

To Study on Ignition Characteristics of Syngas Mixtures by Shock Tube

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ABSTRACT

Ignition delay of undiluted syngas mixtures with different compositions of H₂, CO, CO₂, N₂ and air was measured using a shock tube. Experiments were conducted under various conditions of pressure of 0.2 and 1.0 MPa, temperature from 757 to 1280 K, and equivalence ratio of 0.3 and 1.0. The testing data set was analyzed based on methods including: Arrhenius-type correlation (to assess the effect of pressure, temperature, equivalence ratio, and fuel composition on ignition characteristics), use Davis's mechanism (to calculate ignition delay). The obtained results using Arrhenius-type correlation and Davis's mechanism showed a far difference from experimental values. A detailed analysis was conducted to evaluate the influence of perturbation from shock tube experiments on chemical induction time of the syngas. The ignition delays, considered the effect of non-ideal conditions, are shorter than ones which predicted with ideal conditions. Measured pressure curves were also used to further simulate, the achieved ignition delays have approached to experimental values. Additionally, the most important reactions in combustion process were pointed out and discussed through sensitivity analyses. The research results may contribute to improvement of the current chemical kinetics and mechanisms and be applicable in research and development of equipments operating with syngas fuels.

KEYWORDS

syngas, ignition delay, shock tube, chemical kinetics

1. INTRODUCTION

Since emission levels emitted from industrial equipments have been strengthened, it is necessary to use clean fuel sources and advanced plant designs. As one of the clean fuels, syngas are supposed to play an important role in future energy field [1, 2]. In the future, syngas produced from coal is still a major fuel source for the electric power generations with new technologies to reduce CO₂ emissions and increase energy efficiency [2]. Syngas is also a clean fuel for internal combustion engines, fuel cells, and boilers [1, 2, 3]. Recently, combustion characteristics and chemical kinetics of syngas were studied experimentally under practical conditions relevant to current industrial turbine technologies [4, 5, 6, 7], however, the achievements were very limited. Data studied in the laboratories can provide a comprehensive understanding of the reaction kinetics and other fundamental combustion characteristics of syngas mixtures. Hence, studying of reaction kinetics and combustion characteristics of syngas has a realistic meaningful value from fundamental and practical perspectives [5, 8].

Syngas is a mixture of H₂ and CO. In practice, however, it usually contains some other sub-products of complicated

gasification processes used to convert a solid fuel to syngas. The sub-products can be methane (CH_4), small amounts of heavier hydrocarbons such as C_2H_6 , C_2H_4 , non-combustible constituents namely CO_2 , H_2O , and N_2 (when using air as an oxidant), and several contaminants including small carbonaceous particles, ash, tars, and oils [1]. The components contained in syngas and their fractions vary significantly depending on fuel feedstock, gasification technology and production conditions. Therefore, complexity of syngas compositions can cause difficulty in designing combustion plants as well as in selecting experimental schemes and establishing simulation models.

Experimental investigation of shock tube ignition delay and combustion characteristics of syngas contributes to understanding chemical kinetics and improving capacities of predictive models. The knowledge is useful for designing advanced syngas turbines and combustors applied IGCC (integrated gasification combined cycle) technology [2].

Auto-ignition characteristics of H_2 mixtures at various temperature and pressure conditions were indicated in the work of Mittal et al. [8]. Recently, several detailed kinetic mechanisms of H_2/O_2 system have been developed and validated with experimental data carefully, some of these mechanisms were optimized rely on reported works which relevant to rate constant of the reactions [9, 10].

Ignition characteristics of H_2/O_2 and $\text{H}_2/\text{CO}/\text{O}_2$ mixtures were studied by Mittal et al. [8] in a rapid compression machine (RCM) at pressures of 1.5-5.0 MPa, temperature of 950-1100 K and equivalence ratios of 0.36-1.6. Their study showed that the replacement of small amount of H_2 by CO in the mixture could largely increase its ignition delays. Using available kinetic mechanisms of $\text{H}_2/\text{CO}/\text{O}_2$ still gave large discrepancy to the experimental data at elevated pressures.

Das et al. [11] studied the ignition delays of moist H_2/O_2 mixtures with 0%, 10%, and 40% molar fractions of vapor at temperature of 907-1048 K, pressures of 1.0, 3.0 and 7.0 MPa using a RCM. Their results showed that at pressures of 3.0 MPa and 7.0 MPa, vapor addition of 10% and 40% could promote auto-ignition of mixtures and decrease ignition delay. However, 10% vapor addition would inhibit the reactivity and increase ignition delay at pressure of 1.0 MPa.

Kéromnès et al. [12] investigated the ignition delays of three fuel compositions (100% H_2 , 85% $\text{H}_2/15\%$ CO and 50% $\text{H}_2/50\%$ CO) at pressures of 0.8-3.2 MPa, temperature of 900-1050 K and equivalence ratios of 0.35 and 0.5 using a RCM. A good agreement between experimental and predicted ignition delays was achieved using a modified model. Experimental ignition delays were increased with the addition of CO in the testing mixtures. The inhibited effect became significant when CO concentration is larger than 60%.

The work of Walton et al. [5] focused on the ignition of $\text{H}_2/\text{CO}/\text{CO}_2/\text{N}_2/\text{O}_2$ mixtures with H_2 to CO ratios of 0.25-4.0 at pressures of 0.71-2.64 MPa, temperature of 855-1051 K, and equivalence ratios of 0.1-1.0 using a RCM. The measured values are one order of magnitude lower than the predicted ones at low temperature.

The ignition delays of $\text{H}_2/\text{air}/\text{steam}$ mixtures at temperature of 900-1350 K, pressures of 0.3-1.7 MPa were measured by Wang et al. [13] in a heated shock tube. Their study showed that the ignition delay strongly depends on steam content and temperature, however, the effect of pressure is less significant. The measured ignition delays are lower than model predictions in low temperature range.

Petersen et al. [14] measured the ignition delays of $\text{H}_2/\text{CO}/\text{CO}_2/\text{O}_2/\text{N}_2$ mixtures using a shock tube (at pressure of 2.0 MPa, temperature of 943-1148 K) and CO/H_2 mixture using a flow reactor (at pressure of 0.5 MPa, temperature of 760-783 K). Their study results showed that at temperature of 700-800 K, the measured ignition delays were two-to-three orders of magnitude lower than ones predicted five available mechanisms. They also suggested that reasonable adjustments in reaction constants did not diminish the disagreement.

