

# 燃燒

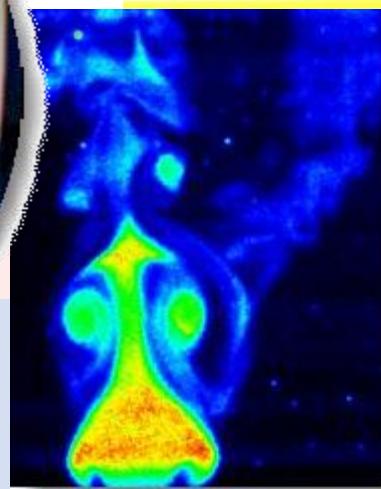


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## 編輯札記

燃燒科技的發展可有效減少消耗能源及資源，有助於減緩對環境生態衝擊，兼顧人類生活環境品質。因而有效提升能源應用效率之技術發展並建置替代能源之最佳化之操作條件參數、將有效減少能源使用與降低污染來改善生活環境並追求生態之永續發展。

本期共刊登六篇專業論文，分別來自大專院校、中鋼等單位之研究成果，長庚大學石心怡教授探討稀釋效應對合成氣體相向噴流擴散火焰的可燃極限、排放氣體、燃燒特徵的分析。崑山科技大學侯順雄教授利用同軸噴流燃燒器，探討不同的氫氣濃度和出口流速，對於反置甲烷-氫氣噴流擴散火焰之火焰形態的影響。高苑科技大學吳志勇教授團隊分析生質柴油B2的噴霧延遲、噴霧速度、噴霧張角以及滲透深度等重要噴霧資訊，相關的結果可以做為使用生質燃料時，引擎性能變異原因之參考。中山大學林汎銓教授使用主要組分分析法精簡生質柴油燃料替代物動力學機制，可有效加快內燃機的燃燒模擬速度。崑山科技大學吳澤松教授設計一套熱電共生裝置，並評估此組共生裝置油品的能源回收率，離形機之能源回收率，於轉速 3600rpm、發電量 3.2 kW，回收效率達 59.8%。中鋼劉永章博士團隊，建立 RH 真空精煉製程冶煉鋼液[C]≤10ppm 的製程技術，以滿足超低碳鋼種的生產需求。

本期的內容豐富多元，理論與實務並重，提供讀者有關基礎燃燒理論、生質柴油有效運用與製程開發之新知。亦請本刊讀者支持及踴躍投稿本期刊，共同推動燃燒、能源與環境科技知識分享及研究交流。

副總編輯 蔡健雄  
總編輯 侯順雄

# 稀釋效應對合成氣體相向噴流擴散火焰燃燒特性之影響

## Dilution Effects on the Combustion and Extinction of Opposed-jet Syngas Diffusion Flames

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### 摘要

本文主要探討稀釋效應對合成氣體相向噴流擴散火焰的可燃極限、排放氣體、燃燒特徵的分析。研究建立固定拉伸率下的一維相向擴散噴流火焰模型，使用詳細的化學反應式及熱力傳輸性質，並搭配窄頻輻射模型。本文也討論了分別加入三種稀釋氣體(二氣化碳、水氣、氮氣)的火焰結構、一氧化氮排放以及合成燃氣的可燃極限。發現當以二氣化碳為稀釋氣體時，最高火焰溫度為三者最低，並導致一氧化氮產生率減少。然而當稀釋氣體為水氣時，富含一氧化碳的合成燃氣的一氧化氮產生率最低，主因為  $\text{NNH}+\text{O}\rightarrow\text{NH}+\text{NO}$  的反應速率減小。但是在接近熄滅時，卻有最高的火焰溫度，因為化學反應與輻射效應的相互影響。本文同時比較不同拉伸率和不同比例的稀釋氣體下的可燃極限圖。透過加入稀釋氣體使合成燃氣熄滅，除了是惰性效應的影響，水氣產生的化學反應也使火焰溫度更高；另外在低拉伸率的可燃極限中，水與二氣化碳的輻射效應扮演很重要的角色。

關鍵字：噴流擴散火焰、合成燃氣、稀釋效應、可燃極限。

### Abstract

The dilution effects on the combustion, emissions, and extinction characteristics of opposed-jet syngas diffusion flames were studied in this paper. A model of one-dimensional counterflow syngas diffusion flames was constructed with constant strain-rate formulations, which used detailed chemical kinetics and thermal and transport properties with flame radiation calculated by statistic narrow band radiation model. The dilution effects from three diluents ( $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{N}_2$ ) on flame structures, NO emission, and extinction limits of  $\text{H}_2/\text{CO}$  synthetic mixture flames were discussed. It was found that the maximum flame temperature is decreased the most with  $\text{CO}_2$  as the diluents, which leads to a significant reduction of NO formation from Zeldovich

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route. However, CO-rich syngas flame with H<sub>2</sub>O dilution has the lowest NO production rate, due to a reduced reaction rate of NNH+O→NH+NO, but highest flame temperature at extinction due to the competition between chemical effect and radiation effect. The flammability maps were also constructed with strain rates and dilution gases percentages as the coordinates. By adding dilution gases to make the syngas non-flammable, besides the inert effect from of diluents, the chemical effect of H<sub>2</sub>O contributes to higher flame temperature, while the radiation effect of H<sub>2</sub>O and CO<sub>2</sub> plays an important role in the flame extinction at low strain rates.

Keywords: syngas flames, opposed-jet diffusion flames, dilution effect, flame extinction.

## 1. Introduction

Syngas or synthetic gas, which generally contains H<sub>2</sub> and CO as the main fuel components, has drawn a lot of attention recently, particularly for stationary power generation with IGCC (Integrated Gasification Combined Cycle) technology [1, 2]. The syngas can be formed through gasification processes [3, 4], and can be produced from many kinds of fossil fuels and solid combustibles, such as coal, biomass, refinery residual and even municipal waste. As the energy demand and environmental concerns continue to grow, syngas is expected to be one of the promising fuels prior to a complete hydrogen-economy society. However, there is a considerable variation of H<sub>2</sub>/CO ratio with the rest being primarily N<sub>2</sub>, CO<sub>2</sub>, and H<sub>2</sub>O. These three diluents may be present or introduced in the syngas mixture during the gasification and refining processes. Regardless of the definition differences in the study of dilution effects in the past, here the possible dilution effects on the flames are categorized as: (1) inert effect, (2) thermal/diffusion effect, (3) chemical effect and (4) radiation effect. First, the inert effect means the addition of the diluents serves as an inert gas that simply reduces the concentrations of the reactants relatively, so that the overall reaction rate decreases. Second, the thermal/diffusion effect indicates the changes of the thermal and transport properties of the syngas mixture due to the presence of the diluents. The flame temperature and species distribution are then altered. Third, the dilution gas is not completely chemically inert and it is involved in the chemical reaction itself. Fourth, H<sub>2</sub>O and CO<sub>2</sub> are strong radiation-participating gases compared to N<sub>2</sub>, and radiation is one of the important heat transfer modes involved in the diffusion flames, especially for the flames at low stretch rates or under low-speed convective flows.

There is a fair amount of research dealing with the dilution effects of H<sub>2</sub>/CO syngas fuels. For the premixed syngas/air combustion, the dilution influence on laminar flame speed (or mass burning rate), flame stability and flammability are the

primary concerns [5-8]. Some previous research works have investigated H<sub>2</sub>/CO syngas diffusion flames with dilution gases, but they may or may not focus on the dilution effects. For example, Drake and Blint [9] numerically examined the effect of flame stretch on the NO<sub>x</sub> formation in opposed-flow diffusion flames with CO/H<sub>2</sub>/N<sub>2</sub> fuel. Park et al. [10] numerically studied the addition effects of CO/CO<sub>2</sub> in H<sub>2</sub>/Ar fuel on the flame structure and NO emission in counterflow diffusion flames. More concentrated on the study of dilution effects, Giles et al. [11] investigated NO<sub>x</sub> emission characteristics of syngas diffusion flames with air-stream dilution. Two representative syngas mixtures with equal molar percentage of H<sub>2</sub>/CO were simulated and three diluents, N<sub>2</sub>, H<sub>2</sub>O, and CO<sub>2</sub> in air were considered. Considerable research efforts have been made in studying the flame characteristics of H<sub>2</sub>/CO syngas diffusion flames diluted with CO<sub>2</sub> by Park et al. [12-14]. The preferential diffusion effects of H<sub>2</sub> on flame behavior were examined for two representative H<sub>2</sub>-enriched and CO-enriched H<sub>2</sub>/CO syngas diffusion flames. The radiative heat loss effects and the chemical effects of added CO<sub>2</sub> as diluents on the flame extinction of H<sub>2</sub>/CO syngas diffusion flames were also studied. Recently, a numerical model of opposed-jet diffusion flames coupled with narrowband radiation model was used to study the combustion, flame extinction, and NO<sub>x</sub> emission characteristics of counterflow syngas diffusion flames by Shih and Hsu [15, 16]. Besides the effects of syngas compositions and pressures, the dilution effects from three diluents of CO<sub>2</sub>, H<sub>2</sub>O, and N<sub>2</sub> were also examined. All these previous studies indicate the dilution effects could be so complex that involves in the combustion system with various contributing effects from inert effect, thermal/diffusion effect, chemical effect, and radiation effect. Therefore, in this work, we attempt to investigate the dilution effects on the combustion extinction, and NO<sub>x</sub> emission of H<sub>2</sub>/CO syngas diffusion flames.

## 2. Mathematical and Numerical Models

### 2.1. Opposed-jet diffusion flames

The physical model considered here is a counterflow, axisymmetric laminar diffusion flame stabilized near the stagnation plane of two opposing jet flows, as shown schematically in figure 1. Two equivalent formulations of the problem exist in the literature [17]: one specifying the flame strain rate (or stretch rate) and the other the jet exit velocity and the distance between two nozzles. The constant strain rate formulation is utilized in this paper, and a constant strain rate, defined as the radial gradient of the radial velocity at the fuel boundary is assumed. The detailed mathematical formulations and governing equations can be found in [18]. Fuel and oxidizer jets are located at negative and positive sides in the axial coordinate, respectively. The fuel is the CO/H<sub>2</sub> mixture as the syngas, and the oxidizer is air. The dilution gases such as CO<sub>2</sub>, H<sub>2</sub>O and N<sub>2</sub> are added at the fuel side for the discussion of

the dilution effects.

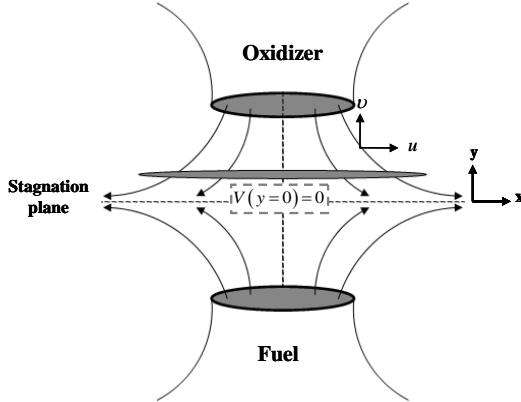


Figure 1. Schematic of opposed-jet diffusion flame

A comparison of different radiation treatments for one-dimensional diffusion flames has been made in the past, indicating that optically thin and gray gas models are inadequate for quantitative predictions [19]. In this work, a statistical narrowband model with the exponential tailed inverse line strength distribution is used to calculate the radiative properties of the participating gases assumed in the calculations (CO, CO<sub>2</sub>, and H<sub>2</sub>O). The combination of detailed kinetics with a narrowband radiation mode provides more accurate prediction on the flame structures and extinction limits quantitatively, which was also used by Daguse et al. [20] for N<sub>2</sub>-diluted H<sub>2</sub>/air diffusion flames at 1 atm pressure and by Shih et al. [21, 22] for H<sub>2</sub>/O<sub>2</sub> diffusion flames with CO<sub>2</sub> dilution.

The narrowband model enables one to calculate spectrally averaged emissivity and absorption coefficients within a small wavenumber range and it is the most accurate other than line-by-line calculations. The nongray radiative transport equation for an absorbing and emitting medium written in terms of the mean transmittance over a narrowband is solved, where both collision and Doppler broadening are considered. The collision half-width and the line spacing parameters are taken from Soufiani and Taine [23]. More details on the narrowband model can be found in [24].

## 2.2. Numerical methods

The opposed-jet constant-stretch diffusion flame equations were solved and coupled with the narrowband radiative transfer equations. The calculations were carried out with a code revised from OPPDIF program by Lutz et al. [25] for opposed-jet diffusion flame configuration. This program was converted from the constant velocity formulation to the constant strain rate formulation, and radiation heat transfer is accounted for by statistical narrowband solver. In the code, upwind and central difference schemes are used for the convective and diffusion terms, respectively. The nongray narrowband radiative transfer equation was solved with the S<sub>8</sub> discrete

ordinates method [24]. A 20-direction Gaussian quadrature set was used. The radiation participating gaseous species included are CO, CO<sub>2</sub>, and H<sub>2</sub>O.

The solution of the governing equations also requires the knowledge of transport coefficients ( $\lambda$ ,  $\mu$ ) and the thermodynamic properties ( $c_p$ ,  $c_{pk}$ ,  $h_k$ ). These data were determined using the CHEMKIN and TRANSPORT package [26, 27]. Both ordinary and thermal diffusion were considered. The detailed kinetics was taken from GRI 3.0 [28]. The Variable grid distribution was used and the grid sizes were adaptable, which are controlled by specifying the acceptable first and second gradients in the solution. The difference between the present results and the ones with much more stringent gradient specifications is minimal. A grid independence check has been performed.

### 3. Results and Discussions

#### 3.1. Dilution Effects on Flame Structures

In the model calculations, detailed flame structures and net reaction rates at different dilution gas percentages are examined for CO-rich (20% H<sub>2</sub>, 80% CO) and H<sub>2</sub>-rich (80% H<sub>2</sub>, 20% CO) syngas flames at normal atmosphere. Figure 2 presents the variations of maximum flame temperatures with three dilution gases (CO<sub>2</sub>, H<sub>2</sub>O and N<sub>2</sub>) for CO-rich syngas flames at strain rates of 10 and 1000 s<sup>-1</sup>, which represents the cases of low-stretch and high-stretch flames respectively. Results show the maximum flame temperatures continually decrease when the mole fractions of dilution gases increase.

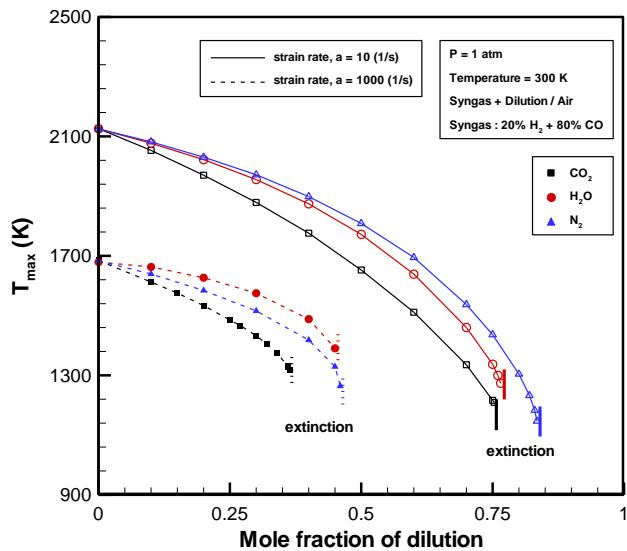


Figure 2. Variations of maximum flame temperature with dilution for CO-rich syngas flames at strain rates of 10 s<sup>-1</sup> and 1000 s<sup>-1</sup>.

At the same dilution percentage, the maximum flame temperature of the low stretch flame decreases the most by adding CO<sub>2</sub>, followed by H<sub>2</sub>O and N<sub>2</sub>. However,

at the high strain rate, the maximum flame temperature of CO<sub>2</sub>-diluted syngas flame still drops the most, but it decreases much less with H<sub>2</sub>O as the diluents. In addition, the flame extinction limits in terms of the dilution gas percentage, where the minimum mole fraction of dilution needed to extinguish the syngas flames, can be reached with less CO<sub>2</sub> compared to the others. The highest dilution percentage for flame extinction is obtained by N<sub>2</sub> dilution at both high and low strain rates, but H<sub>2</sub>O-diluted syngas flame has the highest maximum flame temperature when flame extinction occurs, which has also been found for lean-premixed syngas flames in recent study [29].

Figure 3 then shows the variations of maximum flame temperatures with dilution gases for H<sub>2</sub>-rich syngas flames at strain rates of 10 and 1000 s<sup>-1</sup>. Similar trends have been found that the maximum flame temperatures decrease with increasing mole fractions of dilution gases, and the temperature drops the most for CO<sub>2</sub> dilution. In addition, adding the same amount of dilution gases, the maximum flame temperature decreases much less with H<sub>2</sub>O as the diluents at the high strain rate, although not as obvious as that for CO-rich syngas flames in figure 2. There are two possible reasons for H<sub>2</sub>O-diluted syngas flames to behave so interesting. First, the existence of CO may inhibit the most important chain branching reaction of H+O<sub>2</sub>→O+OH [30]. With H<sub>2</sub>O addition, the reaction of OH+CO→H+CO<sub>2</sub> increases, which then promotes the chain branching reaction and overall reactions. Consequently, the maximum temperatures decrease less with H<sub>2</sub>O addition, and the extinction limit is now extended closer to that of N<sub>2</sub>-diluted gas.

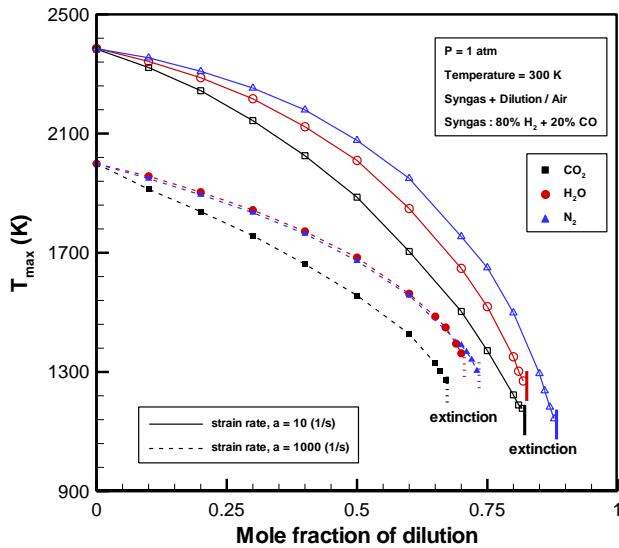


Figure 3. Variations of maximum flame temperature with dilution for H<sub>2</sub>-rich syngas flames at strain rates of 10 s<sup>-1</sup> and 1000 s<sup>-1</sup>.

