hydrocarbons with a dilute gas in the combustible mixture.

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