The measured ignition delays from various facilities were summarized by Dryer and Macos [4]. They reported that the effect of mixture non-homogeneity on chemical process was significant at elevated pressure and low temperature. However, the improved kinetic parameters in the H_2/CO system only influence the predicted ignition delays by factor, not by order of magnitude.

In each reviewed literature, the testing conditions are not the same, for example, range (pressure, temperature and equivalence ratio), mixture (composition and content) and facilities (shock tube, RCM, reactor). In the works, experimental ignition delays of syngas measured by shock tube at moderate-low temperature and elevated pressure conditions are inadequate. Thus, this study aimed to investigate ignition characteristics of syngas in this region. The obtained experimental results were analyzed and compared with predicted ones. The results also contribute to improve the available mechanisms and understand chemical kinetics deeply.

2. EXPERIMENTS

2.1. Shock Tube Setup

All experiments were carried out in a high pressure shock tube shown in [Figure 1](#). The device is a 115 mm inner diameter tube, is divided into a 2.0 m driver section and a 7.3 m driven section by a double diaphragm. Mixtures of high-purity He and N_2 were used as driver gases to create

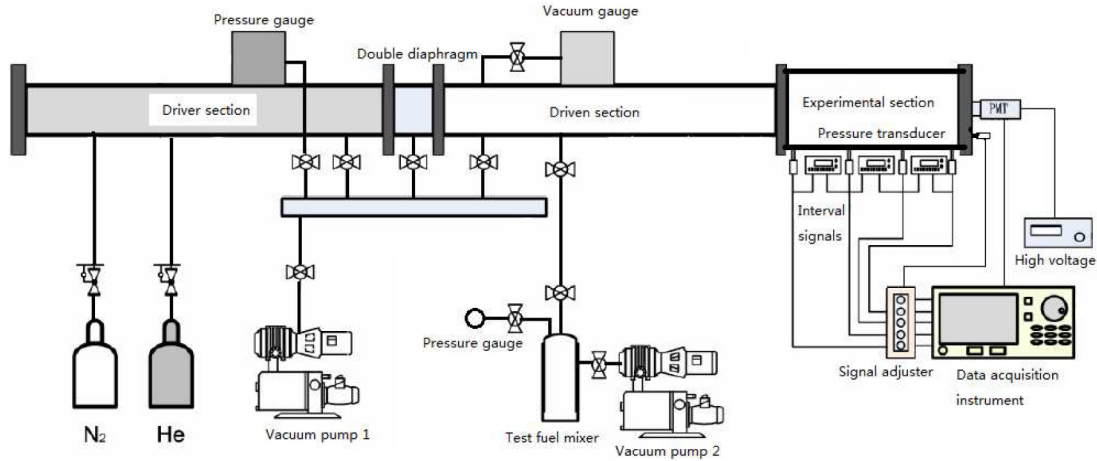


Figure 1. Schematics of shock tube

“tailored” conditions to obtain a long testing time at low temperature. Before each experiment, the shock tube was evacuated to pressure below 10^{-4} Torr. Velocity of shock wave was measured by four fast-response pressure transducers and three time counters located on three 30 cm intervals.

Temperature values behind the reflected shock wave were calculated using the chemical equilibrium software GASEQ [15]. Typical uncertainty in these temperature calculations is ± 20 K.

Reactant mixtures used in this study were prepared in a 128 litre volume stainless-steel tank according to Dalton's law of partial pressure and were kept over 12 hours to ensure a sufficient mixing. Before preparing reactant mixtures, the mixed tank was evacuated to pressure below 10^{-4} Torr. All testing gases have high-purity: H_2 -99.995%, CO -99.99%, CO_2 -99.99%, N_2 -99.995%, O_2 - 99.99%, Ar -99.995%.

Reliability of this facility has been validated in previous works for various fuel mixtures and at different experimental conditions [16, 17, 18].

2.2. Fuels Used and Testing Conditions

The detailed compositions and fractions (by volume) of testing mixtures are given in Table 1.

Table 1. Compositions and fractions of testing syngas

Mixtures	H_2	CO	CO_2	N_2
1	0.333	0.667	0	0
2	0.346	0.353	0.301	0
3	0.114	0.311	0	0.575

The testing regimes were selected considerably the suitability with practical operation conditions of gas turbines [4]. The conditions relevant to gas turbine mixing systems are low temperature, high pressure and lean mixture: $T < 1000$ K, $P \geq$

1.0 MPa and $\phi \leq 1$. Experiments were carried out for three mixtures of syngas (Table 1) in ranges of temperature from 757 K to 1280 K, pressures of 0.2 MPa and 1.0 MPa, and equivalence ratios of 0.3 and 1.0.

3. RESULTS AND DISCUSSION

3.1. Ignition Delay Calculation Method

Calculated ignition delay and kinetic analysis of the syngas were carried out using CHEMKIN software [19] with extensively incorporating SENKIN package [20]. To model the ignition event behind the reflected shock waves, numerical simulation was conducted using the software. Ignition models and computational methods for chemical kinetic calculations were used to compare the ignition delays obtained by shock tube experiments with theoretical predictions based on detailed chemical kinetics. The governing energy and mass conservation equations in the constant-volume model are shown as follow,

$$\frac{dT}{dt} = -\frac{1}{\rho C_v} \sum_{k=1}^K e_k \dot{\omega}_k W_k \quad (1)$$

$$\frac{dY_k}{dt} = \frac{\dot{\omega}_k W_k}{\rho}, \quad k = 1, \dots, K \quad (2)$$

where: T is the temperature, Y_k is the mass fraction of species k , ρ is the density, \bar{C}_v is the mean specific heat at constant volume, e_k is the specific internal energy of species k , $\dot{\omega}_k$ is the molar production rate of species k , and W_k is the molecular weight of species k . The independent variable t represents the time.

3.2. Determination of Experimental Ignition Delay

Ignition is determined by the reflected shock pressure using a piezoelectric pressure transducer. Meanwhile, OH* emission with a wavelength of 307 nm was selected and measured by a photomultiplier through a narrow band pass filter. Ignition delay is defined as the time interval between the incident shock wave arrived at the end-wall and the onset of ignition, which is determined by the intercept point of the maximum tangent line of the OH* emission curve with the baseline, as shown in [Figure 2](#).