Second,  $\text{CO}_2$  and  $\text{H}_2\text{O}$  are radiation-participating media. Radiation heat loss from those species would cause the flame temperature to drop, and the decrease of temperature is more pronounced for flames at low strain rates. Therefore, the chemical effect of  $\text{H}_2\text{O}$  dilution enhances the overall reactions, especially for the CO-rich syngas flame, which increases flame temperature and extend the extinction limit. But the flame temperature of  $\text{H}_2\text{O}$ -diluted syngas flame become lower than that of  $\text{N}_2$ -diluted flame at lower strain rate due to the radiation effect from  $\text{H}_2\text{O}$  dilution.

Evidence can be seen in figure 4, where the chemical heat generations and the ratios of radiation heat loss to the chemical heat generations (R/C ratio) for CO-rich syngas flames with dilutions are shown. Results indicate the heat generations are larger for high-stretch flames, and it decreases with increasing dilution percentage due to the inert gas effect. At the same percentage of dilution, the heat generation is the largest with  $\text{H}_2\text{O}$  as the diluents, attributed to the chemical effect from  $\text{H}_2\text{O}$ . On the other hand, the R/C ratios demonstrate the radiation heat loss has a more significant role at low strain rate. With  $\text{N}_2$  dilution, the R/C ratio gradually decreases with increasing the percentage of dilution. However, for  $\text{H}_2\text{O}$  and  $\text{CO}_2$  dilution, it exhibits some non-monotonic behaviors. The ratios could slightly increase at intermediate dilution percentages and then drop quickly before flame extinction. Although  $\text{CO}_2$  gas is a stronger radiation-participating media than  $\text{H}_2\text{O}$ , both the production rates of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  are beneficial from the addition of  $\text{H}_2\text{O}$ . Therefore, with  $\text{H}_2\text{O}$  addition, the ratio of radiation heat loss over the chemical heat generation is the largest rather than that from  $\text{CO}_2$  dilution.

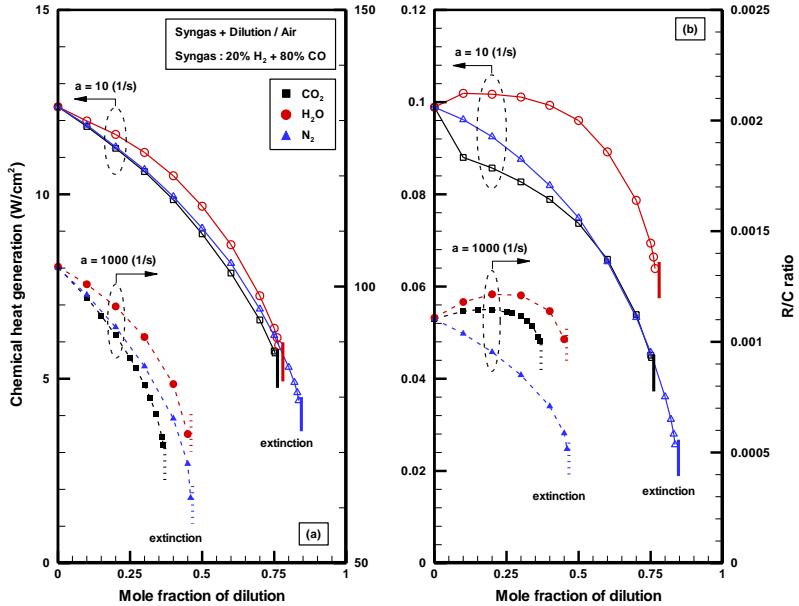


Figure 4. The comparisons of chemical heat generation and R/C ratio for CO-rich syngas flames with dilution gases at strain rates of  $10$  and  $1000 \text{ s}^{-1}$ .

### 3.2. Dilution Effects on NO<sub>x</sub> emissions

Figure 5 and figure 6 show the variations of NO production rates from the main reactions with different mole fractions of dilutions for H<sub>2</sub>-lean and H<sub>2</sub>-rich syngas, respectively. Four categories of reactions are discussed, which includes the NO formations from N<sub>2</sub> directly (reversed R178: N<sub>2</sub>+O→NO+N), through N<sub>2</sub>O-, NNH-intermediate and N species (R179: N+O<sub>2</sub>→NO+O, R180: N+OH→NO+H). In figure 5, even with dilution, NO is produced mainly through R179 and R180, and all the NO production rates from these reactions for H<sub>2</sub>-lean syngas decrease with increasing the dilution percentages. At the same dilution percentages, the NO production rates for these reactions discussed are still the highest with N<sub>2</sub>-dilution, but when using H<sub>2</sub>O as the diluent, it shows a significant decrease for the reaction sequences from NNH to NO. The reduced reaction rate of R208 (NNH+O→NH+NO) is believed to be the reason why NO mole fraction decreases the most with H<sub>2</sub>O-dilution despite the flame temperature and thermal NO production from the reversed reaction of R178 (N<sub>2</sub>+O→NO+O) are decreased the most with CO<sub>2</sub>-dilution.

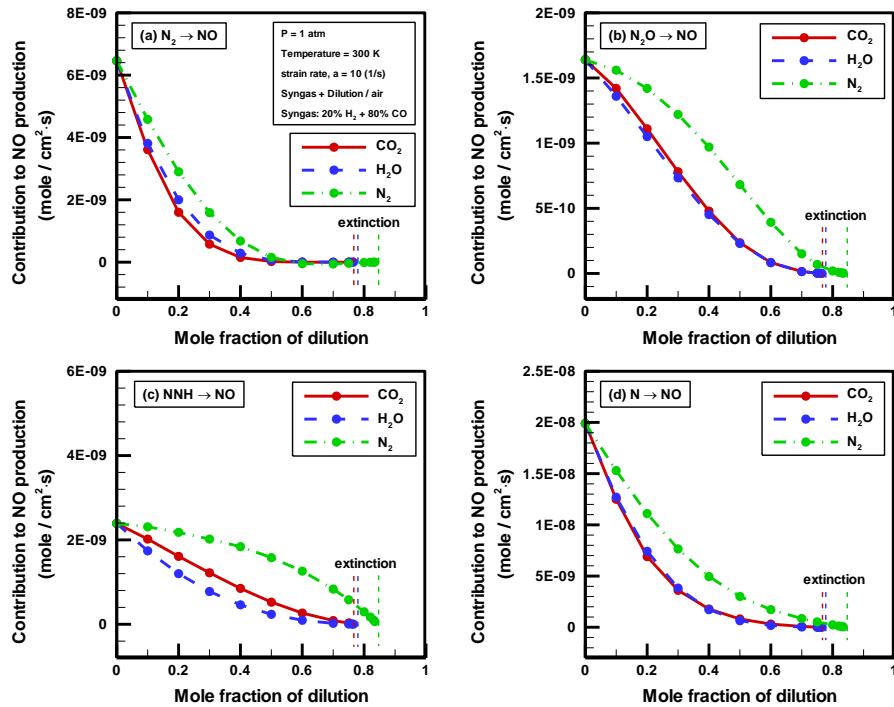


Figure 5. The contribution to NO production from main reactions at different dilution percentages for H<sub>2</sub>-lean syngas flames.

Figure 6 then compares the variations of the NO production rates from these particular reactions for H<sub>2</sub>-rich syngas at different mole fractions of dilution gases. Except the NO production from N<sub>2</sub>O-intermediate, where the production rates exhibit a non-monotonic behavior over the dilution percentages, the production rates of NO from other reactions continually decrease with increasing mole fraction of dilutions. Since

the thermal NO route (reversed R178, R179, R180) is dominant, the H<sub>2</sub>-rich syngas with N<sub>2</sub> dilution has the highest temperature and NO production rate. The interesting responses of the N<sub>2</sub>O-intermediate route to the dilution gases are due to the competition between forward and backward reactions of R199 (NH+NO=N<sub>2</sub>O+H). When forward reaction prevails, NO is converted to N<sub>2</sub>O for H<sub>2</sub>-rich syngas flame, however, when backward reaction rate is larger, NO is produced through the N<sub>2</sub>O-intermediate route as that for H<sub>2</sub>-lean syngas. Therefore, at a small amount of dilution gases, the backward reaction rate of R199 is larger than that of forward reaction rate, consequently, NO production rates increase with the percentage of dilution.

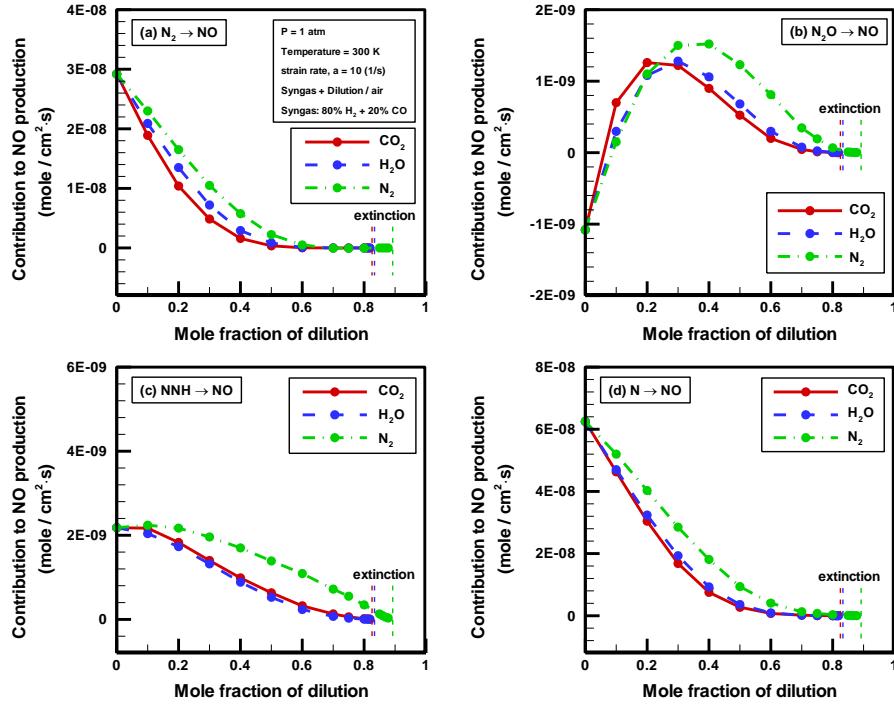


Figure 6. The contribution to NO production from main reactions at different dilution percentages for H<sub>2</sub>-rich syngas flames.

The phenomenon is then reversed when the dilution percentage becomes large, that is, NO production rates decrease with increasing dilution percentage. However, the non-monotonic changes of NO production rates from N<sub>2</sub>O-intermediate are different with the type of dilution gases. At smaller amount of dilution, CO<sub>2</sub>-diluted syngas produces more NO through N<sub>2</sub>O-intermediated than other dilutions, but at higher dilution percentage, N<sub>2</sub>-diluted syngas creates more NO instead. The differences may come from several aspects though. First, when adding a small amount of CO<sub>2</sub>, the CO<sub>2</sub> gas is not totally chemically inert, and it can be converted to CO near high temperature region through one of the primary CO reaction mechanism in backward direction: reversed R99 (CO<sub>2</sub>+H→CO+OH). The H<sub>2</sub>-rich syngas actually becomes leaner, and for leaner-H<sub>2</sub> syngas, NO is produced more through

$\text{N}_2\text{O}$ -intermediate, therefore NO production rate from  $\text{N}_2\text{O}$ -intermediate increases the most with  $\text{CO}_2$  as the diluent. On the other hand, at higher dilution percentages with  $\text{N}_2$ ,  $\text{N}_2\text{O}$  is then produced more with enhanced reaction rate of reversed R185 ( $\text{N}_2+\text{O}(\text{+M}) \rightarrow \text{N}_2\text{O}(\text{+M})$ ) compared to that with  $\text{CO}_2$  and  $\text{H}_2\text{O}$  diluents. The NO production rate with  $\text{N}_2$  dilution is then the highest among these three diluents. Overall speaking, the NO production rate drops the most with  $\text{CO}_2$  as the dilution. It is attributed to the reduced flame temperature and NO formation from thermal route (reserved R178, R179 and R180).

### 3.3. Dilution Effects on the Flame Extinction

To determine the dilution extinction boundary in this study, an equal-molar  $\text{H}_2/\text{CO}$  syngas (50%  $\text{H}_2$ , 50%  $\text{CO}$ ) are computed with various diluents on the fuel side of the opposed jet. The dilution extinction boundary is constructed in figure 7 with strain rate and dilution gases percentage as the coordinates. Two groups of inversely U-shaped curves are obtained. One is for fuel-side diluted syngas combustion in air, and the other is  $\text{CO}_2$  dilution on both fuel and oxidizer sides, just for reference. Apparently, with dilution gas on both sides, the flammable range is narrower. In addition, the dilution percentages drop drastically for quenching limits at lower strain rates compared to those for blowoff limits at higher strain rates, indicating the significance of radiation effect than chemical effect from the diluents for low-stretched flames. More importantly, the limiting dilution percentages for quenching extinction drop the most with  $\text{H}_2\text{O}$  dilution, but it decreases the most with  $\text{CO}_2$  dilution for blowoff extinction. A crossover of extinction boundaries between  $\text{CO}_2$  and  $\text{H}_2\text{O}$  dilutions are found and the reasons require further investigations.

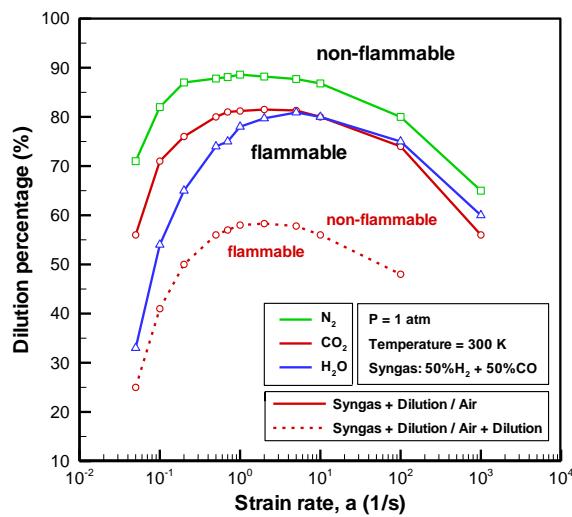


Figure 7. Flammability boundary constructed with strain rates and dilution gas percentages for equal-molar syngas.

The dilution effects analyses are made to explain the crossover and the same approaches are applied as described in Ref. [31], where the dilution effect were categorized as four individual effects: inert effect, thermal/diffusion effect, chemical effect, and radiation effect. Figure 8 plots the variations of maximum flame temperature versus dilution percentage of three dilution gases ( $\text{N}_2$ ,  $\text{CO}_2$ , and  $\text{H}_2\text{O}$ ) for equal-molar  $\text{H}_2/\text{CO}$  syngas fuel at low strain rate of  $0.1 \text{ s}^{-1}$ . Results show the maximum flame temperature continually decreases with increasing dilution percentage until flame extinction by quenching. The peak flame temperature drops the most for  $\text{H}_2\text{O}$ -diluted flames, followed by  $\text{CO}_2$  and then  $\text{N}_2$  dilution. The extinction dilution percentage with  $\text{N}_2$ -diluted syngas fuel is the highest, and it is least with  $\text{H}_2\text{O}$  dilution. To distinguish the individual effects on the extinction limit, first we assume that  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{N}_2$  are inert gases with the same thermal and transport properties as the syngas fuel. Therefore, at the strain rate of  $0.1 \text{ s}^{-1}$ , 89.3% dilution is needed to extinguish syngas flames regardless the types of diluents. The differences in the extinction dilution percentages then come from the thermal/diffusion, chemical, and radiation effects, and the radiation effect contributes to flame extinction significantly among them. Normally,  $\text{CO}_2$  gases are more active in radiation participation than  $\text{H}_2\text{O}$ . However, adding  $\text{H}_2\text{O}$  can produce more OH radicals through the reactions of  $\text{H}_2\text{O}+\text{H}\rightarrow\text{H}_2+\text{OH}$  and  $\text{H}_2\text{O}+\text{O}\rightarrow\text{OH}+\text{OH}$ . This actually leads to the promotion of  $\text{CO}_2$  production rate through  $\text{CO}+\text{OH}\rightarrow\text{H}+\text{CO}_2$ . Therefore, the  $\text{H}_2\text{O}$ -diluted flame has larger amounts of radiatively species including  $\text{CO}_2$ ,  $\text{CO}$  and  $\text{H}_2\text{O}$ , despite  $\text{CO}_2$  gases is more radiative and less reactive than  $\text{H}_2\text{O}$ . With  $\text{H}_2\text{O}$  addition, the flame temperature before extinction is highest due to chemical effect, but the lowest extinction dilution percentage at lower strain rates attributes to the radiation effect.

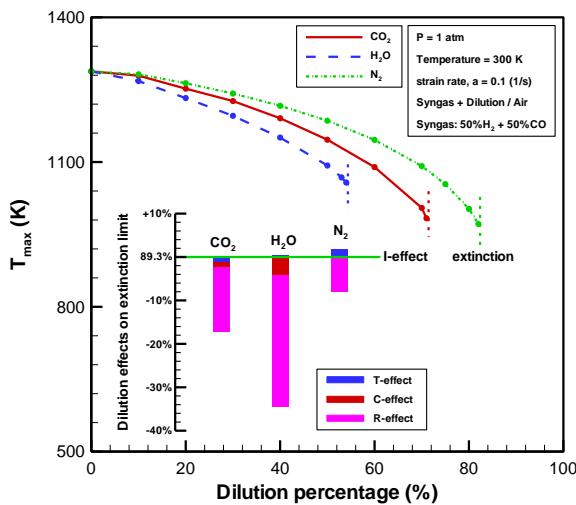


Figure 8. The dilution effects analysis for equal-molar syngas at strain rate of  $0.1 \text{ (1/s)}$ .

Further dilution analysis was made for the flame at high strain rate of  $1000\text{ s}^{-1}$  to provide the comparisons and better understanding of the crossover. The results are shown in figure 9, where the peak flame temperature drops the most for  $\text{CO}_2$ -diluted flames, followed by  $\text{N}_2$  and then  $\text{H}_2\text{O}$  dilution.  $\text{N}_2$ -diluted syngas fuel has the highest extinction dilution percentage, and  $\text{CO}_2$  dilution has the least. Results indicate 62.5% dilution is required for syngas flame extinction from the inert effects. For  $\text{CO}_2$  dilution, the thermal/diffusion, chemical and radiation effects all favour flame extinction, and thermal/diffusion effect actually contributes the most with higher heat capacity. Flame goes out earlier due to lower flame temperature at very high  $\text{CO}_2$  dilution percentage. On the other hand, although  $\text{H}_2\text{O}$  dilution assists flame extinction with chemical and radiation effects, especially from chemical effect, the thermal/diffusion effect can extend the extinction dilution percentage. As for  $\text{N}_2$ -diluted syngas flames, the flammability is extended mainly by the thermal/diffusion effect. Consequently, the maximum extinction dilution percentage for syngas flames at high strain rates is the highest for  $\text{N}_2$  dilution, followed by  $\text{H}_2\text{O}$  and  $\text{CO}_2$ . This is quite different from the flames at low strain rate and the crossover of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  extinction boundaries are then well explained.

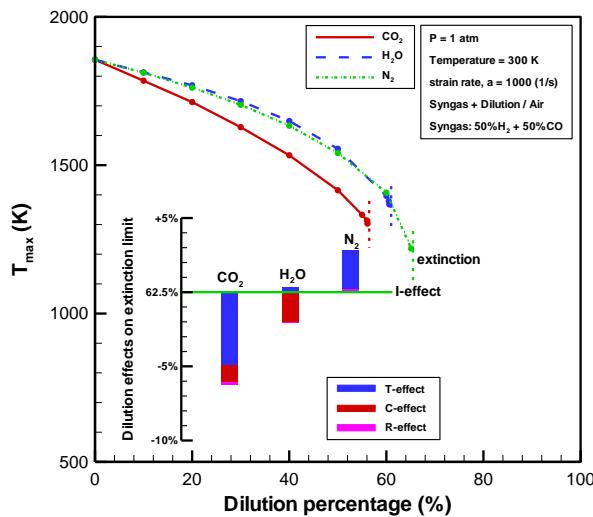


Figure 9. The dilution effects analysis for equal-molar syngas at strain rate of  $1000\text{ (1/s)}$ .

#### 4. Conclusions

The dilution effects on the combustion and extinction of  $\text{H}_2/\text{CO}$  syngas mixture indicate the overall chemical reactions are inhibited due to inert gas effect of dilution gases. At a low strain rate, the flame temperature is lower and the dilution percentage for flame extinction is higher with  $\text{CO}_2$  and  $\text{H}_2\text{O}$  as the dilution gases. However at a

high strain rate, the flame temperature of H<sub>2</sub>O-diluted syngas flame is the highest and its dilution percentage for flame extinction is then close to that of N<sub>2</sub>-diluted flame, because the chemical effects of H<sub>2</sub>O are more conspicuous than radiation effects. The dilution effect analysis indicated the shift of the lowest extinction dilution percentage from CO<sub>2</sub> to H<sub>2</sub>O dilutions at lower strain rate is attributed to the radiation effect. The dilution effects on NO production rates indicate, for H<sub>2</sub>-lean syngas flame, the NO production rate for CO<sub>2</sub>-dilution is slightly higher than that with H<sub>2</sub>O-dilution. It is because the H<sub>2</sub>O-diluted syngas reduces the reaction rate of R208 (NNH+O→NH+NO), which causes a significant decrease in NO production from NNH route, despite a higher flame temperature and thermal NO production for H<sub>2</sub>O-diluted syngas flame.

## 5. Acknowledgments

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# 甲烷-氫氣反置擴散火焰之火焰型態分析

## Appearances of Methane-Hydrogen Inverse Diffusion Flames

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### 摘要

本研究利用同軸噴流燃燒器，探討不同的氫氣濃度和出口流速，對於反置甲烷-氫氣噴流擴散火焰之火焰形態的影響。結果發現，在固定的第一環燃料出口速度( $V_F$ )下，逐漸增加第二環(內管)空氣出口速度( $V_A$ )，火焰型態會依序出現 Type A、Type B、Type C 和 Type D 四種類型的火焰。其中 Type A 為單層(外層)焰端閉合之擴散火焰，Type B 為內層和外層擴散火焰共存之雙層焰端閉合之擴散火焰，Type C 為內層和外層擴散火焰共存之雙層焰端開口(破孔)M 型擴散火焰，Type D 為內層火焰上飄(lift-off)並與外層火焰連接之雙層火焰。研究亦發現摻混氫氣比例越高，內層擴散火焰穩定性越佳。此外，摻氫比例增加，根部藍焰增長，但火焰高度會逐漸降低。在相同的第二環燃料出口速度( $V_F$ )下，火焰高度隨著第一環空氣出口速度( $V_A$ )的增加而有逐漸降低的趨勢。

關鍵字：反置擴散火焰、火焰型態、同軸噴流燃燒器、氫氣

### Abstract

In this study, combustion characteristics of inverse methane jet diffusion flames were investigated using a coaxial Jet burner. The results showed that as the fuel (methane) stream velocity ( $V_F$ ) are fixed, the gradual increase in the center stream (air) velocity ( $V_A$ ) will result in the transition of inverse diffusion flame from a single cone-shaped flame (Type A) to a double cone-shaped flame (Type B) to a M-shaped flame with inner and outer open tips, so-called Type C flame, to a Type D flame with

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lifted inner flame. When  $V_F$  is kept constant, flame height decreases with increasing  $V_A$ . As  $V_F$  was fixed, the critical velocities of center air stream ( $V_A$ ) corresponding to the occurrence of Type B, Type C and Type D flames increased with the hydrogen concentration in the methane/ hydrogen mixture.

Keyword: Inverse diffusion flame, Flame appearances, Coaxial jet burner, Hydrogen

## 一、前言

依燃燒結構或燃料及空氣供給方式的不同，氣態噴流火焰通常可分為噴流擴散火焰 (jet diffusion flame)、噴流部份預混火焰(partially-aerated jet partially premixed flame)和噴流預混火焰 (fully-aerated jet premixed flame) 三種型式[1]。第一種型式為無空氣混合的噴流擴散火焰，其燃燒現象出現在燃料和外圍空氣的分界面，火焰面位置取決於燃料跟氧化物的擴散能力。第二種型式則是部分空氣混合的噴流部份預混火焰，係由噴管中混入部份空氣而造成預混效果。一般而言，部份預混火焰的基本定義為火焰從反應區的兩邊獲得燃料或氧化物或兩者兼具[2-4]。如果預混空氣量足夠，將會出現內層富油預混火焰和外層擴散火焰同時並存的雙層火焰結構[1-4]。若燃空比超過富油可燃極限，內層預混火焰必需靠著外層擴散火焰的支持，與外層擴散火焰並存於流場中。第三種型式為充分空氣混合的噴流預混火焰，當預混空氣量等於或超過氣態燃料的化學計量比 (stoichiometric)，則外環的擴散火焰將消失不見，此時噴流場中將僅存一化學計量比的預混火焰或一貧油預混火焰。

一般而言，單管噴流燃燒器較少使用在工業應用或家用燃燒器。為了得到大面積的加熱面或增加混合效果，多噴口噴流燃燒器常使用在工業燃燒系統及家用燃氣設備上[4]。而依安全性、適用性、燃燒效率與污染防治等觀點，其所使用的火焰型態亦會有所不同[6, 7]。一般而言，噴流擴散火焰的火焰長度較長，容許較長的加熱腔，焰色呈黃色，其顯示熱輻射傳遞較強；火焰的穩定性受制於上飄 (lift-off)，吹掉 (blow-out) 和回駐 (drop back) 等現象，狹窄的火焰高溫區會減少高溫 NO (thermal NO) 的形成，但黃色火焰卻代表大量不完全燃燒的碳顆粒 (soot) 分布[8-12]。

為防止回火現象的發生，工業用途的燃燒器大都使用噴流擴散火焰，如原動力廠和汽電共生廠中的鍋爐、工業用加熱爐和氣渦輪機等。擴散火焰不需考慮回火的問題，因此在實際工業應用系統是比較安全的。但因擴散火焰之燃料與氧化物是以化學計量比燃燒(stoichiometric burning)，且燃燒氣體在高溫區的遲滯時間較長因而通常會導致較嚴重的污染排放，特別是氮氧化物( $\text{NO}_x$ )和碳顆粒(soot)。由於經由燃燒火焰所衍生的環境污染， $\text{NO}_x$  排放一直扮演著很重要的角色，因此在在工業應用中，經常利用分段燃燒(stage combustion)技術來降低  $\text{NO}_x$  排放量 [3]。

反置擴散火焰因具有降低氮氧化物( $\text{NO}_x$ )和碳顆粒(soot)的優點，因此引起眾多學者的關注[13, 14]。本研究所探討之中心管噴氧化物、第二環噴燃料與第三

環噴氣化物之三環同軸反置噴流擴散火焰可用來模擬工業用燃燒爐之分段燃燒技術。氫-碳氫化合物混合氣之相關研究大都聚焦在燃燒污染物排放，而在火焰形態(flame appearances)和火焰穩定性的著墨仍相當匱乏。

本研究探討不同氫氣含量之氫-甲烷混合氣體燃料的火焰形態和火焰穩定性，以瞭解不同氫氣含量對富氫製程剩餘氣體燃料與碳氫燃料混燒之燃燒特性的影響。

## 二、實驗設備與研究方法

本實驗探討甲烷-氫氣反置噴流擴散火焰，固定第二環( $V_F = 30 \text{ cm/s}$ )下，改變第二環甲烷/氫氣混合氣的氫氣濃度( $\Omega_{H_2} = 0\%、20\%、40\%$ )，由小而大逐漸增加內管空氣速度( $V_A$ )，分析內管空氣速度和第二環氫氣濃度對甲烷-氫氣反置擴散火焰之火焰型態、結構之影響。圖 1 所示為本實驗中所使用的儀器設備包含有：噴流燃燒器、氣體供應系統、影像擷取系統、火焰高度量測系統、抽氣系統，如下所述：

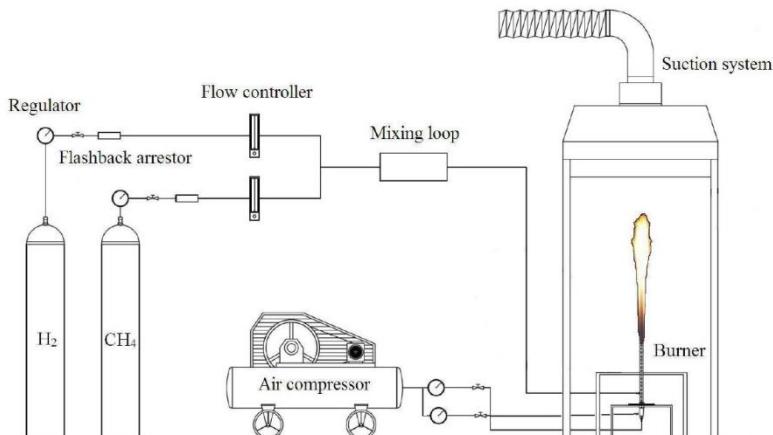


圖 1、實驗設備配置圖。

本研究所使用的同軸擴散火焰燃燒器系統，係由一個雙環同軸噴流燃燒器所構成。雙環同軸噴流燃燒器中的內管與外環之內徑尺寸分別為管徑 10 mm 與 16 mm。其中內管之管壁厚為 1 mm，外環管壁厚為 2 mm。外環直接由螺紋固定於燃燒器底座，內管則是先套接在中心圓管轉接座，再利用內管轉接座上的螺紋與燃燒器底座固定。最後將燃燒器底座固定於圓盤，再將圓盤架設於實驗平台上。最後於最外環之外側裝置一矩形之陶瓷纖維板或金屬塊以形成一垂直壁，用於模擬一般加熱爐內之壁面存在與噴流燃燒器的距離(側壁間距, Separation Distance，簡稱 S)。研究中，內管的氣體由下方進入，外環管的氣體則由兩側進入。各管內部均以三層之蜂巢網與細目鐵絲網進行整流，使各噴流出口之速度為均勻分布。內管供給空氣，外環管供給燃料，如此可以形成一個雙環同軸反置擴散火焰(double-coaxial inverse diffusion flame)。若內管供給空氣(僅為靜止空氣)，外環管

供給燃料，則可形成環狀反置擴散火焰(annular inverse diffusion flame)。實驗中使用圓柱座標系，其原點設定於中心圓管出口平面上的圓心， $r$  為噴流之徑向， $Z$  方向為噴流之軸向。利用雙環同軸噴流燃燒器，可進行各操作參數的實驗分析。

氣體供應系統包含輸送管路、壓力控制閥與氣體流量計三項。實驗使用的燃料為高壓鋼瓶裝之高純度甲烷(98%)、及氬氣(99.9%)，內管供給的空氣則是由空氣壓縮機所提供之。甲烷、氬氣及空氣分別由各自的調壓閥(regulator)控制壓力，使供給壓力固定在錶壓 1.5 atm，然後甲烷、氬氣和空氣則是經由各自的氣體質量流量控制器(flow controller)，分別來控制各自所需的流量。空氣經由管線直接從燃燒器底部進入內管，而甲烷和氬氣則是先經由混合管路(mixing loop)充分混合後，再經由進氣導管從燃燒器側面進入外環管兩側圓環，然後經點火燃燒形成所需的火焰形態。

研究中將利用影像系統來進行火焰的形態和焰色的觀察以及高度的量測。在本實驗中所使用的影像處理系統為數位單眼照相機(Nikon D80)。數位單眼照相機為 1020 萬畫素，拍攝不同實驗條件下的火焰形態。解析度為  $2896 \times 1944$  pixel，擷取之影像可看出火焰長度，以利於火焰高度量測。

火焰高度的量測，將一鐵尺立於噴環右方，以利於拍攝火焰高度後，輸入電腦觀測的比例尺。利用影像擷取系統所拍攝之圖片，輸入至電腦，並利用電腦軟體 Image Tool，進行量測火焰高度；量測值為火焰頂端擾動最高點至火焰最低點和管口，並以二十組的數據取平均值。

噴流燃燒器距離上方錐狀不鏽鋼抽氣罩約 1.2 m，利用伸縮鋁環將抽氣罩連接於 1hp 的抽氣馬達，藉以排出燃燒產物。此外，利用錐狀抽氣罩，以降低燃燒時所產生的高溫廢氣。

### 三、火焰型態分析

首先針對本研究中可能產生之火焰型態進行定義和說明。如圖2所示，在固定的外環燃料流速( $V_F$ )下，火焰結構隨著內管空氣出口流速  $V_A$  逐漸增加而可分為 Type A、Type B、Type C、Type D 四種型態。其中 Type A 為單層(外層)焰端閉合之擴散火焰，Type B 為內層和外層擴散火焰共存之雙層焰端閉合之擴散火焰，Type C 為內層和外層擴散火焰共存之雙層焰端開口(破孔)M 型擴散火焰，Type D 為內層火焰上飄(lift-off)並與外層火焰連接之雙層火焰。



圖 2、火焰型態說明(Type A、B、C、D)。

Type A為單層(外層)焰端閉合之擴散火焰，意指由於內管空氣流速未到達某一臨界速度，以至於內層反置擴散火焰尚未形成，而外層標準擴散火焰以橘黃色火焰為主體，僅在火焰底部為些許藍色火焰存在。Type B為內層和外層擴散火焰共存之雙層焰端閉合之擴散火焰，意指由於內管空氣流速逐漸增加，使內層擴散火焰足以形成於內管出口處上方，火焰尖端為閉合情況(closed flame tip) 之內層圓錐形反置擴散火焰；外層則由外環燃料和靜止之外圍空氣形成外層標準擴散火焰。Type C為內層和外層擴散火焰共存之雙層焰端開口(破孔)M型擴散火焰，意指由於內管空氣流速逐漸增加，使得外層火焰焰端由閉合轉變為開口(tip opening)，此時內層火焰亦為開口情況，且內外火焰之焰端連接在一起形成M型火焰頂端。Type D為內層火焰上飄(lift-off)並與外層火焰連接之雙層火焰；內層擴散火焰浮火是因為管口附近流體流動之特徵時間小於化學反應所需的特徵時間，以至於內層擴散火焰無法建立，促使火焰離開管口，而飄高於噴口上方流場強度較小的區域。因此逐漸增加內管空氣出口流速的情形下，當 $V_A$ 流速過快，內管空氣與外環燃料於管口附近沒有足夠的時間進行燃燒反應，使得內層火焰無法於管口附近形成附著火焰(attached flame)，遂產生焰端開口之上飄火焰(lifted flame)，且內層擴散火焰之焰端與外層擴散火焰相連接。

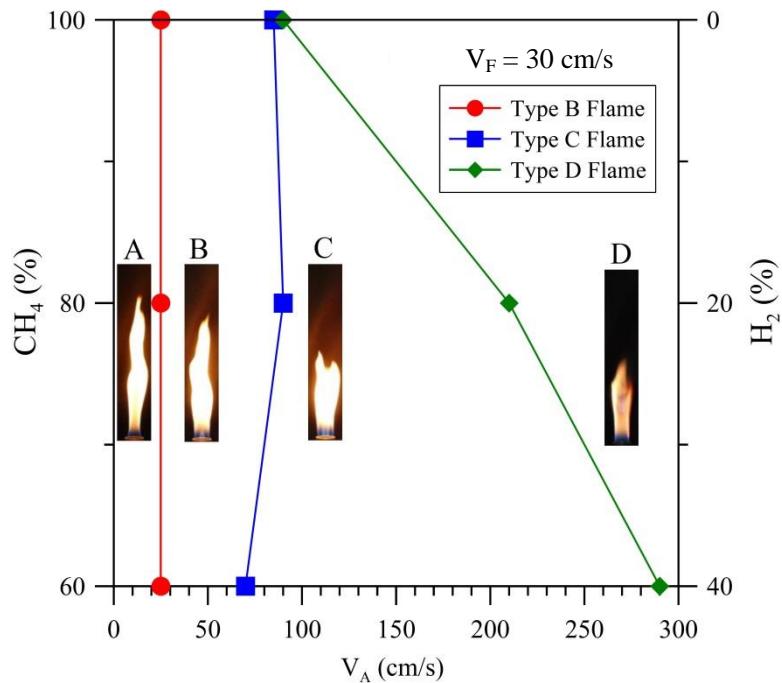


圖 3、固定  $V_F = 30 \text{ cm/s}$ ，不同摻混氫氣比例  $\Omega_{H_2}$  (0%、20% 和 40%  $H_2$ )下，火 焰型態與  $V_A$  之變化關係圖。