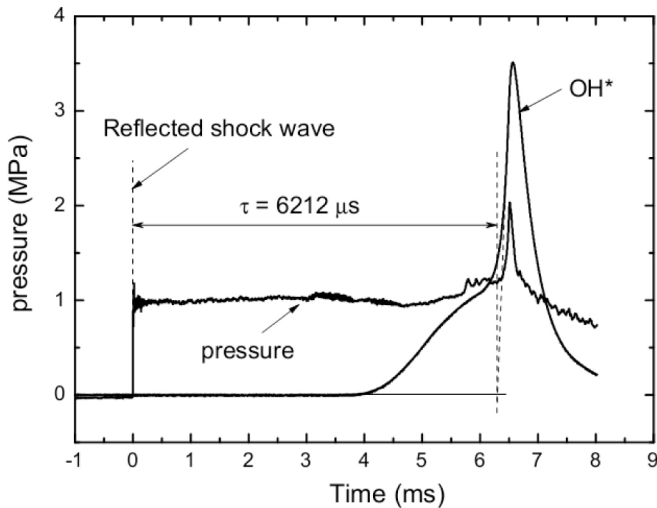


Figure 2. Determination of ignition delay for mixture of 5.28% H₂/5.39% CO/4.6% CO₂/17.79% O₂/66.94%N₂ at pressure of 1.0 MPa, temperature of 921 K.

3.3. Arrhenius-Type Correlation

Experimental results were analyzed using the multiple linear regression method to determine exponential factors that used to normalize measured data. A comparative analysis was carried out between the obtained data and predictive results used current kinetic mechanisms with CHEMKIN software at corresponding regimes.

The effect of various parameters including temperature, pressure, equivalence ratio and reactant components on ignition delays of syngas mixtures was fitted as an Arrhenius-type expression by the multi-linear regression analysis. With a data set, it can take out several forms of regression depend on selected state and mixture parameters. In this study, firstly, all parameters were considered to get a general correlation. Then, exponents, which their influences are insignificant, were eliminated. A general expression is given by,

$$\tau = Ap^{\alpha}\phi^{\beta}X_{H_2}^{\gamma}X_{CO}^{\eta}X_{CO_2}^{\kappa}X_{N_2}^{\lambda}X_{O_2}^{\mu}\exp\left(\frac{E_a}{RT}\right) \quad (3)$$

where, p is pressure in atm, T is temperature in K, ϕ is equivalence ratio, E_a is global activation energy in cal/mol, X are mole fraction of reactants and R is the universal gas constant in cal. Mol⁻¹.K⁻¹. The coefficients A , α , β , γ , η , κ , λ and μ are determined from the multi-linear regression analysis. The final Arrhenius-type correlation is shown as follows,

$$\tau = 6.87 * 10^{-10} p^{0.5} X_{H_2}^{0.93} X_{CO}^{-0.84} X_{N_2}^{-2.2} X_{O_2}^{-1.82} \exp\left(\frac{35694}{RT}\right) \quad (4)$$

The correlation coefficient of the regression analysis is 0.88. In the regression analysis, the high correlation coefficient indicates that experimental results are reliable. All experimental data, related the multi-linear regression analysis, are shown in the [Appendix](#).

The factor of equivalence ratio was eliminated prior to regression analysis because all experiments were carried out at equivalence ratios of 0.3 and 1.0, consequently quantities in matrix of linear equation system presented only two different values. This affects regression analysis quality.

The exponent of CO₂ content is small, the effect of CO₂ on ignition delays is negligible, so the factor CO₂ was ignored. This also eliminates the cases of lack of CO₂ ($X_{CO_2} = 0$ in mixture 1 and mixture 3). Exponent of pressure of hydrocarbon fuel is usually negative at high temperature, but in the expression (4), it has a positive value due to the experiments were carried out at low temperature. The effect of CO₂ content, equivalence ratio and other factors on ignition delays is integrated into the coefficient A ($6.87*10^{-10}$).

3.4. Experimental and Predictive Results

The reviewed literatures indicated that an “S” shape with a high radian of ignition delay curves were presented for syngas mixtures [5, 14, 21], herein, the higher activation energy is exhibited at moderate temperature range, and the the lowest activation energy is presented at low temperature range.

In [Figure 3](#), the solid lines show predictive results at ideal conditions and pressure of 0.2 MPa; the dash lines show predictive results at ideal conditions and pressure of 1.0 MPa; the circle and solid lines show predictive results that considering the effect of facility at pressure of 0.2 MPa; the circle and dash lines show predictive results that considering the effect of facility at pressure of 1.0 MPa; the star-discrete points show predictive results that taking account of real pressure histories from measured data. The square and triangular discrete points show the experimental results at pressures of 0.2 and 1.0 MPa, respectively.