圖3所示為固定外環燃料出口速度  $V_F = 30 \text{ cm/s}$  時，不同摻混氫氣比例(0%、20%和40%  $H_2$ )情況下之火焰形態與內管空氣出口速度( $V_A$ )之火焰高度的變化圖。結果發現：在固定  $V_F$  的情況下，火焰高度會隨著  $V_A$  的增加而逐漸降低的趨勢；且隨著混氫比例的增加，火焰底部藍焰部分亦隨之增高，而火焰高度則隨著混氫比例的增加而逐漸降低。此外，當固定  $V_F$  時，隨著混氫比例增加，Type C 火焰型態，因整體火焰高度下降，導致外層火焰提前產生開口(tip opening)，因此形成之臨界  $V_A$  值減少，但是 Type D 火焰初始形成之臨界  $V_A$  值隨著混氫比例增加亦逐漸增加，此一特性顯示氫氣摻混比例增加，內層火焰穩定性更佳，上飄延後於較大的內管出口速度才出現。

圖4所示為固定外環甲烷出口速度  $V_F = 30 \text{ cm/s}$  時，逐漸增加內管空氣出口速度  $V_A$  的火焰照片圖。可知當  $V_A = 20 \text{ cm/s}$ ，內層反置擴散火焰尚未形成，而外層標準擴散火焰以橘黃色火焰為主體，僅在火焰底部為些許藍色火焰存在(Type A 火焰)。逐漸增加內管空氣出口速度達  $V_A = 25 \text{ cm/s}$  時，火焰尖端為閉合情況(closed flame tip)之內層圓錐形反置擴散火焰開始形成於內管管口上方，此為內層火焰初始生成的臨界速度，亦即 Type B 火焰開始形成。再逐漸增加  $V_A$ ，可發現內焰高度逐漸增加，且火焰頂端碳煙狀黃焰(sooty yellow flame)也增多，整體火焰亮黃色部分也更加明亮。再逐漸增加  $V_A$  ( $30 - 80 \text{ cm/s}$ )，可發現整體火焰高度亦有逐漸下降之趨勢。當  $V_A = 85 \text{ cm/s}$  時，外層火焰焰端由閉合轉變為開口(tip opening)，此時內層火焰亦為開口情況，且內外火焰焰端連接在一起形成M型火焰頂端，此即為 Type C 火焰開始形成。當  $V_A = 90 \text{ cm/s}$  時，內層火焰出現上飄(lift-off)現象，整體火焰黃色火焰部分明顯大幅下降，呈現淡黃色火焰。

由內層和外層淡藍色火焰包覆著的火焰型態(Type D火焰)。此外，由 $V_A = 0$  cm/s逐漸增加至85 cm/s的過程亦可發現外層火焰底部有由內而外逐漸擴張的現象。而當內層火焰出現上飄之後，外層火焰底部又有向內縮的現象。

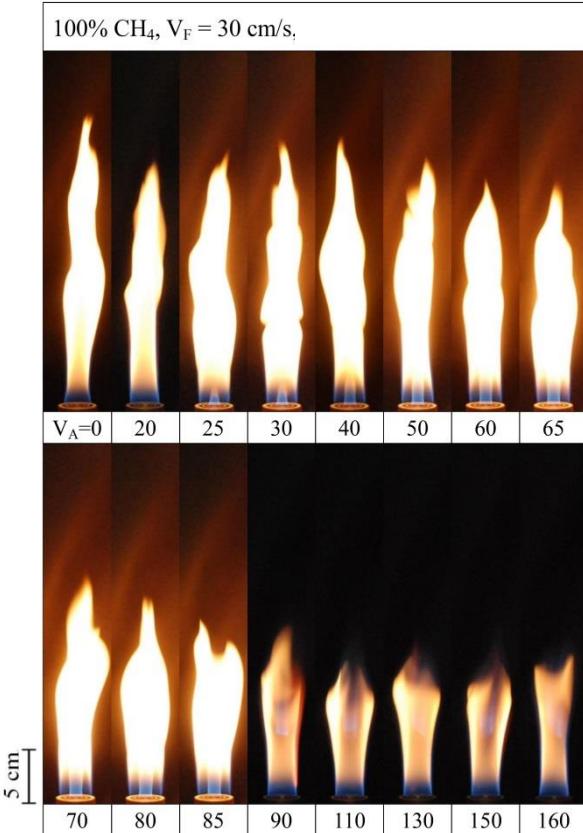


圖 4、固定第二環甲烷出口速度  $V_F = 30$  cm/s，逐漸增加內管空氣出口速度  $V_A$  的火焰型態變化圖。

圖 5 所示為固定外環摻混氫氣比例為 20%  $H_2 + 80\%$   $CH_4$  之結果，當固定  $V_F = 30$  cm/s 時，逐漸增加  $V_A$ ，火焰型態會依序出現 Type A、Type B、Type C 和 Type D 四種類型的火焰型態。當  $V_A = 20$  cm/s，內層反置擴散火焰尚未生成，而外層標準擴散火焰係以橘黃色火焰為主體，但在火焰底部可發現添加氫氣時有較長的藍色火焰存在(Type A 火焰)。逐漸增加內管空氣出口速度達到  $V_A = 25$  cm/s 時，亦即 Type B 火焰開始形成。再逐漸增加  $V_A$ ，可觀察發現內焰高度逐漸升高，且其火焰頂端碳煙狀黃焰也增多，整體火焰亮黃色部分也更多且更加明亮。當  $V_A = 90$  cm/s 時，外層火焰之焰端由閉合轉變為開口，此時內層火焰亦為開口情況，且內外火焰焰端連接在一起形成 M 型火焰頂端，亦即 Type C 火焰開始形成。再逐漸增加  $V_A$  (90 - 190 cm/s)，可發現 M 型火焰頂端更加明顯，且整體火焰高度有逐漸下降之趨勢。當  $V_A = 210$  cm/s 時，內層火焰出現上飄現象，且整體火焰之黃色火焰部分明顯地大幅下降，而呈現內層有破孔的暗藍色火焰和外層淡黃色火焰所包覆著的火焰型態(Type D 火焰初始生成)。此外，由  $V_A = 25$  cm/s

逐漸增加至 210 cm/s 的過程亦可發現外層火焰底部有由內而外逐漸擴張的現象。而當內層火焰出現上飄之後，外層火焰底部又有逐漸向內縮的現象。

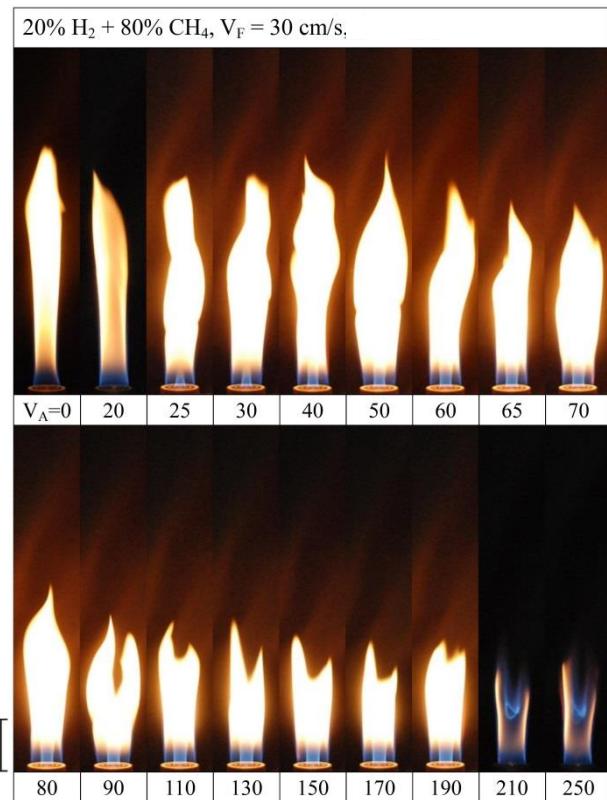


圖 5、固定第二環摻混氫氣比例為 20%(20% H<sub>2</sub> + 80% CH<sub>4</sub>)，V<sub>F</sub> = 30 cm/s，逐漸增加內管空氣出口速度 V<sub>A</sub> 的火焰型態照片圖。

圖 6 所示為固定外環摻混氫氣比例為 40% H<sub>2</sub> + 60% CH<sub>4</sub>，V<sub>F</sub> = 30 cm/s 時，逐漸增加內管空氣出口速度 V<sub>A</sub> 的火焰照片圖。當 V<sub>A</sub> = 20 cm/s，內層反置擴散火焰尚未生成，而外層標準擴散火焰係以橘黃色火焰為主體，但在火焰底部可發現添加氫氣越多時有越長的藍色火焰存在(Type A 火焰)。逐漸增加 V<sub>A</sub> 達到 25 cm/s 時，Type B 火焰開始形成。再逐漸增加 V<sub>A</sub>，可觀察發現內焰高度逐漸升高，且其火焰頂端碳煙狀黃焰也增多，整體火焰亮黃色部分也更多且更加明亮。當 V<sub>A</sub> = 70 cm/s 時，Type C 火焰開始形成。再逐漸增加 V<sub>A</sub> (70 - 280 cm/s)，可發現 M 型火焰頂端更加明顯，且整體火焰高度有逐漸下降之趨勢。當 V<sub>A</sub> = 290 cm/s 時，內層火焰出現上飄現象，且整體火焰轉變成藍色火焰及些許的桃紅色火焰，而呈現內層有破孔的暗藍色火焰和外層藍色帶有些許之桃紅色火焰所包覆著的火焰型態(Type D 火焰初始生成)。此外，由 V<sub>A</sub> = 25 cm/s 逐漸增加至 290 cm/s 的過程亦可發現外層火焰底部有由內而外逐漸擴張的現象。而當內層火焰出現上飄之後，外層火焰底部又有逐漸向內縮的現象。

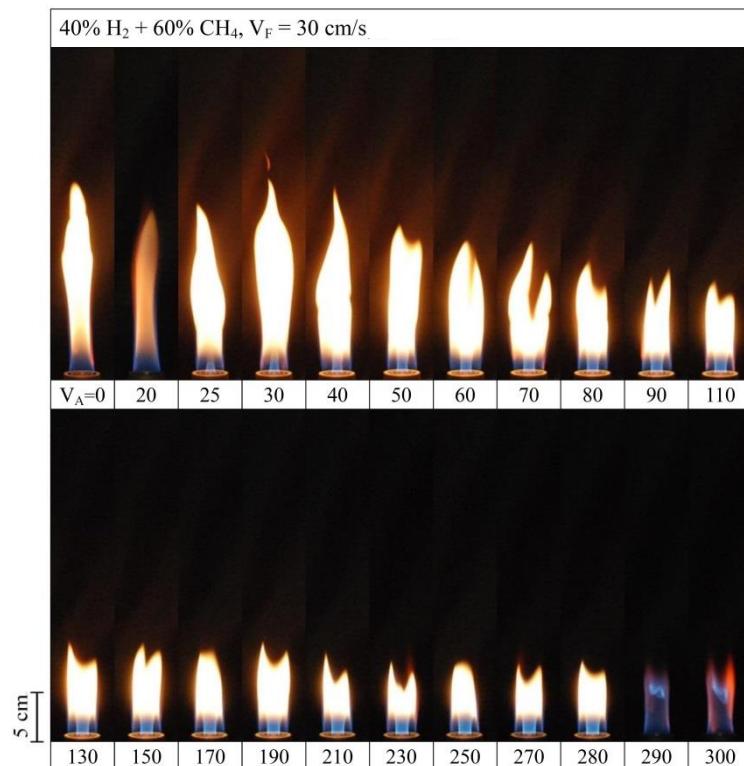


圖 6、固定第二環摻混氫氣比例為 40%(40% H<sub>2</sub> + 60% CH<sub>4</sub>)，V<sub>F</sub> = 30 cm/s，逐漸增加內管空氣出口速度 V<sub>A</sub> 的火焰型態照片圖。

圖 7 所示為，V<sub>F</sub> 固定時，不同摻混氫氣比例(0%、20% 和 40% H<sub>2</sub>)情況下，逐漸增加 V<sub>A</sub>，火焰型態會依序出現 Type A、Type B、Type C 和 Type D 四種類型的火焰。但隨著混氫比例增加，Type C 火焰高度下降，導致外層火焰提前出現開口，因此 Type C 火焰形成之臨界 V<sub>A</sub> 值減少；然而隨著氫氣摻混比例增加，內層火焰穩定性更佳，上飄延後於較大的內管出口速度才出現，亦即 Type D 火焰初始形成之臨界 V<sub>A</sub> 值增加。

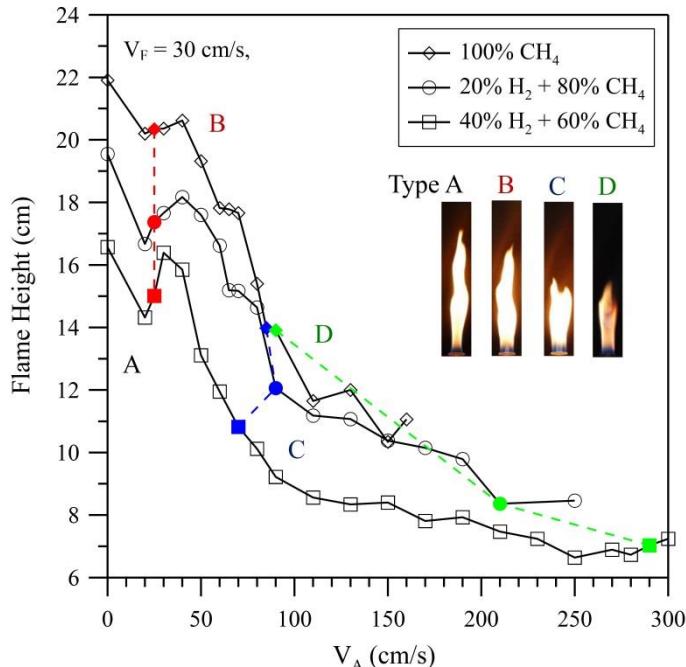


圖 7、固定  $V_F = 30 \text{ cm/s}$ ，不同摻混氫氣比例(0%、20%和 40%  $\text{H}_2$ )下，火焰高度與  $V_A$  之變化關係圖。

#### 四、結論

- 當固定第二環燃料出口速度( $V_F$ )時，隨著第一環空氣出口速度( $V_A$ )的增加，火  
焰型態大致可分為四種型態：
  - Type A為單層(外層)焰端閉合之擴散火焰。
  - Type B為內層和外層擴散火焰共存之雙層焰端閉合之擴散火焰。
  - Type C內層和外層擴散火焰共存之雙層焰端開口(破孔)M型擴散火焰。
  - Type D為內層火焰上飄(lift-off)並與外層火焰連接之雙層火焰。
- 當固定第二環燃料出口速度( $V_F$ )、內管空氣速度( $V_A$ )時，隨著摻混氫氣比例增  
加，火焰的高度會降低。而固定第二環燃料之氫濃度與  $V_F$  時，火焰高度隨著  
第一環  $V_A$  值增加而降低。
- 在相同的第二環燃料出口速度( $V_F$ )下，Type D 火焰型態開始形成之臨界  $V_A$  值  
隨氫氣濃度增加而增加，顯示摻混氫氣比例越高，內層擴散火焰穩定性越佳。

#### 五、致謝

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# 柴油/生質柴油噴注之特徵-BOSCH 6M60 為例

## Characterization of Diesel/Bio-diesel Injection: BOSCH 6M60

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### 摘要

本研究以一個由 BOSCH 公司所生產 6 孔柴油共軌噴嘴在使用柴油與生質柴油後的影響，並且與一個全新的噴嘴進行比較，而實驗的進行則是使用雷射光學技術分析噴霧特徵為主要的目的。藉由雷射時間的準確操控可以用來分析噴霧延遲、噴霧速度、噴霧張角以及滲透深度等重要噴霧資訊，其中噴霧速度使用數位影像處理進行影像邊緣強化搭配數位化雷射顆粒影像測速法進行速度分析，相關的結果可以用為未來使用生質燃料時，引擎性能變異原因之參考。

關鍵字：生質柴油、雷射診測、數位化粒子影像測速、噴霧

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## Abstract

The objective of this study is to compare the spray characteristics of aged injectors using diesel/bio-diesel with a new one based on the spray pattern and performance of the injectors. The experiments are conducted using a 6-holes injector manufactured by BOSCH. The fuel is injected into the chamber and is measured using laser-based imaging technique. The measured spray characteristics in environments of different pressures can suitably provide bases for diesel engine modifications and feasibility verification for bio-diesel application.