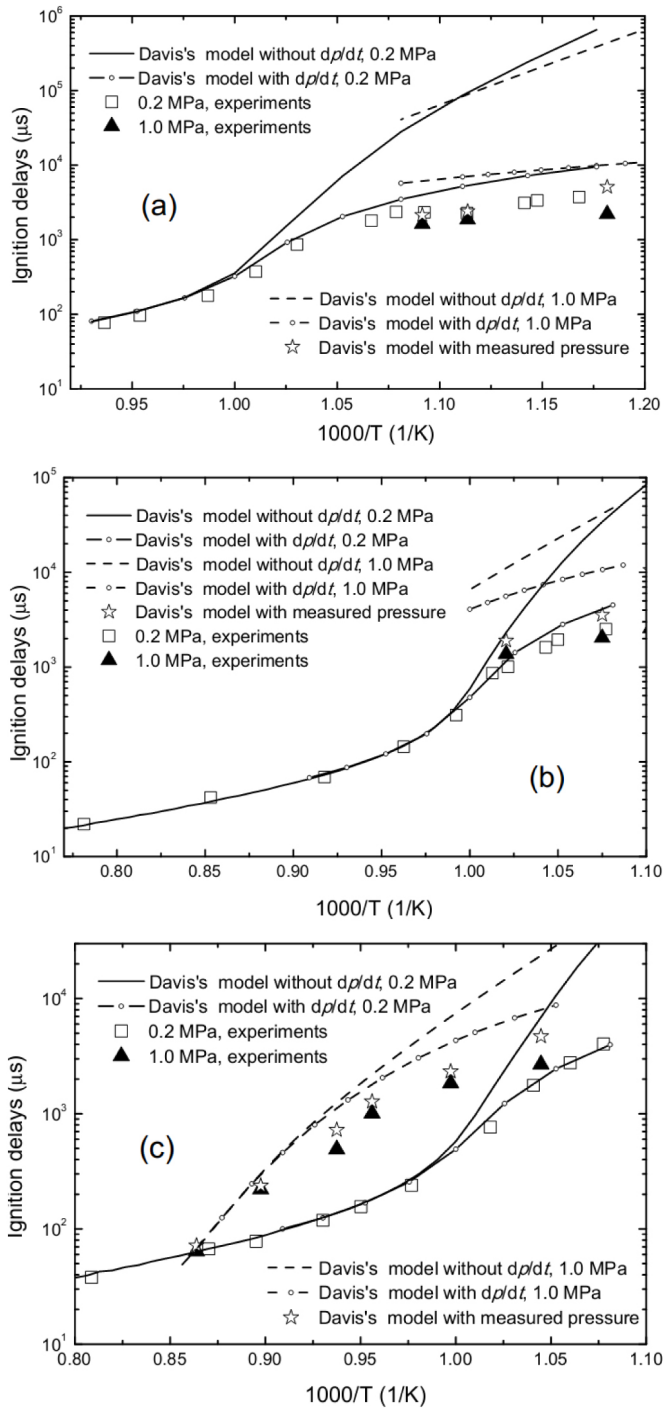


Figure 3. Experimental and predictive results of ignition delay of of three stoichiometric mixtures at pressures of 0.2 and 1.0 MPa a) Mixture #1; b) Mixture #2, c) Mixture #3

The like “S” curve of ignition delays was also observed for syngas mixture in the work of Petersen [22]. The radius of ignition delay curves presented for syngas is higher than that of hydrocarbon fuel. This is a characteristic of fuel mixtures with hydrogen.

The results in Figure 3 indicate the ignition delays at pressure of 0.2 MPa are usually shorter than ones at 1.0 MPa in this study region. The results are also in agreement with the expression (4) in which the exponent of pressure is positive, it means increasing pressure is cause of longer ignition delay. This will be explained more detailed by Figure 7 in section 4.

Figure 3 shows the comparison of ignition delays between measured and simulated results of syngas mixtures at low-moderate temperature. The results show that the measured ignition delays are an order of magnitude lower than ones simulated using Davis's kinetic mechanism at low temperature, however, they are identical at high temperature.

The results are similar to phenomena which were found in previous works [4, 5, 14, 23]. The reasons that could cause the discrepancy between experimental data and predictive values are: (1) effect of boundary layer on movement of gas flow in apparatus [18]; (2) non-idealities of the driver and driven gas mixtures [18]; (3) impurities of the experimental gases [4, 5]; (4) uncertainty in measurements and calculations [16, 17]; (5) accuracy and reliability of the measured instruments [16]; (6) the pre-ignition energy release [24], local ignition kernels and the “hot ignition” phenomena [4]. Therefore, it is necessary to find out methods to eliminate the effects.

According to the classical shock wave theory, the pressure and temperature behind the reflected shock wave are constant at ideal condition. However, an obvious pressure rise behind the reflected shock wave was observed in practical experiments due to effect of boundary layer and non-ideal condition. The observations were presented in previous studies of Pang et al. [24] for $H_2/O_2/Ar$ mixtures at low temperature and Chaos et al. [25] for iso-octane, n-Decane, Toluene and syngas. In addition, Davidson et al. [26] also observed a linear rise of pressure behind the reflected shock wave prior to onset of ignition.

In this study, the facility-related pressure rise rate behind the reflected shock wave is about 4.22% per millisecond ($dp/dt=4.22\%/ms$). Additionally, the pre-ignition energy release has occurred violently and the pressure rise does not conform to linear law for undiluted or low-dilutability mixtures. A typical time-pressure history obtained at the endwall is shown in Figure 4.

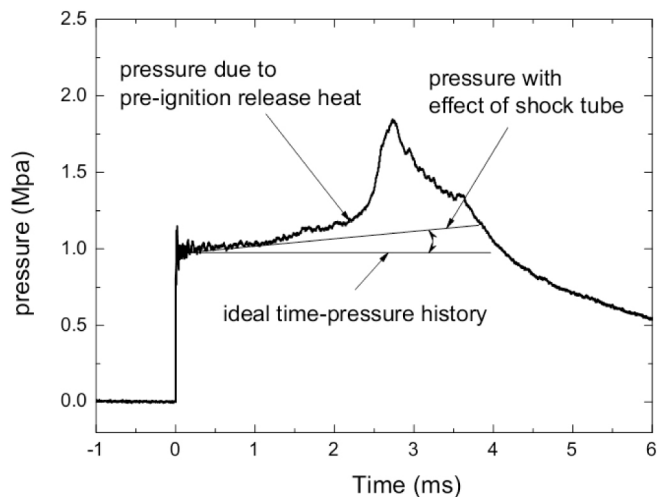


Figure 4. The typical time-pressure history.

The pressure curve does not increase significantly at around 1.7 millisecond. Combining the pressure curve with OH* curve shows that both pressure and OH* emission have risen observably at this timing, however, the reactants have not been combusted yet. The simulation result using CHEMKIN software also indicates temperature of the mixture has not increased at the around timing but has suddenly developed at around 2.5 millisecond. The definitions of onset ignition of CHEMKIN software and ours (as the Figure 2) are identical. Increasing pressure and OH* emission at around 1.7 millisecond is due to the pre-ignition energy release. The phenomena have also reported by Pang et al. [24], Chaos et al [25] and Davidson et al. [27].

At high pressure and low temperature conditions, the pressure behind the reflected shock wave is complex (not linear) as shown in Figure 5. Therefore, using each real pressure trace from measured signal for simulating obtains results closed to experimental data, as shown by the star-discrete points in the Figure 3. The star-discrete points are close to the measured ignition delays (presented by the triangular discrete points).

For premixed combustible mixtures with high hydrogen fraction, their combustion process often accompanies with a detonation phenomenon. It is well known that the strong ignition limit indicates the capability of given reactants for transition from deflagration to detonation, which is a very important combustion problems, for example, unconfined explosion and knocking in thermal engines. Thus, controlling detonation phenomenon is necessary for the operation safety of equipment as well as human. The pressure peaks and the CJ (Chapman-Jouguet) detonation pressures were derived from the pressure curves shown in Figure 5 and corresponding testing conditions. These parameters are useful and applicable to study further detonation characteristics of syngas mixtures.

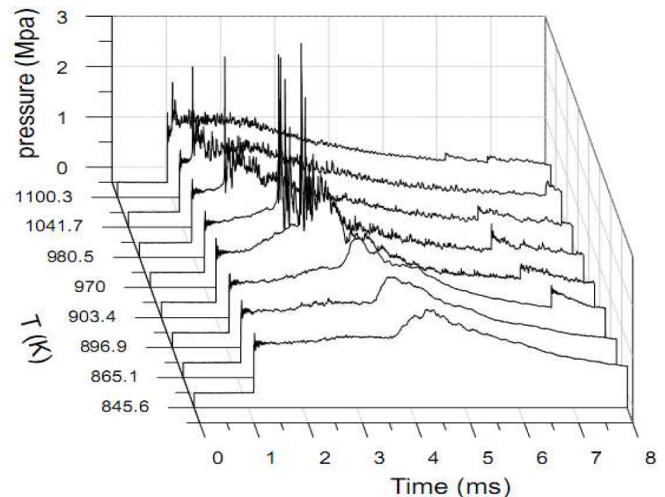


Figure 5. The typical time-pressure histories at different temperatures

Figure 5 shows pressure profiles at the end-wall for the stoichiometric mixture 2 at pressure of 1.0 MPa. The results indicate that in the transitional region (temperature of 903-981K), the pressure peak is higher than the ones of other regions. It could appear the secondary explosion and a high overpressure value between the reflected shock wave and the end-wall. When testing temperature is higher than the strong limit, the pressure peak value decreases. The gathered characteristics illustrate a map related detonation of the syngas mixture, it is an important practical guidance to avoid the dangerous operation regions.

4. SENSITIVITY AND KINETIC ANALYSIS

The predictive results, taken account of the non-ideal effects, approach to experimental data. However, the difference between experimental and predictive results is still remaining. To solve this problem, sensitivity and kinetic analyses were conducted to determine the key reactions which dominate ignition process of syngas.

Sensitivity analysis using the Davis mechanism [28] was conducted to identify the most important elementary reactions which dominate the ignition delays of syngas. Brute force sensitivity calculations for ignition delay were carried out at pressures of 0.2 and 1.0 MPa and the temperatures of 900 K and 1100 K. Sensitivity S is defined as,

$$S = \frac{\tau(2k_i) - \tau(0.5k_i)}{1.5\tau(k_i)} \quad (4)$$

where: τ is ignition delay of the mixture and k_i is specific rate coefficient of reaction i^{th} .

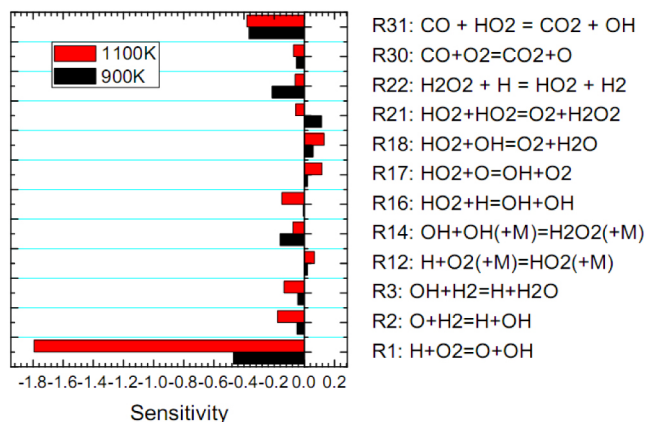
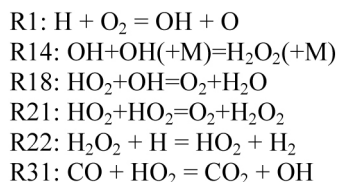


Figure 6. Sensitivity analysis of elementary reactions of mixture 1, at pressure of 1.0 MPa, equivalence ratio of 0.3 with Davis mechanism

Negative value of sensitivity coefficient indicates a promotion effect on total reaction rate and vice versa. Figure 6 depicts the most sensitive reactions in ignition process. The most important reactions are,



The main branching chain reaction R1 exhibits an extreme high negative sensitivity coefficient, it dominates the ignition process of H_2 systems at high and moderate temperature. The effect of reaction R1 on ignition is weaker at low temperature. The reactions R2, R3 and R16 produce strong active radicals at the high-moderate temperatures and also promote the ignition process, whereas the reactions R12, R17 and R18 play the role of inhibition of the ignition process. The reactions R17 and R18 consume two active radicals (O or OH) and HO_2 meanwhile only produce one OH radical (R17) or no radical (R18). At low temperature, the reactions R14 and R22 become more important to ignition process. The reaction R22 produces strong active HO_2 radical (the most important at low temperature) and the reaction R14 produces H_2O_2 radical to provide to the reactions R22 and R31. The reaction R21 has a peculiar effect, it is promotive at high-moderate temperature (it produces O_2 to provide to the reactions R1) and is inhibitive at low temperature (it consumes active radical HO_2). The reactions R12 and R14 are very important at high pressure, because they are main chain termination reactions.

Reason of the effect of the reaction R12 on ignition delays is due to drastically competition between the chain branching reaction R1 and the chain termination reaction R12 in the temperature region. Mertens et al. [29] determined that the

termination reaction R12 plays a controlling role in ignition process at high pressure and low temperature.