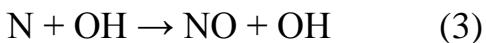
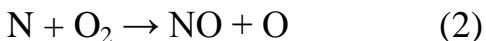
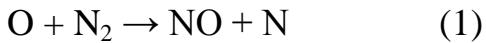
Keywords: Bio-diesel, laser diagnostics, PIV, spray

## 一、前言

生質能(Biomass Energy 或 Bio-energy)，係指利用生質物，經轉換所獲得之可用能源，如電與熱。根據國際能源總署 (International Energy Agency ; IEA) 2008 年統計資料顯示，生質能為全球第四大能源，供應全球約 10.0% 的初級能源需求，同時也是目前最廣泛使用的一種再生能源，約占世界所有再生能源利用的 78%<sup>1</sup>。根據工研院的資料指出：2010 年國內生質柴油，使用 B2 作為摻配時，可以達到 70,000 公秉的使用量，預計於 2015 年使用 B5 並達到 250,000 公秉的使用量。使用生質燃料於柴油引擎中並非完全沒有缺點，例如：熱值略低、凝固點比石化柴油高而容易雲化、微生物孳生、與潤滑油產生交聯影響等問題，但是在噴嘴裂化上卻尚無很確切的研究顯示其對噴嘴的影響，這些影響包含腐蝕、積碳，以及磨損等，這些效應都會直接影響到噴霧的功能與特性。不僅如此，在基礎燃燒特性上更缺乏完整且系統性的評估，這些相關的資訊更是許多從事內燃引擎研究工作者所需要的資料庫。

以食物作物為來料源的第一代生質燃料，因有與糧食競爭的疑慮，所以其可行性與道德風險需要進一步的考量。非糧料生質柴油的種類包括來自：廢棄食用油回收、蓖麻籽、痲瘋樹甚至是來自於藻類，廢棄食用油回收因來源穩定度不足，且廢棄食用油尚有作為肥皂之用途，因此產量不大，另外兩個較具有潛力的則是蓖麻油以及痲瘋樹油，這兩種油類因本身分別具有蓖麻毒素以及痲瘋樹毒蛋白，所以使用上必須考慮燃燒不完全可能殘留的毒性。藻類部份雖然產油量很高，但在基因工程、養料供應等技術仍然相當的昂貴，因此仍在積極開發中。使用以糧料為基礎的生質柴油會讓許多原本生產食品的農地改種植經濟作物，很可能造成糧價上漲，威脅貧窮人口；如果因燃料需求而開墾新的農地則會破壞生態，其所造成的二氧化碳排放則是一個不環保且不具道德的措施。雖然使用生質柴油可以達到減少二氧化碳排放的效果，但是使用生質柴油時，其氮氧化物的排放卻有增高的趨勢，其原因來自於成份中含有氧原子分子以及較高燃燒溫度所致，不過生質柴油燃燒時所產生的 CO 與 HC 的排放卻有明顯的減少。

在使用生質柴油時，氮氧化物卻增加的趨勢，在大自然界中自然界閃電等高溫所產生的氮氧化物( $\text{NO}_x$ )之外，人類所產生的  $\text{NO}_x$  約佔百分之七十，而產生的過程大部分發生在各種內外燃機中所產生；而  $\text{NO}_x$  的來源主要是來自於(1)空氣中氮氣的參予反應以及(2)含氮物質的燃燒(生質能)。一般來說對於不含氮燃料的反應中， $\text{NO}_x$  的生成主要可以分成兩種：(一)Thermal NO；(二)Prompt NO。高溫的燃燒過程中，因高溫產生的  $\text{NO}_x$ (Thermal NO)主要是以 Zeldovich<sup>5</sup> 的生成機構來表示：



其中的關鍵速率在於氮氣三鍵的裂解，該鍵的裂解能量需要 941KJ/mole<sup>6</sup>，而其裂解溫度必須大於 2000K 以上，也因此當燃氣溫度高於 2000K 以上時 Thermal NO 會急速增加。Miller<sup>7</sup> 在先前的研究中提到，以甲烷為燃料且  $\phi$  值於 1.37 之情況下燃燒溫度約為 1800K，此時 Thermal NO 的產生量可以被忽略；其研究更說明 Thermal NO 僅在  $\phi$  值介於 0.8 至 1.0 之間為最大。因為 Thermal NO 的反應速率比較慢所以大部分 Thermal NO 主要產生於後火焰(Post-flame)中，所以無法以 Zeldovich 的反應機制來描述主要反應區中快速產生的 NO，而 Fenimore 在 1971 年認為在主反應區中快速產生的 NO 必定與碳原子或碳氫原子有關聯，故提出關於在主要反應區中快速產生 NO 的反應機制，也就是說碳氫燃料的火焰中透過以下的反應式：



再轉化出 NO 的反應機構。在 Bartok<sup>8</sup> 的文獻中發現到當  $\phi$  值大於 0.9 之後 NO 主要由 Fenimore NO 所主導，而且當  $\phi$  值接近 1.2 時達到最大值當  $\phi$  值大於 1.2 之後 Fenimore NO 逐漸減少。Fenimore NO 的產生機構屬於 Prompt NO 中重要的一種反應機構，但是 Prompt NO 還包含了另外兩種：Superequilibrium NO 與  $\text{N}_2\text{O}$  轉化 NO，前者係當  $\phi$  值小於 0.8 時也就是 Thermal NO 與 Fenimore NO 的量均很少時才顯得重要，而後者主要是針對燃煤或者燃重油的系統中才顯得重要<sup>9</sup>。一般而言，欲降低  $\text{NO}_x$  的方式主要有兩種：修改燃燒技術，例如：分層燃燒(Staging Combustion)<sup>10</sup>，回燒(reburning)<sup>11</sup> 與漩渦燃燒(Swirling Combustion)<sup>12-14</sup>；以及後處理技術：例如觸媒後處理。

本研究與生質柴油適用性有關，搭配財團法人車輛研究測試中心之實車道路與引擎耐久測試，利用 Malvern 雷射繞射粒徑分析儀、雷射系統、高速攝影機、高壓封閉艙體與時序控制器等，進行分析生質柴油之噴嘴噴霧特徵與噴霧型態(包括噴霧延遲、貫穿距離、噴霧速度、噴霧張角、粒徑分佈與 PIV 噴霧流場等分析)，評估長期使用生質柴油噴油嘴內腐蝕以及積碳對於噴霧之影響研究。

## 二、研究方法

過去本研究團隊已經建立成熟的雷射噴霧診測技術[19]，本研究係採用雷射進行相關噴射時間的掌控，對於攝影來說可以取得較為清晰的影像。

### 2.1 燃料供應系統

本研究使用一共軌柴油用加壓泵進行增壓，該加壓泵與一 3hp 變頻馬達連結，可適當控制轉速以進行相關試驗，如圖 1 所示為高壓柴油供應裝置與高壓環境模擬艙之連結。除此之外，為了有效控制噴油壓力，本研究使用壓力感測以及快速電磁閥進行壓力操控，如圖 1 所示為共軌系統之壓力感測與快速電磁閥。



圖 1、雷射系統與高壓艙本體配置

### 2.2 雷射與光學系統

目前應用於定性視流以及數位化粒子影像測速技術所使用的雷射光源為美國 NewWave 公司生產之 Solo III-15 雷射，該雷射系統是專供粒子影像測速儀之照明專用雷射，如圖 2 所示，雷射頭中含有雙管鈸鐠石榴石雷射，並且由雙套閃光燈以及控制電路所激發，雷射之能量為 50mJ/pulse，每一管雷射擁有獨立的觸發點亮線路，可以分別受到不同訊號的控制，足以應付空氣流場中的數位粒子影像測速技術以及噴霧視流之用。

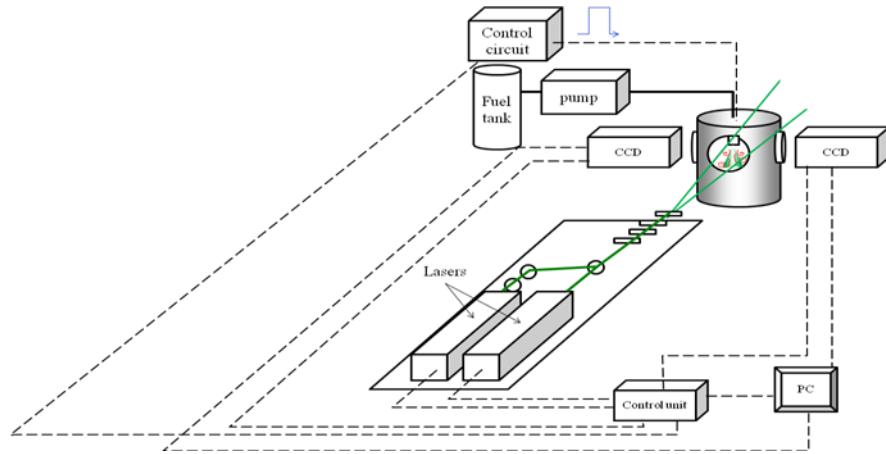


圖 2、雷射系統與高壓艙本體配置

進行相關影像視流擷取時，必須使用適當的影像擷取裝置，而且在執行研究的工作中包含有速度流場的定量，所以非一般 CCD 可以勝任。Pixel fly 為德國 Cooke 公司所生產之 CCD，其造型與尺寸非常精簡，非常適用於小空間之配置，其中可指定選配俱備畫幅內分(interframe)功能，使其具備顆粒影像測速技術之取像功能，該機型屬於較為經濟的機種，其畫幅內分的時間能達到  $10\mu s$ 。影像擷取後儲存於電腦記憶體中，配合各種數位影像處理技術來取得噴嘴所需釐清之各種物理量。

粒徑方面則是應用 Malvern 雷射繞射粒徑分析儀，進行流體霧化液滴平均粒徑之量測，Malvern 雷射繞射粒徑分析儀可以快速即時之方式量測噴霧流場中，區域之 SMD 分布。一般 Malvern 雷射繞射粒徑分析儀之量測原理是由一部氦-氖(He-Ne)雷射產生紅光雷射，藉著光束擴大器將雷射光擴束為直徑 9mm 之平行光後，穿過噴霧流場中之液滴產生繞射光，再經由傅立葉透鏡收集液滴產生之繞射光後，通過光檢測器。光檢測器是由 31 道環狀光強度感應器組成，可偵測不同粒徑分布產生之繞射光，光訊號經由固態晶體轉換成電子訊號，再經放大後透過類比數位轉換器轉換成光訊號傳送至電腦上，由軟體來執行處理。當使用之傅立葉透鏡焦距為 300mm 時，其有效量測粒徑範圍為  $1.5\mu m \sim 564\mu m$ ，其粒徑量測範圍與光學透鏡焦距及長度有關。

### 2.3 分析方法

噴霧延遲、噴霧型態、滲透深度、與噴霧張角主要是根據噴霧攝影所得，本研究團隊均是使用雷射系統作為攝影光源，其主要的優點在於雷射的高發光時機控制以及極短的脈衝時間，在極短的脈衝時間內，噴霧型態的現象可以被定瞬凝固，而噴霧延遲主要是利用雷射準確的出光時機尋找燃料從管孔逸出的時機，噴霧延遲主要跟燃料的壓力以及環境壓力有關，但是環境壓力的效應不是很明顯。根據 SAE J2715 文件[16]，噴霧型態攝影共計有三種方式可以呈現：1.背光照明法；2.體光照明；3.光頁照明。這三種方法均可以使用雷射作為光源，再依照光源、光學鏡片、以及攝影裝置的配置差異加以實現，因此均能善盡運用脈衝雷射

的優點。在噴霧型樣分析方面，將使用液滴對雷射光束所產生 Mie Scattering 訊號加以估算。滲透深度、噴霧張角的估算也將依據 SAE J2715 [16] 規範加以定義如前所述，目前尚無針對柴油噴嘴所定義之標準規範，因此引用的均是缸內直噴噴嘴所使用之 SAE J2715，柴油噴嘴與汽油缸內直噴噴嘴的最大差異在於噴嘴孔的配置方式，因此在噴霧定義上並沒有影響，唯有影響的是在於噴嘴原點座標定義以及多孔噴嘴的量測方式。

PIV 粒子影像測速[17、18]的基礎原理是利用顆粒或者液滴在空間中受到雷射光照明後所產生的米散射(Mie scattering)，米散射是一種彈性散射過程，當粒子直徑與入射光波長的比值遠小於 1 ( $d/\lambda \ll 1$ ) 時稱之為芮利散射(Rayleigh scattering)，當前述的比值非遠小於 1 時則稱之為米散射。米散射的強度與粒子的尺寸有很大的關係，其強度為直徑的平方成正比。懸殊的噴霧尺度將會造成粒子散射光的極大差異，當雷射能量設定較高以觀察微細顆粒時，較大的粒子所產生的散射光強度很容易使 CCD 上被投影的像素(pixel)飽和(saturated)，除此之外飽和區的像素(pixel)也容易產生電子溢出而產生影像暈染(blooming)，所以大顆粒區域的曝光與大顆粒的實際尺度都會造成較大的誤差。雖然較大的顆粒會有前述的問題，不過在速度量測上並不會造成影響，因為速度的分析是根基於影像中所擷取審視窗(interrogation window)中的交叉關連分析(cross-correlation)統計而得。對於缸內直噴引擎用燃油噴嘴噴霧而言，由於噴霧上游處之燃油為液膜型態，因此使用以粒子影像測速方法量測該處較無意義，而本研究是使用該技術量測噴霧下游處之速度分佈，且噴霧之液滴速度以粒子影像測速技術來量測屬直接量測，意即量測到的數據即為噴霧之液滴速度，而該速度特徵將可作為內燃引擎燃燒室與活塞頂設計之基礎。影像分析系統在軟體上是以 Matlab 為開發平台的程式，該程式自從開發以來以經歷 7-8 年的修改，並且成功的應用於燃燒場中速度的分析與處理[17, 19-20]，透過 Matlab 開發更可以快速的建立訊號處理的數學模型並且可以快速方便地轉換成效率較高的 C/C++ 語言程式。影像取得後必須針對影像的特性進行前處理，前處理完成後進行影像分割以成為審視窗，在訊號處理上主要使用交叉關連分析加以處理，並進行後續處理的工作。

### 三、結果與討論

#### 3.1 噴射延遲與噴霧外型

本研究使用雷射光學法直接測定噴嘴之噴霧延遲，所謂噴霧延遲係指噴嘴接收到驅動信號後，燃料出現於噴嘴口的這一段延遲時間，噴嘴噴霧的延遲時間會影響到引擎的操控方法與策略，在噴嘴診斷中可以將噴嘴出現當成時間的原點，所有從此原點開始的現象描述均可以加註 SOI 為記號，其定義如圖 3-1 所示；另外一方面，液體出現在噴嘴口的現象如圖 3-2 所示，目前經由此方式所測得之各噴嘴噴霧延遲結果如表 1 所列。



圖 3、實驗時間控制與液體於噴嘴口出現示意圖

表 1 噴嘴延遲時間

噴嘴形式	噴射壓力(bar)	延遲時間(ms)
全新	1600	0.25
使用 B2	1600	0.25
使用 D100	1600	0.25

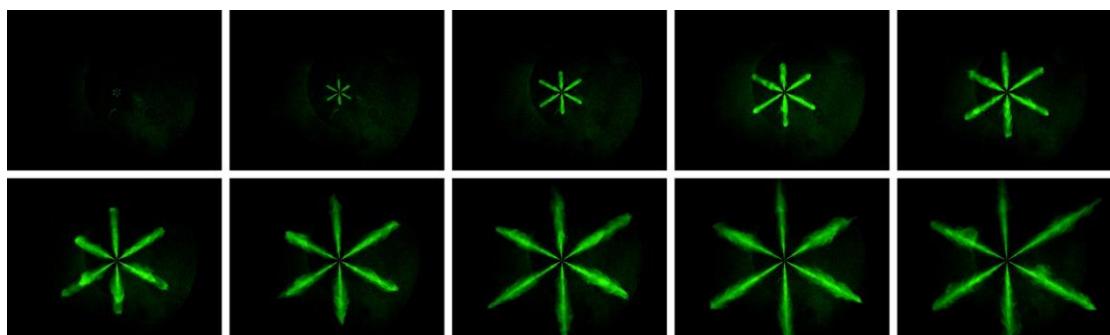


圖 4、6M60 新噴嘴在大氣環境壓力下隨 SOI 之噴霧變化

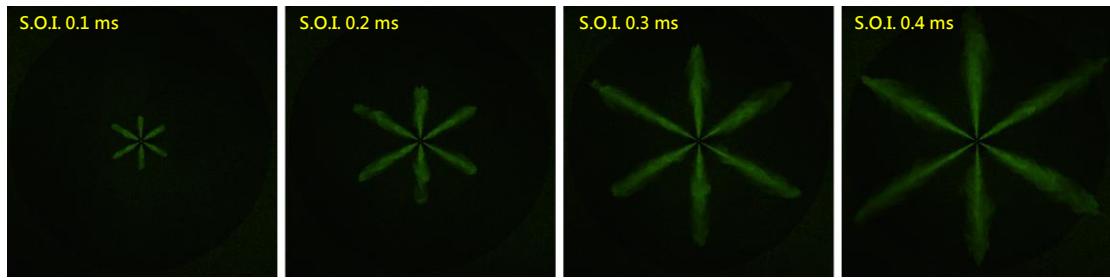


圖 5、6M60 使用過 B2 後舊品噴嘴在大氣環境壓力下隨 SOI 之噴霧變化

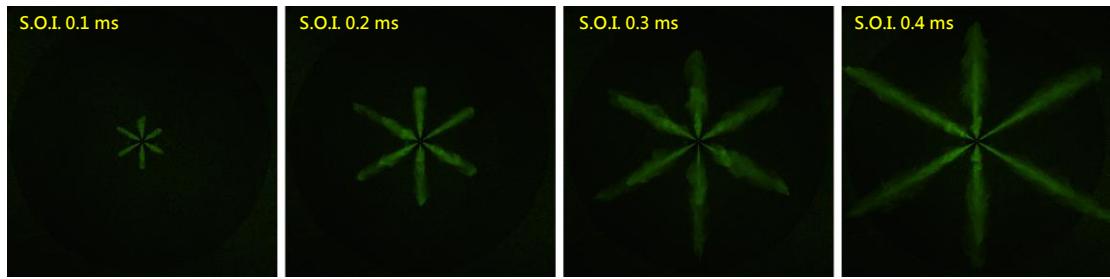


圖 6、6M60 使用過 D100 後舊品噴嘴在大氣環境壓力下隨 SOI 之噴霧變化

### 3.2 滲透深度與噴霧張角

為了判斷各噴嘴孔的滲透深度，因此將噴嘴各噴注予以代號。如圖 7 所示分別為 6M60 新品噴嘴滲透深度隨噴霧演進時間之變化，大致上來說，每個孔滲透深度的變化差異隨著時間增加而逐漸變大。如圖 8(左)為 6M60 噴嘴使用過 B2 後之滲透深度隨時間之演變，而圖 8(右)所示為 6M60 噴嘴使用過 D100 後之滲透深度隨時間之演變。新噴嘴與使用過 B2 與 D100 之 6M60 之噴霧張角之比較則展示於圖 9。