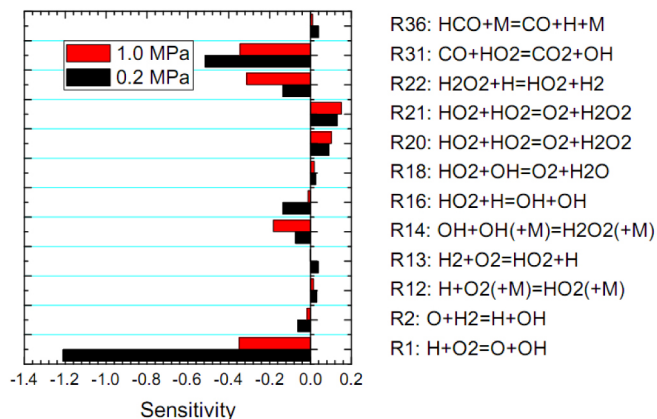


Figure 7. Sensitivity analysis of elementary reactions of mixture 2, at temperature of 900K, equivalence ratio of 1.0 with Davis mechanism

Figure 7 shows the sensitivity analysis results which indicate the effect of the elementary reactions at different pressures on ignition process. Two reactions R1 and R31 are the most important at the different pressures, the effect reduces when increasing pressure. The promoted effect of R1 and R31 on ignition characteristic can explain for the results shown in Figure 3. The reaction R12 inhibits ignition at low pressure because it consumes the strong active H radical. As increasing pressure, the inhibition effect of R12 has decreased due to the R12 is a chain termination reaction. The reaction R14 promotes strongly ignition process at high pressure because it is also chain termination reaction and it produces H_2O_2 radical to supply to the reaction R22. The reactions R20 and R21 inhibit ignition because they consume the strong active radical HO_2 and produce a less active radical H_2O_2 .

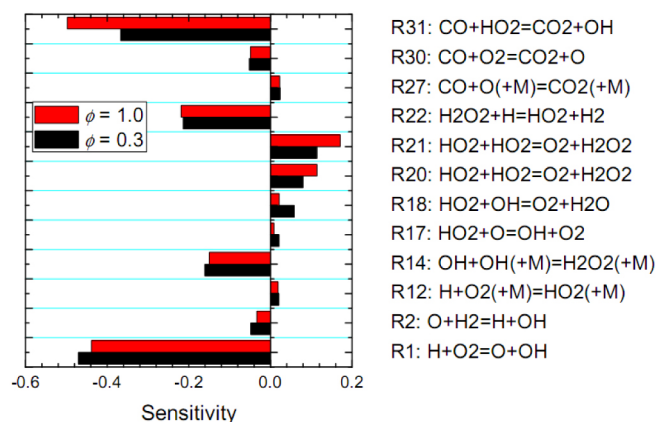


Figure 8. Sensitivity analysis of elementary reactions of mixture 1, at pressure of 1.0 MPa, temperature of 900K with Davis mechanism

Figure 8 shows the sensitivity analysis results which indicate the effect of the key elementary reactions on ignition process at different equivalence ratios. Two reactions R1 and R31 are still the most important at different equivalence ratios, however, their promoted effect is opposite. The effect of the reaction R1 of lean mixture is stronger than one of the stoichiometric mixture due to the O₂ content in lean mixture is higher than one in another. Oppositely, the CO content in rich mixture is higher than one in lean mixture, consequently the influence of reaction R31 is stronger in the stoichiometric mixture.

The effect of the elementary reactions of different stoichiometric mixtures on ignition process is shown as in Figure 9. The sensitivity analysis results indicate that because of additional CO₂ in the mixture 2, other composition contents become lower, so its ignition delays is longer than others. The high CO₂ content also inhibits directly on reactions R30 and R31.

The change of mole fraction of species versus time at temperature of 1100 K is shown as in Figure 10. Radicals H, O, OH are strong active species and have high concentrations during combustion process. Concentrations of other radicals, such as HO₂, HCO, H₂O₂ are not significant. These are not shown in the Figure. The results, shown Figure 6 and Figure 10, indicate that the reaction R1 dominates absolutely ignition process of the syngas mixture at 1100 K, H radical is the most important species promoted the general combustion process. Figure 10 also shows the H₂ consumption rate is much faster than that of CO. At the end of the combustion process (1.2 millisecond), CO content is still left significantly (about 4% in products), while H₂ has been nearly used up.

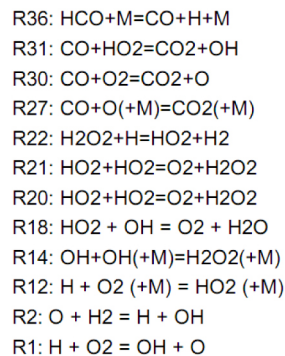
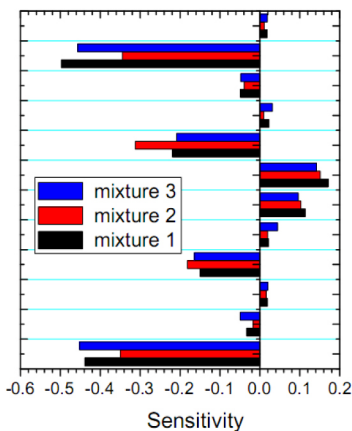


Figure 9. Sensitivity analysis of elementary reactions of stoichiometric mixtures, at pressure of 1.0 MPa, temperature of 900K with Davis mechanism

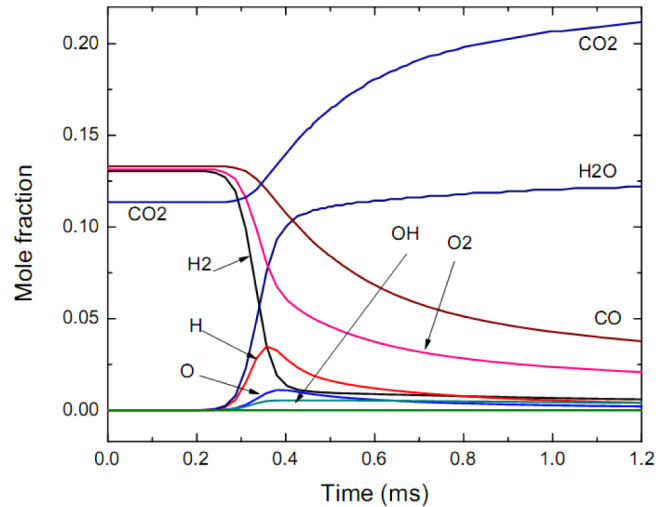


Figure 10. Time - mole fraction histories of species of stoichiometric mixture 2 at pressure of 1.0 MPa, at temperature of 1100 K.

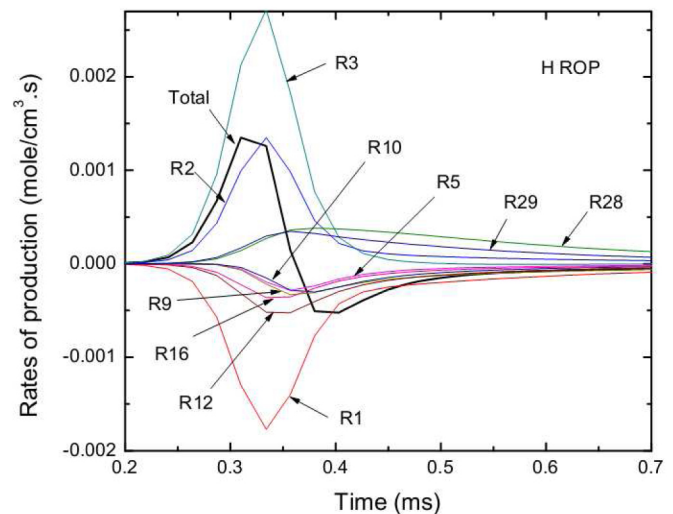


Figure 11. Rates of production of radical H of stoichiometric mixture 2 at pressure of 1.0 MPa, at temperature of 1100 K

Figure 11 shows production rates of typical and active H radical. Positive value implies that the reaction produces the H radical and vice versa. The result also shows that, the reactions R2 and R3 produce H radical in the first stage, whereas the reaction R1 consumes H radical. The main source produced H radical during the last stage are the reactions R28 and R29. The results also indicate that the reaction R12 has consumed H radical considerably, it competes with the reaction R1 for H and O₂ compositions.

The sensitivity and kinetic analyses pointed out the key reactions. Rate constants of these reaction were modified by some related studies to improve the Davis's mechanism suitability. The results indicated that the ignition delays

predicted with the modified mechanism are in good agreement with experimental data.

5. CONCLUSIONS

The results have provided a new data set that could be useful to further research and design plants applied the IGCC technology. This data is a reliable foundation to compare with the simulation results. It is also a firm basis to modify and improve the current chemical kinetic mechanisms. The experimental results provide a more comprehensive insight into the effect of the state and mixture compositions on ignition characteristics of syngas mixtures. The Arrhenius-type expression in Equation 4 is able to quickly calculate ignition delay at the particular conditions. Using a kinetic mechanism and CHEMKIN software to predict ignition delay is complicated and costly.

The expression (4), the experimental results, predictive results and modelling analytic results show that pressure has an inhibition effect on combustion process of syngas mixtures at moderate and low temperature. This is a important difference from characteristics of the typical hydrocarbon fuels.

The predictive results are in good agreement with actual measurements at high temperature. However, the experimental results are about an order of magnitude lower than predictions with Davis's mechanism at low temperature. After using methods to minimize the influence of the practical conditions and considering the effect of facility, the results show that the measured and predictive ignition delays are quite identical.

The sensitivity analyses indicate that the reaction $H+O_2=OH+O$ (R1) plays the most important role in ignition process of syngas mixtures in this study region, the effect is reduced when decreasing temperature and/or increasing pressure. It always competes violently with the chain termination reaction $H+O_2(+M)=HO_2(+M)$ (R12) for O_2 molecule and active H radical. The chain termination reactions R12 and $OH+OH(+M)=H_2O_2(+M)$ (R14) have promotive influence in ignition process at high pressure. The reaction $CO+HO_2=CO_2+OH$ (R31) is also very important at every corresponding regime for carbonmonoxide mixtures.

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APPENDIX*Testing data*

No	p₅ (atm)	φ	X_{H2} (by volume)	X_{CO} (by volume)	X_{CO2} (by volume)	X_{N2} (by volume)	X_{O2} (by volume)	T₅ (K)	t_{ign} (μs)
1.	1.93	1	0.098492	0.19728	0	0.55634	0.14789	1067.8	77
2.	1.94	1	0.098492	0.19728	0	0.55634	0.14789	1048.4	96
3.	2.10	1	0.098492	0.19728	0	0.55634	0.14789	970.5	861
4.	1.98	1	0.098492	0.19728	0	0.55634	0.14789	1013.2	178
5.	2.09	1	0.098492	0.19728	0	0.55634	0.14789	989.8	373
6.	2.06	1	0.098492	0.19728	0	0.55634	0.14789	937.5	1800
7.	2.18	1	0.098492	0.19728	0	0.55634	0.14789	927	2356
8.	2.12	1	0.098492	0.19728	0	0.55634	0.14789	915.3	2323
9.	2.23	1	0.098492	0.19728	0	0.55634	0.14789	898.2	2269
10.	2.11	1	0.098492	0.19728	0	0.55634	0.14789	876.2	3111
11.	2.16	1	0.098492	0.19728	0	0.55634	0.14789	871.4	3364
12.	2.08	1	0.098492	0.19728	0	0.55634	0.14789	856.2	3738
13.	9.92	1	0.098492	0.19728	0	0.55634	0.14789	916.2	1627
14.	10.10	1	0.098492	0.19728	0	0.55634	0.14789	898	1866
15.	9.87	1	0.098492	0.19728	0	0.55634	0.14789	846.3	2205
16.	1.91	0.3	0.037262	0.074636	0	0.7016	0.1865	1086.6	112
17.	2.02	0.3	0.037262	0.074636	0	0.7016	0.1865	1028.5	205
18.	2.17	0.3	0.037262	0.074636	0	0.7016	0.1865	1024.2	297
19.	2.15	0.3	0.037262	0.074636	0	0.7016	0.1865	970.9	1321
20.	2.12	0.3	0.037262	0.074636	0	0.7016	0.1865	947.1	2123
21.	2.16	0.3	0.037262	0.074636	0	0.7016	0.1865	925.2	3669

Testing data (cont.)