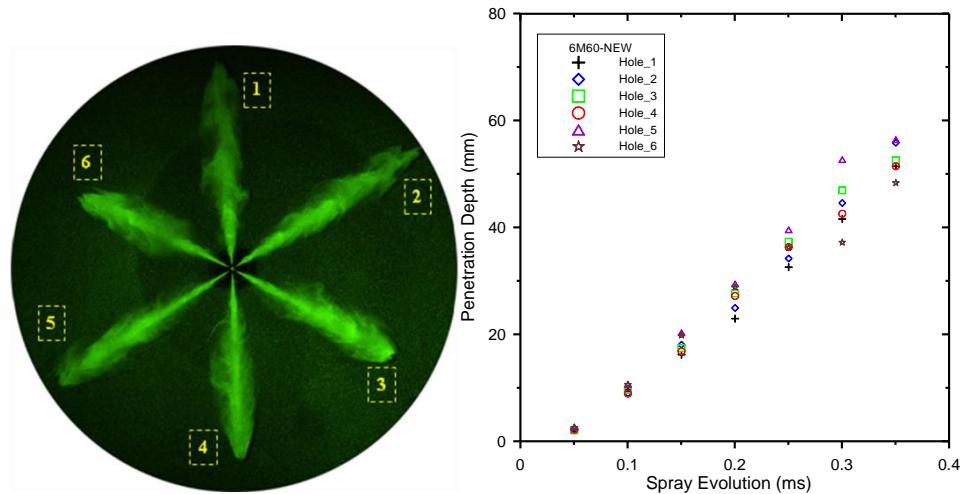


圖 7、6M60 噴嘴出口編號與新噴嘴之滲透深度隨噴霧演進時間之變化

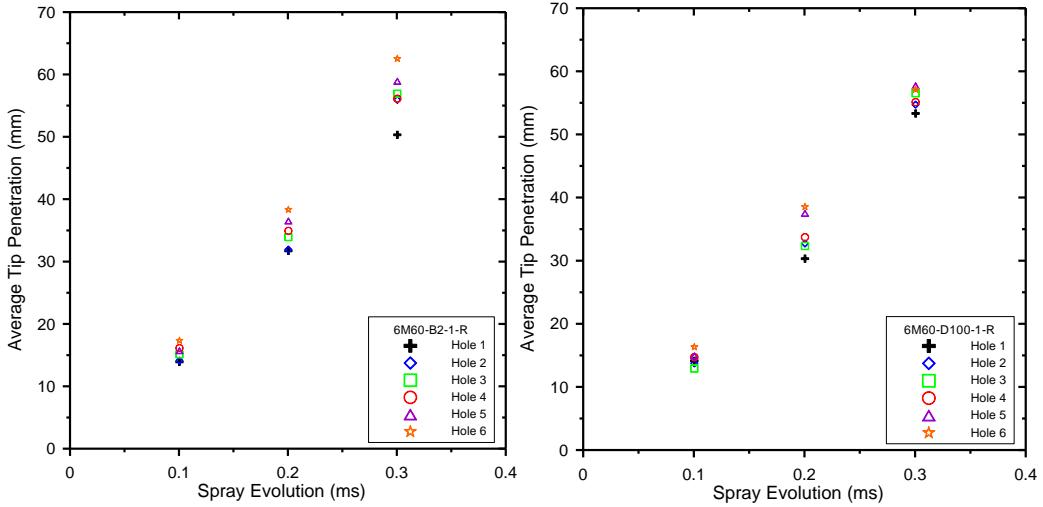


圖 8、使用過 B2(左)與 D100(右)之 6M60 之滲透深度隨噴霧演進時間之變化

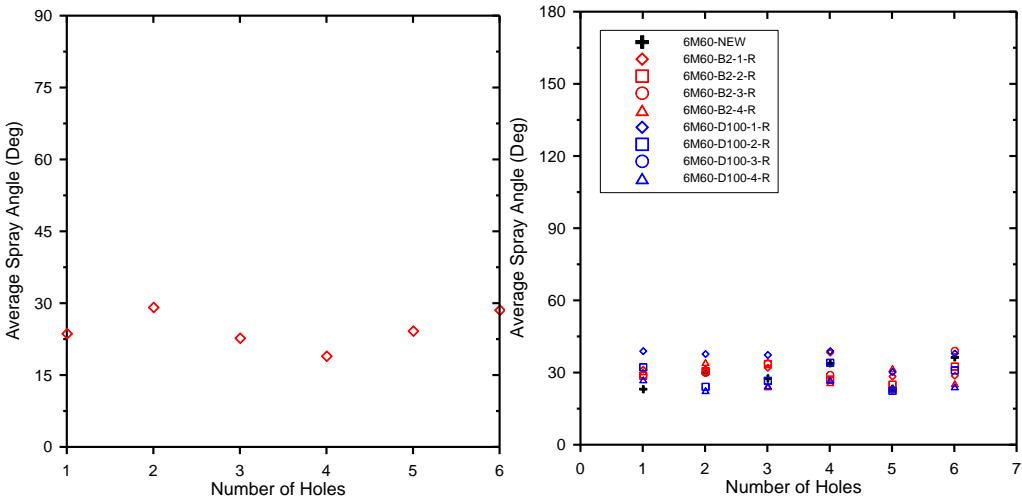


圖 9、新噴嘴(左)與使用過 B2 與 D100(右)之 6M60 之噴霧張角之比較

### 3.3 噴霧粒徑

針對新品與使用過之噴嘴進行噴霧粒徑之比較，如圖 10 所示為 6M60 新噴嘴之噴霧粒徑，圖 11 為使用過 B2 舊噴嘴之噴霧粒徑；圖 12 為使用過 D100 舊噴嘴之噴霧粒徑，從數據比較來看，噴嘴粒徑似乎沒有什麼變化，其數值都約略在  $6\text{-}7\mu\text{m}$ 。

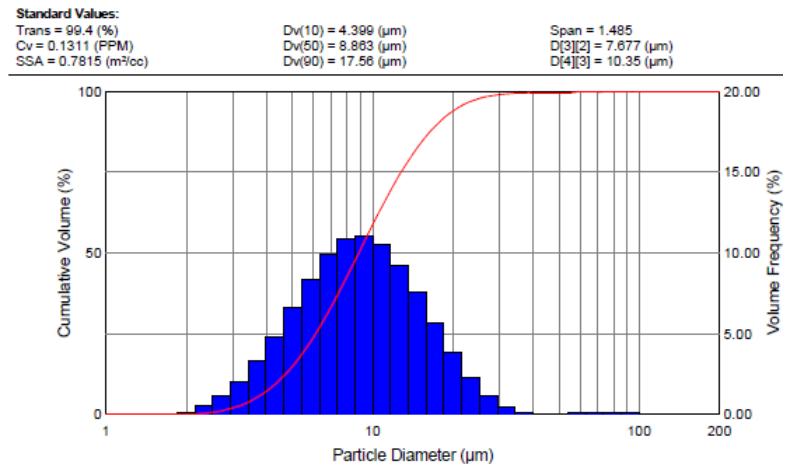


圖 10、新噴嘴之噴霧粒徑

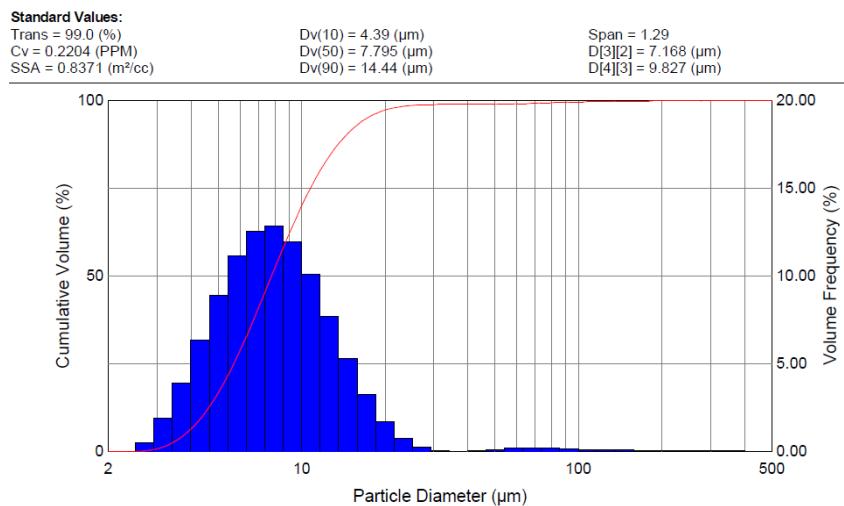


圖 11、使用過 B2 之噴霧粒徑

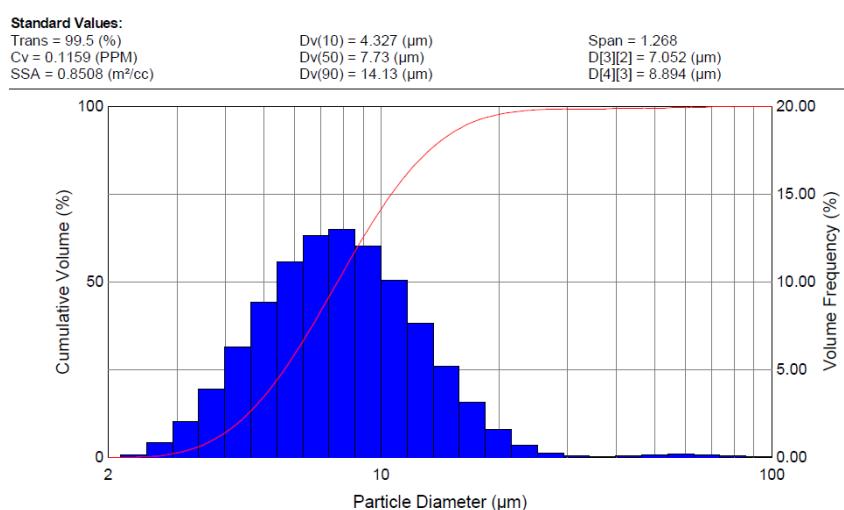


圖 12、使用過 D100 之噴霧粒徑

#### 四、結論

本研究應用雷射診測影像技術進行生質柴油與石化柴油混合燃料應用於噴嘴使用後進行噴霧特徵探討，噴嘴長時間使用的燃料分別有 D100 與 B2，解析噴嘴之噴霧延遲、噴霧張角、滲透深度與噴霧粒徑等重要資訊。根據有限的結果顯示：使用 B2 與 D100 之改變相當微小，因此可以預計，當柴油引擎使用 B2 與 D100 燃料時，經過特定時間後並不會產生巨大的影響。

#### 五、致謝

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# 應用主要組分分析法對生質柴油燃料替代物動力學 機理的精簡

## Kinetic Mechanism Reduction Using a Principal Component Analysis for a Biodiesel Fuel Surrogate Mechanism

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### 摘要

有關生質柴油動力學機理的研究是基礎研究及工業發展領域的研究熱點。如何能夠在不損失預測精準度的前提下對詳盡的動力學機理進行精簡，成為影響用來模擬反應流和內燃機工作、包含詳細化學動力學的計算流體力學及多區域模型發展的主要因素。本文的目的在於呈現作為有著與生質柴油燃料類似結構的最小酯分子—丁酸甲酯(MB)的一個新的熱分解機理。基於主要組分分析 (PCA)，原本由 267 種中間體和 2054 個反應組成的機理被按不同程度進行精簡，以用來了解如何在機理的規模大小和它的預測能力之間進行權衡。主要組分分析法會對每一個反應進行敏感度分析，因而應用此方法能夠從原反應機理中去掉那些就選定的反應器操作條件而言並不重要的反應。我們從一組精簡的丁酸甲酯機理中精選出了一個計算成本合理、同時又能夠很好地複製從 1200K 到 1700K 的溫度範圍內激波管實驗的結果。

關鍵詞：主要組分分析，機理精簡，丁酸甲酯，熱分解

### Abstract

Kinetic mechanisms for biodiesel are of great interest for both fundamental research and industrial development. Understand how to reduce detailed mechanisms without losing the prediction accuracy is the main challenge to develop computational fluid dynamics and multi-zone models with detailed chemical kinetics for simulations of reacting flow and internal combustion engines. The aim of this paper is to present a novel reduced pyrolysis mechanism for methyl butanoate (MB), the smallest ester with a structure similar to the ones of biodiesel fuels. Based on the Principal on the

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Principal Component Analysis (PCA), the original mechanism composed of 267 species and 2054 reactions has been reduced to various levels for proper understanding the trade-off between the mechanism size and its prediction capability. The PCA, which uses a sensitivity analysis for each reaction, is able to eliminate unimportant species and reactions from the original mechanism based on the criteria that weight the contribution of each reaction to selected reactor operating conditions. From a pool of reduced MB mechanisms, we have been able to identify a computationally economical reduced mechanism that is able to well reproduce shock tube experimental data over a wide temperature range of 1200 to 1700 K.

Keywords: Principle component analysis, Mechanism reduction, Methyl butanoate, Pyrolysis.

## 1. Introduction

The great concerns over the disadvantages of fossil fuels, including diminishing resources, greenhouse gases and other pollutants, have raised the strong interest in the application of alternative fuels as a renewable source of energy for road transport, aviation, and electricity generation. One potential solution as an alternative fuel is biodiesel, which is considered as CO<sub>2</sub> neutral. Most commonly, biodiesel produced from local crops, such as soybeans, rapeseed, palms, olives, or sunflowers is comprised primarily of fatty acid methyl esters (FAMEs, -C(=O)-O-CH<sub>3</sub>) [1, 2]. In terms of power system costs, biodiesel has the advantage of being generally compatible with current combustion technologies and fuel infrastructure [3, 4].

Chemical kinetic modeling, which enables detailed chemical analyses on pyrolysis and oxidation characteristics of fuels, is one way to greatly increase the efficiency of studying a variety of biodiesel-powered combustors. Because of the biodiesel complexity and molecular size, direct kinetic modeling of its combustion has been historically unfeasible. Alternatively, a common practice is the use of surrogate molecules or blends that match the characteristics of real fuel but have much lower computational requirements [5]. For the past decade, biodiesel surrogate mechanisms have been widely used to model fuel reactions in simplified reactors such as, jet stirred reactors, shock tubes and rapid compression machines [6]. For the past several years, methyl butanoate (MB), as shown in Fig. 1, is the most commonly used surrogate for studies of biodiesel fuel combustion [7]. However, species and reaction numbers in developed biodiesel surrogate mechanisms are generally too large to be numerically embedded into computational fluid dynamics engine models that provide detailed description of in-cylinder gas motion and detailed reactions on a local level.

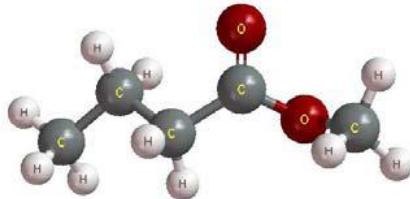


Fig. 1. Structure of methyl butanoate

One way to address the issue of size and computational cost is the mechanism reduction. Skeletal reduction, which is typically the first step in a comprehensive scheme, is to remove unimportant species and reactions in achieving certain computational targets, such as species concentrations, temperatures or pressures. The methods based on the strategy of skeletal reduction include sensitivity analysis [8, 9], principle component analysis (PCA) [10, 11], Jacobian analysis [12], direct relation graph (DRG) and other DRG-based methods[13, 14]. These advantages and disadvantages of the reduction processes have been described and summarized in the review article of Lu and Law [13]. Among the methods above, PCA is easier to apply, because principle components offer an effective means for extracting useful kinetic information from derived sensitivity tables.

Although reduced mechanisms for biodiesel surrogates have been extensively proposed in the past years [15-17], relatively little is known about how reduced mechanisms produced by skeletal reduction are influenced by the selected targets such as, initial temperature, pressure, and concentrations. In the present study, we attempt to develop a reduced mechanism for MB pyrolysis mechanism using principle component analysis with difference initial temperatures in a shock tube study. The findings of this study extend the applicability of the PCA to reduction for the mechanisms of oxygenated fuel pyrolysis.

## 2. Methodology

### 2.1 Numerical Details

In this study, the principle component analysis is performed using KINTECUS and ATROPOS [18]. The details of the numerical method is given in references [10, 11]. In the first step, the original mechanism is used in KINTECUS to obtain each species concentration as a function of time under isothermal and isobaric conditions. The systematic ordinary differential equations for each species concentration are numerically solved with given initial conditions. In each time step, where concentrations are solved, a matrix of Normalized Sensitivity Coefficients (NSC) is generated by the equation:

$$NSC = \left( \frac{\partial \ln[A]}{\partial \ln k_j} \right) \quad (1)$$

where  $[A]$  is the concentration of each species ( $\text{mol}/\text{cm}^3$ ) and  $k_j$  is the rate coefficients for reaction each reaction  $j$ .

Each calculated NSC matrix ( $S$ ) is used to determine the degree of influence of reactions on each species. The produced NSC matrices,  $S_1$ ,  $S_2$ ,  $S_3\dots S_n$ , are concatenated to form a big matrix, BS. The removal of each reaction requires the satisfaction of the inequality constrains:

$$\lambda \leq m \times q \times \text{safe values} \quad (2)$$

where  $\lambda$  is the eigenvalues of  $BS^T \times BS$  (the superscript T is transpose),  $m$  is the species number and  $q$  is the time points. The safe values are the threshold numbers that we use to generate different levels of skeletal mechanisms.

## 2.2 Parameter Selections for Mechanism Reduction

The MB mechanism of the University of Michigan [19] that is composed of 267 species and 2054 reactions is reduced by eliminating reactions and species that have less influence on MB pyrolysis in a shock tube [20]. The initial conditions used for calculating NSC matrices are 0.5% mole fraction of MB in Ar. at The pressure of 1.5 atm is selected for two temperatures ( $T = 1200$  and  $1600$  K) based on the experimental conditions used in the study of Farooq et al [20]. The safe number(threshold number) employed in this study is in the range of 10-14 to 17.

## 3. Results and Discussion

In the present study, we attempt to identify a reduced mechanism that achieves the best trade-off among the mechanism complexity, mechanism accuracy and the range of temperature applicability. We first analyze the relationship between the size of skeletal mechanisms and safe numbers. Then, these generated skeletal mechanisms are employed to reproduce existing experimental results for MB pyrolysis in a shock tube. The discrepancies between experimental and predicted values for different skeletal mechanism are presented accordingly.

Fig. 2 and 3 show the presence of reaction and species numbers, respectively, in different skeletal mechanism generated by a wide range of safe numbers. As expected, the increase of safe number results in the decrease of mechanism complexity. It is seen that the complexity of the mechanism against safe values drops faster at  $T = 1200$  K when compared to that at  $T = 1600$  K. This observation indicates that the reaction connectivity is strongly reduced at relatively low reaction temperatures. At safe value of  $10^{-8}$ , where the mechanism complexity is seen to be reduced significantly, approximately 75% reactions and 33% species in the original mechanism are removed. This result implies that the 25% reactions remained are strongly bound with about 67% of the total species in the original mechanism. In the

range, where the safe value is increased from  $10^{-8}$  to  $10^{-4}$ , the removal of unimportant species and reactions becomes less effective. Although the species numbers are further significantly reduced at the safe number greater than  $10^{-2}$ , the accuracy to predict MB pyrolysis (produced at safe number  $> 10^{-2}$ ) may dramatically drop in these skeletal mechanisms.