22.	2.17	0.3	0.037262	0.074636	0	0.7016	0.1865	921.7	3831
23.	2.15	0.3	0.037262	0.074636	0	0.7016	0.1865	978.2	948
24.	2.07	0.3	0.037262	0.074636	0	0.7016	0.1865	1014.3	333
25.	2.19	0.3	0.037262	0.074636	0	0.7016	0.1865	994.3	726
26.	2.03	0.3	0.037262	0.074636	0	0.7016	0.1865	1060.3	122
27.	2.16	0.3	0.037262	0.074636	0	0.7016	0.1865	1110.8	74
28.	2.10	0.3	0.037262	0.074636	0	0.7016	0.1865	1157.9	57
29.	10.13	0.3	0.037262	0.074636	0	0.7016	0.1865	1041.9	1234
30.	10.19	0.3	0.037262	0.074636	0	0.7016	0.1865	1105.6	431
31.	10.09	0.3	0.037262	0.074636	0	0.7016	0.1865	1128.4	241
32.	10.61	0.3	0.037262	0.074636	0	0.7016	0.1865	1167.5	64
33.	10.56	0.3	0.037262	0.074636	0	0.7016	0.1865	1232.1	17
34.	10.60	0.3	0.037262	0.074636	0	0.7016	0.1865	1086.7	731
35.	10.90	0.3	0.037262	0.074636	0	0.7016	0.1865	1055.7	1161
36.	10.52	0.3	0.037262	0.074636	0	0.7016	0.1865	1015	1398
37.	10.87	0.3	0.037262	0.074636	0	0.7016	0.1865	999.2	1582
38.	10.67	0.3	0.037262	0.074636	0	0.7016	0.1865	979.1	1811
39.	11.01	0.3	0.037262	0.074636	0	0.7016	0.1865	966.6	1922
40.	2.12	1	0.12986	0.13249	0.11297	0.49349	0.13118	1089.7	69
41.	2.14	1	0.12986	0.13249	0.11297	0.49349	0.13118	1279.9	22
42.	2.19	1	0.12986	0.13249	0.11297	0.49349	0.13118	1007.8	311
43.	2.02	1	0.12986	0.13249	0.11297	0.49349	0.13118	978.8	1005
44.	2.02	1	0.12986	0.13249	0.11297	0.49349	0.13118	987.3	862
45.	2.09	1	0.12986	0.13249	0.11297	0.49349	0.13118	958.7	1611
46.	2.14	1	0.12986	0.13249	0.11297	0.49349	0.13118	952.4	1941

Testing data (cont.)

47.	2.13	1	0.12986	0.13249	0.11297	0.49349	0.13118	928.4	2513
48.	1.97	1	0.12986	0.13249	0.11297	0.49349	0.13118	1172.1	42
49.	1.99	1	0.12986	0.13249	0.11297	0.49349	0.13118	1038.9	144
50.	10.33	1	0.12986	0.13249	0.11297	0.49349	0.13118	907.1	2570
51.	10.15	1	0.12986	0.13249	0.11297	0.49349	0.13118	930.2	2040
52.	10.06	1	0.12986	0.13249	0.11297	0.49349	0.13118	979.8	1374
53.	2.14	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	980.2	1177
54.	2.13	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1027.7	199
55.	2.03	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1049.8	132
56.	2.11	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1080.8	88
57.	2.13	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	991.3	864
58.	2.02	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1008.6	447
59.	2.14	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1038.6	178
60.	2.13	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	972.8	1583
61.	2.08	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	951.7	2777
62.	2.07	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1223.6	32
63.	2.02	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1115.2	66
64.	2.12	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1146.9	54
65.	9.91	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	970	2644
66.	9.77	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	980.5	2327
67.	10.06	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1041.7	1187
68.	10.10	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1100.3	539
69.	10.09	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1139.7	182
70.	10.07	0.3	0.052843	0.053912	0.04597	0.66935	0.17792	1174.9	52
71.	2.12	1	0.056662	0.15458	0	0.68314	0.10562	1052.4	156

Testing data (cont.)

72.	2.03	1	0.056662	0.15458	0	0.68314	0.10562	1023.8	239
73.	2.13	1	0.056662	0.15458	0	0.68314	0.10562	982.2	764
74.	2.03	1	0.056662	0.15458	0	0.68314	0.10562	943.3	2767
75.	2.02	1	0.056662	0.15458	0	0.68314	0.10562	928	4025
76.	2.13	1	0.056662	0.15458	0	0.68314	0.10562	960.8	1766
77.	2.11	1	0.056662	0.15458	0	0.68314	0.10562	1117.2	78
78.	2.03	1	0.056662	0.15458	0	0.68314	0.10562	1075	119
79.	2.18	1	0.056662	0.15458	0	0.68314	0.10562	1149.1	67
80.	2.00	1	0.056662	0.15458	0	0.68314	0.10562	1236.6	38
81.	10.54	1	0.056662	0.15458	0	0.68314	0.10562	1002.7	1840
82.	10.22	1	0.056662	0.15458	0	0.68314	0.10562	957.2	2684
83.	10.07	1	0.056662	0.15458	0	0.68314	0.10562	1046	1008
84.	9.44	1	0.056662	0.15458	0	0.68314	0.10562	1066.7	491
85.	10.06	1	0.056662	0.15458	0	0.68314	0.10562	1157.7	64
86.	10.00	1	0.056662	0.15458	0	0.68314	0.10562	1114.2	221
87.	2.03	0.3	0.026069	0.071117	0	0.74084	0.16198	1046.2	205
88.	2.08	0.3	0.026069	0.071117	0	0.74084	0.16198	1004.6	567
89.	2.03	0.3	0.026069	0.071117	0	0.74084	0.16198	991	885
90.	2.13	0.3	0.026069	0.071117	0	0.74084	0.16198	969.2	1905
91.	2.22	0.3	0.026069	0.071117	0	0.74084	0.16198	956.3	2311
92.	2.20	0.3	0.026069	0.071117	0	0.74084	0.16198	949	3048
93.	2.18	0.3	0.026069	0.071117	0	0.74084	0.16198	1019.1	342
94.	2.23	0.3	0.026069	0.071117	0	0.74084	0.16198	1076.7	139
95.	2.19	0.3	0.026069	0.071117	0	0.74084	0.16198	1128.6	102
96.	2.14	0.3	0.026069	0.071117	0	0.74084	0.16198	1171.5	73

Testing data (cont.)

97.	10.01	0.3	0.026069	0.071117	0	0.74084	0.16198	1037.1	2434
98.	10.41	0.3	0.026069	0.071117	0	0.74084	0.16198	1072.9	1298
99.	10.19	0.3	0.026069	0.071117	0	0.74084	0.16198	1122.2	414
100.	9.97	0.3	0.026069	0.071117	0	0.74084	0.16198	1127.6	293
101.	9.89	0.3	0.026069	0.071117	0	0.74084	0.16198	1155.7	118
102.	9.87	0.3	0.026069	0.071117	0	0.74084	0.16198	1218	29
103.	9.88	0.3	0.026069	0.071117	0	0.74084	0.16198	1011.1	3387

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