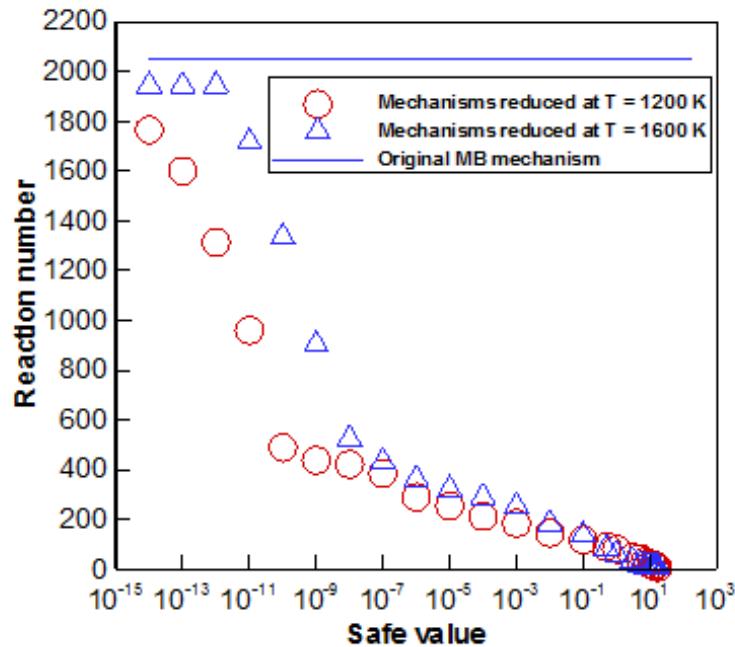


Fig. 2. Reaction number of each reduced mechanism derived from different safe values.

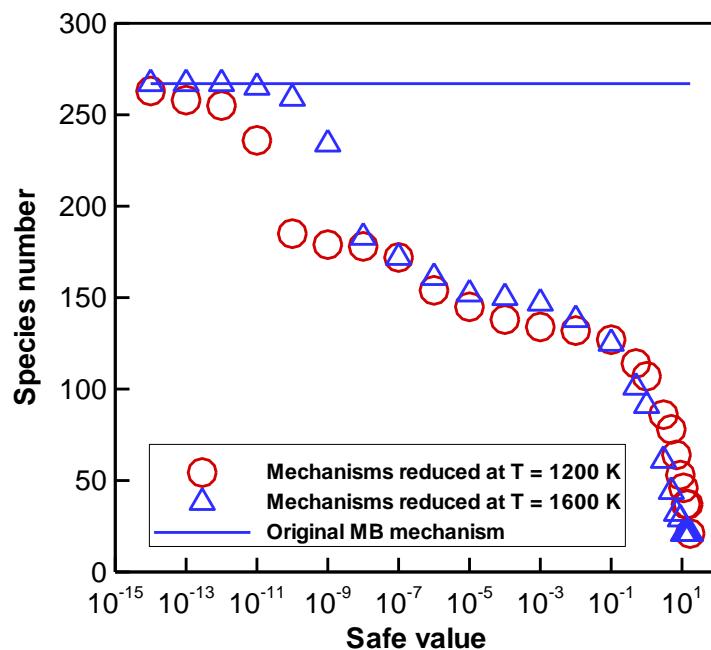


Fig. 3. Species number of each reduced mechanism derived from different safe values.

In order to test the mechanism accuracy and range of temperature applicability, ten skeletal mechanisms selected (five from each temperature in Fig. 2-3) are employed in KINTECUS to reproduce transient CO formation from MB pyrolysis in a shock tube experiment. These skeletal mechanisms are chosen by the safe numbers of  $10^{-14}$ ,  $10^{-9}$ ,  $10^{-5}$ , 0.5 and 17, respectively. The predicted results for CO formation at T of 1700 K are shown in Fig. 4 and 5. In Fig. 4, where the mechanisms are reduced at the condition of T = 1600 K, the most computationally economic and acceptable skeletal mechanisms is the case with the safe number of  $10^{-5}$ . This skeletal mechanism has 152 species and 321 reactions. Unlike the results in Fig. 4, the skeletal mechanisms which are produced at T = 1200 K (Fig. 5) indicate that favored mechanism is seen in the case with the safe number of  $10^{-9}$ . The complexity of this skeletal mechanism, however, is not efficiently reduced. Further, the mechanisms in Fig 4 and 5 are used to predict CO formation from MB pyrolysis at T of 1250 K and the results are presented in Fig. 6 and 7, respectively. As seen in Fig. 6, the predicted results from the skeletal mechanisms (created at T = 1600 K) reveal that the case with the safe number of  $10^{-5}$  is consistently reliable for predicting CO formation at low temperature. In Fig. 7, where the mechanisms are reduced at the condition of T = 1200 K, the most favored skeletal mechanism is the case with the safe value of 10-5. This mechanism that is composed of 145 species and 254 reactions shows a significant discrepancy when used to predict the CO formation at T = 1700 K. In this study, we observe that safe values need to be reduced to obtain reliable skeletal mechanisms created at relatively low temperature.

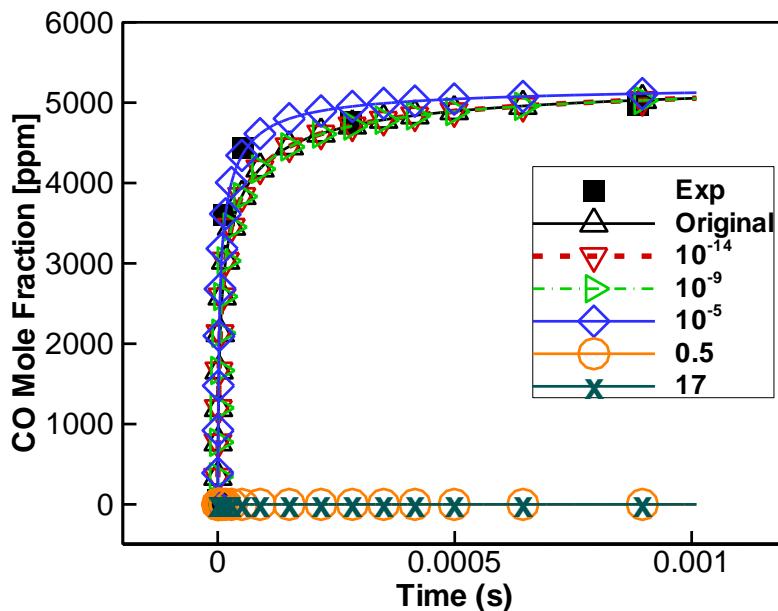


Fig. 4. CO mole fraction versus time in shock tube simulation at  $T = 1700$  K using reduced mechanisms derived from different safe values at  $T = 1600$  K.

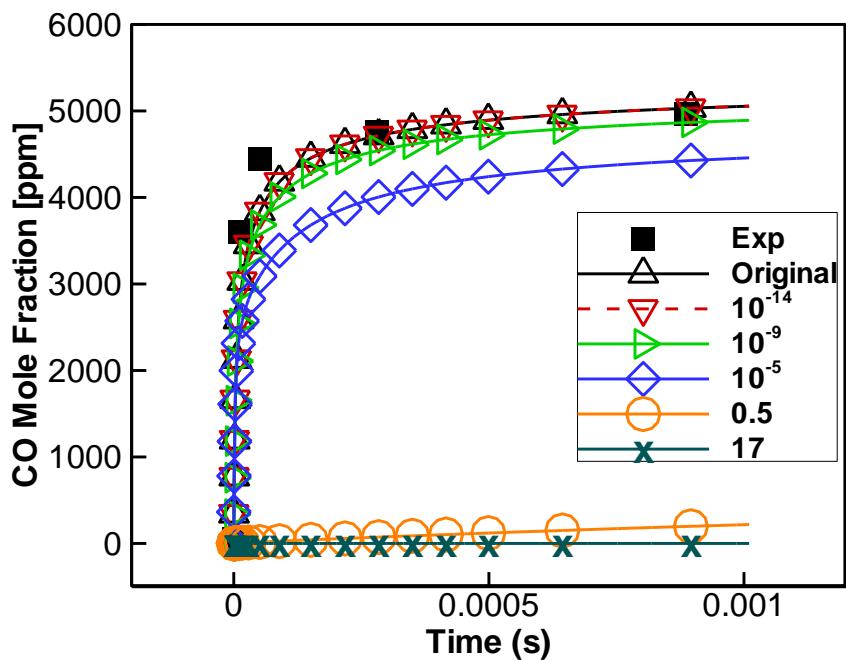


Fig. 5. CO mole fraction versus time in shock tube simulation at  $T = 1700$  K using reduced mechanisms derived from different safe values at  $T = 1200$  K.

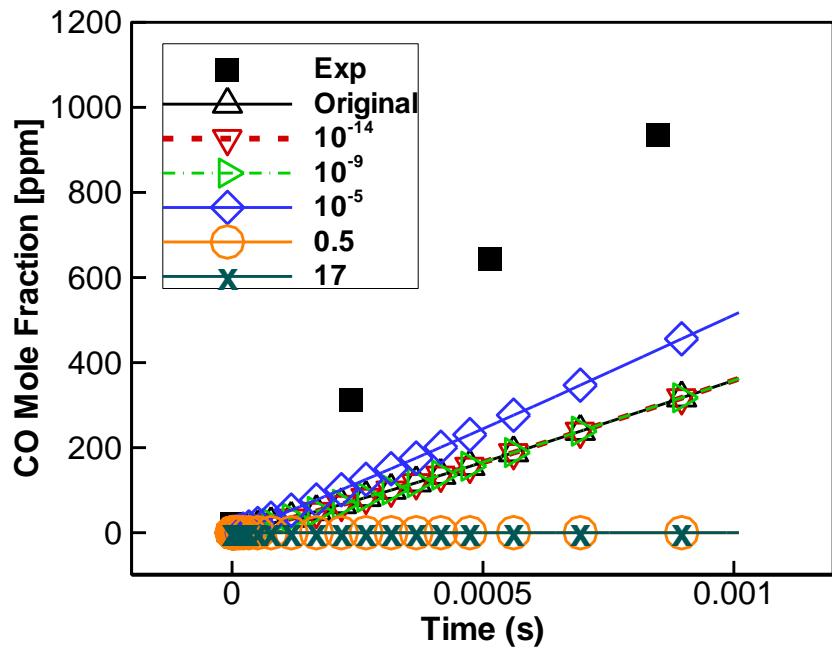


Fig. 6. CO mole fraction versus time in shock tube simulation at  $T = 1250$  K using reduced mechanisms derived from different safe values at  $T = 1600$  K.

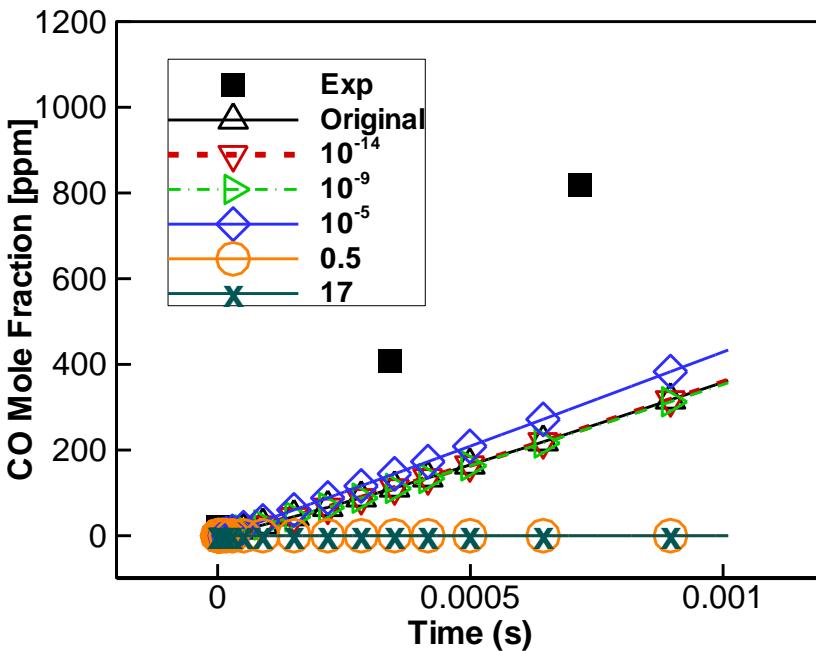


Fig. 7. CO mole fraction versus time in shock tube simulation at  $T = 1250$  K using reduced mechanisms derived from different safe values at  $T = 1200$  K.

#### 4. Conclusions

The objective of this study is to identify a skeletal mechanism that is able to predict CO formation from methyl butanoate pyrolysis at temperatures in the range of 1250 K and 1700 K. This study is achieved by a Principle Component Analysis applied to an existing mechanism in the literature. The most favorable skeletal mechanism generated at 1600 K is able to well reproduce the CO formation at both 1250 and 1700 K, whereas the most preferable skeletal mechanism created at 1200 K shows no consistency in terms of the accuracy for the prediction of CO formation at different pyrolysis temperature. Future work will be dedicated to validating the reduced mechanisms with different products formed in methyl butanoate pyrolysis.

#### 5. Acknowledgments

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# 熱電共生裝置之能源回收率影響研究

## Energy Retrieve Efficiency of Thermal and Electric Co-generation Device

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### 摘要

本研究之目的在設計一套熱電共生裝置，並評估此組共生裝置油品的能源回收率。系統以一具小型機車引擎帶動一個發電機，並將引擎所產生的熱，加以回收儲存；發電機產生之電，可以利用電瓶儲存或連結到一般家庭使用的電器上，此系統研究目標有兩個：第一為汽油之熱電共生裝置，如何提高其能源回收率；第二為導入車用液化石油氣燃料，評估能源回收效率。車用液化石油氣與汽油相比，車用液化石油氣售價較汽油低廉，相同單位重量下能提供較高的熱值。

研究結果顯示，離形機之能源回收率，於轉速 3600rpm、發電量 3.2 kW，回收效率達 59.8%；無發電負載時，加熱時間較長熱傳損失相對較多，能源回收率約僅有 23.4%；為了提升能源回收率，在設備上裝填了隔熱材料，減少回收廢熱時的熱傳損失，實驗結果顯示，在汽油燃料在 3000rpm、發電量 3.2 kW 時，運轉 26 分 51 秒，便足以提供足夠的能量，來供給一般的家庭做使用；能源回收效率更可以達到 71.4%；受排氣熱損失之影響，在低負載時，能源回收率約 20%，隨負載增加而提升，在高負載時會升至約 71.4%，比一般汽油引擎之 25%~30% 热回收率，高出約 40% 的利用率；在車用液化石油氣 4000rpm、負載值 3.2kW 時，運轉 24 分 30 秒，便足以提供能量；在 3600rpm、負載值 3.2kW 時能源效率可達到 63.1%，仍然比一般汽油引擎高出約 30% 的熱回收率。

關鍵字：熱電聯產、燃燒特性、熱交換器、能源回收率

### Abstract

In this study, the internal combustion engine cogeneration system is experimented to produce thermal energy and electrical power. The energy retrieve efficiency of this equipment will also be evaluated. The system studied consists of a commercial motorcycle engine, an electric generator and a heat exchanger to recover residual heat from the refrigeration water and exhaust gases. There are two objectives in this research. The first topic is to improve the original gasoline used thermoelectric devices. The second objective is the expanding of system's energy diversity, based on this high-energy retrieve system.

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The multi-fuel device will be developed in this system for liquid and gaseous fuels. The automotive fuels-liquefied Petroleum Gas (LPG) will be conducted through the engine fuel system modification. The energy retrieve efficiency will also measured for using gaseous fuel.

The experimental results showed that the prototype cogeneration device worked in rotate speed 3600rpm, and load values 3.2 kW, the energy retrieve efficiency was reached up to 59.8%. By the effect of exhaust gas heat loss at low load, energyretrieve efficiency was reached up to 23.4%. In order to improve the energy retrieve efficiency, the device packed with insulating material to reduce heat transfer loss. Then, it showed that the cogeneration device worked twenty-seven minutes in rotate speed 3000rpm, and load values 3.2 kW, could provide one household use and energy retrieve efficiency was reached up to 71.4%. By the effect of exhaust gas heat loss at low load, energy retrieve efficiency was reached only 20%, At high load would rise to about 71.4%, but still higher about 40% than generally gasoline heat recovery efficiency. When the gaseous fuel LPG was conducted to this system, the energy retrieve efficiency was reached 63.1% for working twenty-four minutes in rotate speed 3600rpm, and load values 3.2 kW. It was about 30% higher than generally gasoline heat recovery efficiency.

Keywords: Cogeneration System, Heat Exchanger, Energy Retrieve Efficiency, Multi-Fuel Device

## 一、前言

美國能源局(DOE)能源指出，一般高溫加熱的能量中，約有 10%~25%是藉由熱傳導的機制而造成熱損失，而對於能源的損失，將是關鍵因素之一；所以該如何減少熱損失，是降低能源消耗非常重要的課題。該局提出的儲熱三大目標：1. 發展進階工作流體與儲能(冷/熱)系統、2. 發展儲能(冷/熱)材料與系統以降低儲熱成本、3. 把儲能(冷/熱)成本及效能與聚光行太陽能系統整合。現代社會科技發展迅速，造成人類生活需求急速改變，近年來經濟快速發展，以及生活水準提昇，對於能源之需求逐年提升；然而環境保護意識日益覺醒，使得開發自產能源、能源回收及應用的重要性，逐漸成為人們當前要研究的課題。

現今全球暖化的問題越來越嚴重，南北極冰帽不斷地融化，造成海平面的升高，使得許多低窪的地區逐漸被海淹沒，而氣候的變遷使得冷暖異常，極熱極冷的氣候，導致世界各地，凍傷及曬傷的傷亡更不斷傳出，在這樣的低溫，或者是因電力無法供給，肯定會造成人類生活上的不便利，且在生命上也會造成強大的威脅，所以在熱的回收並加以應用，是個很重要的課題。。

熱傳是熱能從高溫向低溫部分轉移的過程，是一個分子向另一個分子傳遞振動能的結果。各種材料的熱傳導性能不同，傳導性能好的，如金屬，還包括了自由電子的移動，所以傳熱速度快，可以做熱交換器材料，而金屬傳導能力依次為

銀>銅>金>鋁；傳導性能不好的，如石棉，可以做熱絕緣材料；材料選擇在經濟考量上採用銅做為熱交換材料，在隔熱的部分用石綿作為隔熱材料降低熱傳損失。

LPGITA 出版之液化石油氣概論[1]，液化石油氣燃燒時所需之理論空氣量， $1\text{m}^3$  液化石油氣需要  $24 \sim 31 \text{ m}^3$  空氣量，為取得完全燃燒的效果，在使用時一般採用降壓法將液態轉換為氣態。當液態直接用於燃燒時，應採用霧化的方法使其與空氣充分接觸，以提高燃燒效率；LP-GASES HANDBOOK, 4ED, NFPA, 1995[2]，液化石油氣的主要成分為丙烷、丁烷，其熱值丙烷為  $12,034\text{kcal/kg}$ ，丁烷為  $11,832\text{kcal/kg}$ ，相較於汽油的  $11,200\text{kcal/kg}$ ，熱值上液化石油氣來的高。

## 二、實驗設備與研究方法

本實驗使用，國內知名機車廠牌，三陽機車(SYM)之 RV150 水冷引擎，作為實驗用引擎；本實驗設備配置如圖 1，引擎規格如表 3-1 (1)控制負載發電機，(2)傳動盤，(3)轉速計，(4)排氣管，(5)噴油嘴，(6)熱交換器溫度計，(7)熱交換器內銅管，(8) 數位微處理型溫度表，(9)水箱，(10)副水箱，(11) 量測油耗磅秤，(12)燃油桶，(13)燃油泵浦，(14)廢氣分析儀，(15) 負載控制面板，(16)引擎發電機電盤，(17) 電瓶。

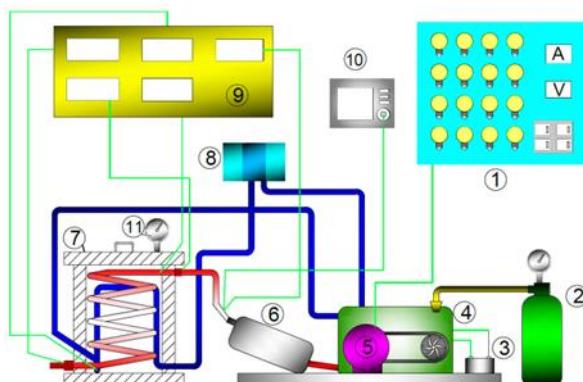


圖 1、實驗設備配置。

本實驗是以三陽(SYM) RV150 汽油引擎，實驗流程如圖 2，在轉速  $3000 \text{ rpm}$ 、 $3600 \text{ rpm}$ 、 $4000 \text{ rpm}$  時，負載值  $0 \text{ kW}$ 、 $0.8 \text{ kW}$ 、 $1.6 \text{ kW}$ 、 $2.4 \text{ kW}$ 、 $3.2 \text{ kW}$ ，來記錄溫度上升變化，為求實驗準確性及一定標準，本實驗將熱交換器內的溫度起訖時間，設定在  $35 \sim 60^\circ\text{C}$ ，記錄溫度上升的時間、廢氣汙染值、油耗、發電量、能源回收率等...值，再將數據填入至計算軟體、繪製成圖表，來計算出油品的能源回收率。

本實驗以水做為儲熱液體，此可得知水吸收之熱量來計算熱回收量；量測引擎發電機之輸出總功及發電機發電總功率可得到引擎輸出總功率；量測燃料消耗，可得到實驗過程中所輸入的總熱能；從引擎發電機及負載用發電機加上儲熱桶之

水所吸收熱量在除掉燃料供給總熱能即可得到能源回收率之數據。

儲熱桶之水吸收熱量=質量×比熱×溫度差(儲熱桶容量 66 公升)

總輸出功=引擎發電機輸出總功+發電機負載總功率

燃料供應總熱能=消耗燃料總重量×燃料熱值

能源回收率=(輸出功率+儲熱桶之水的吸收熱量)/ 燃料供應總熱能

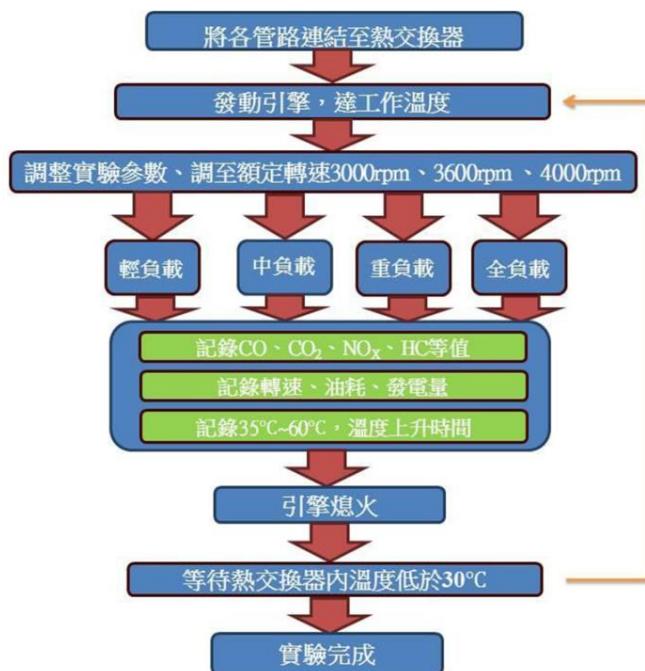


圖 2、實驗流程。

### 三、實驗結果及分析

本研究首先探討引擎排放出來的廢氣，廢氣可以提供引擎燃燒狀況之資訊，可以看出引擎狀況；如圖 3~5 所示，CO 於引擎 3000 rpm，未使用隔熱材料狀況 CO 排放量比明顯比使用隔熱材料多，液化石油氣在 CO 排放量在低發電負載的狀況下 CO 排放明顯比汽油燃料少；CO 於引擎 3600 rpm，提高轉速後，CO 排放量有逐漸減少，未使用隔熱材料狀況 CO 排放量已經趨近於使用隔熱材料後的排放量，液化石油氣提高轉速後，CO 的排放並無明顯變化；CO 於引擎 4000 rpm，整體 CO 排放量比起 3000 rpm 有明顯的減少，提高轉速後對 CO 的排放有一定程度的改善。

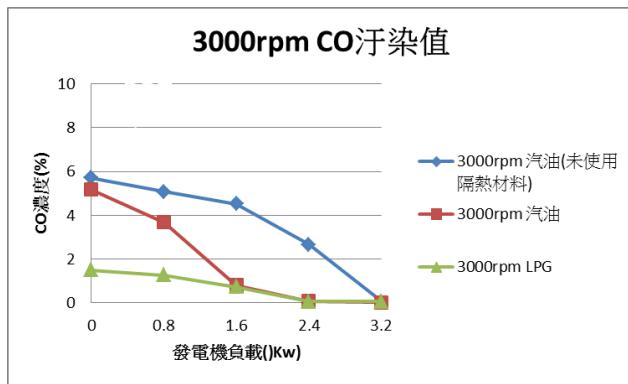


圖 3、化學當量乙 烯/氧氣焰於引擎 3000 rpm 之 CO 排放量。

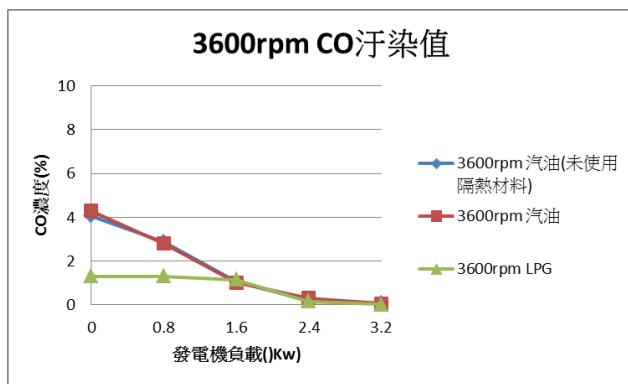


圖 4、化學當量乙 烯/氧氣焰於 3600 rpm 之 CO 排放量。

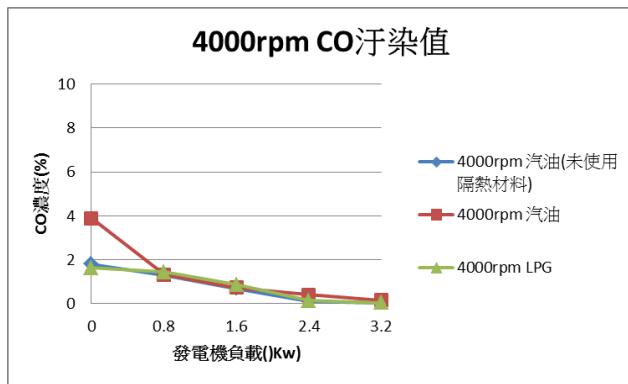


圖 5、化學當量乙 烯/氧氣焰於 4000 rpm 之 CO 排放量。

圖 6~8 所示，HC 於引擎 3000 rpm，低發電負載狀況下，使用隔熱材料汽油燃料所排放 HC 比未使用隔熱材料明顯比較高，隨著發電負載提高後，引擎燃燒狀況改善燃燒更完全，HC 排放量有明顯有比未使用隔熱材料來的少，液化石油氣的部分，隨著發電負載的提升，HC 排放有漸漸減少；HC 於引擎 3600 rpm，發電負載為零的時候，使用隔熱材料後 HC 排放比未使用隔熱材料來的高，提高發電負載後，HC 排放有明顯的下降，液化石油氣的部分，則是和 3000 rpm 差異甚

小；HC 於引擎 4000 rpm，汽油燃料所排放之 HC，明顯比 3000 rpm 和 3600 rpm 低，隨著發電負載的增加，HC 排放濃度皆低於 5 ppm。

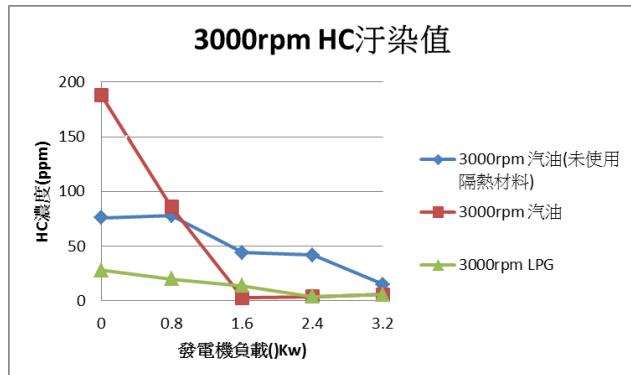


圖 6、3000rpm HC 汚染值。

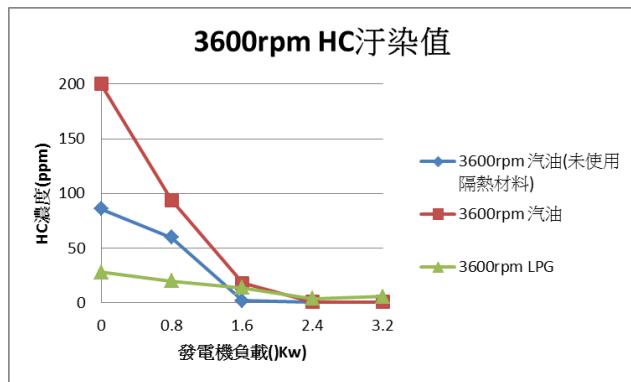


圖 7、3600rpm HC 汚染值。

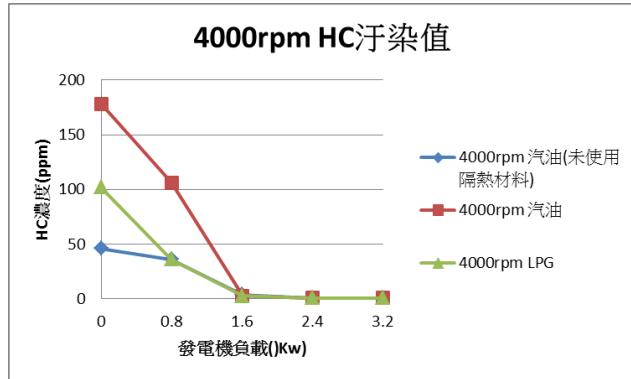


圖 8、4000rpm HC 汚染值。

圖 9~11 所示，NO<sub>x</sub> 於引擎 3000 rpm，使用隔熱材料後 NO<sub>x</sub> 排放明顯比未使用隔熱材料多，並隨著發電負載的增加，排放量也明顯增加，液化石油氣的部分，NO<sub>x</sub> 排放量則是明顯比汽油燃料來的多；NO<sub>x</sub> 於引擎 3600 rpm，轉速的提升，NO<sub>x</sub> 排放量也比 3000 rpm 多；NO<sub>x</sub> 於引擎 4000 rpm，NO<sub>x</sub> 排放比 3600 rpm 有小幅度的增加；在這三個參數下的實驗結果中也得知在低發電負載的狀況下引擎燃燒效率比高發電負載差，在低發電負載並且使用隔熱材料後，NO<sub>x</sub> 排放量就極為接近。

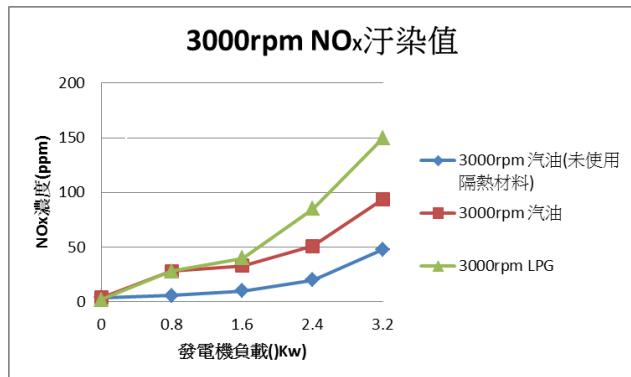


圖 9、3000 rpm NO<sub>x</sub>汙染值。

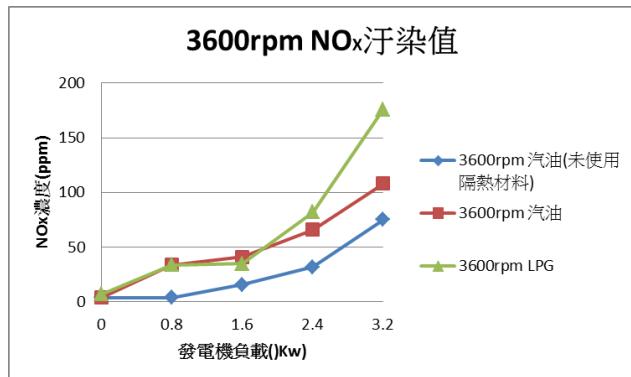


圖 10、3600 rpm NO<sub>x</sub>汙染值。

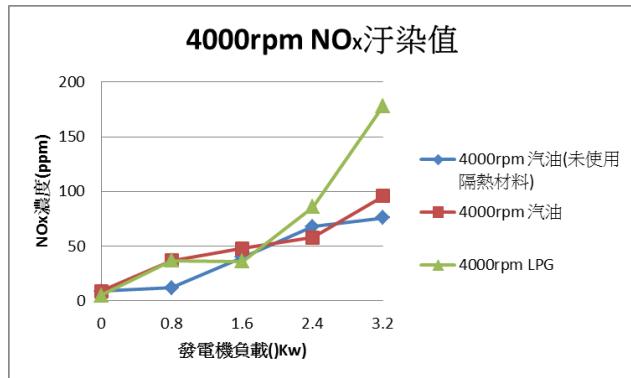


圖 11、4000rpm NO<sub>x</sub>汙染值。

儲熱方面的結果，溫度上升時間，隨著發電機負載增加而縮短，而發電機負載的增加也代表引擎輸出功率也要相對地增加，輸出功率增加使得燃料消耗也跟功率成正比增加；如圖 12~14 所示，引擎轉速 3000 rpm 到 3600 rpm，加熱時間有明顯的縮短，而在 3600 rpm 到 4000 rpm，加熱時間縮短的量有漸漸緩和的趨勢，加裝隔熱材料對加熱時間上有的一定程度的縮短，在低發電負載的狀況下有明顯的效果，液化石油氣在轉速提高後，加熱時間有明顯的接近汽油燃料的加熱時間。

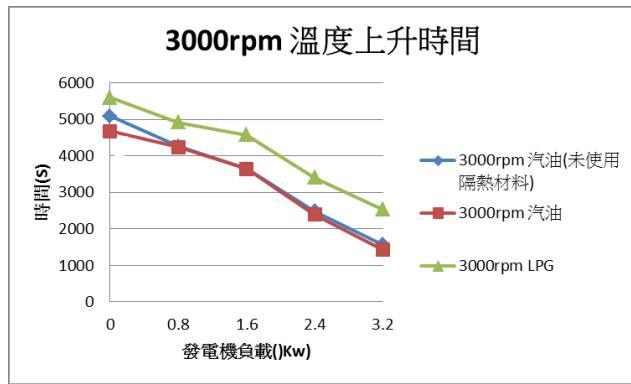


圖 12、3000 rpm 溫度上升時間。

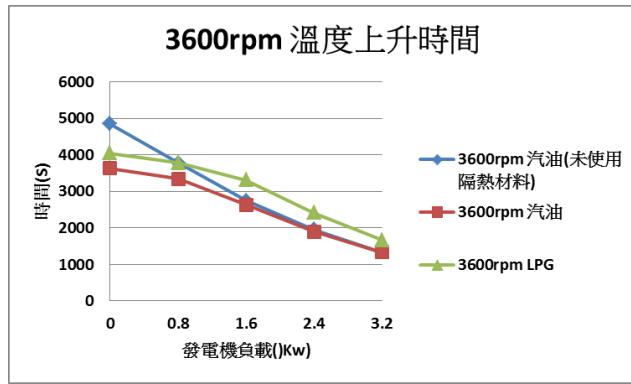


圖 13、3600 rpm 溫度上升時間。

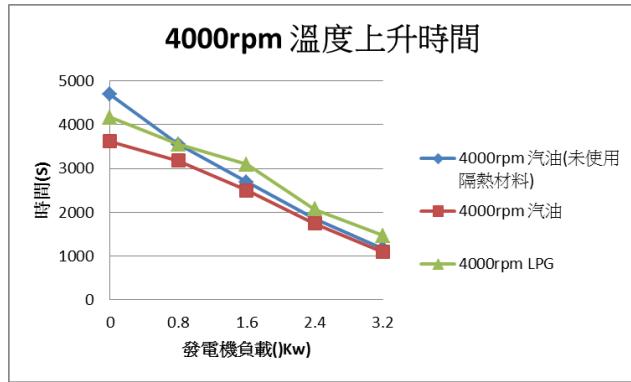


圖 14、4000 rpm 溫度上升時間。

圖 15~17 所示，能源回收率在引擎 3000rpm，未使用隔熱材料的狀況下，能源回收率比使用隔熱材料低約 10%左右，隨著發電負載的增加，汽油燃料最高回收率達到 71%，液化石油氣在無發電負載回收率就有 40%，但提高發電負載後，回收率就比汽油燃料低；引擎 3600rpm，能源回收率比引擎 3000rpm 來的低，液化石油氣則隨著引擎轉速提升，回收率比 3000rpm 最高有 8%的提升量；引擎 4000rpm，能源回收率比 3000rpm 和 3600rpm 有減少的趨勢，從實驗結果得知在轉速提高的狀況下，燃料消耗相對較多，回收率不一定會有所提升。

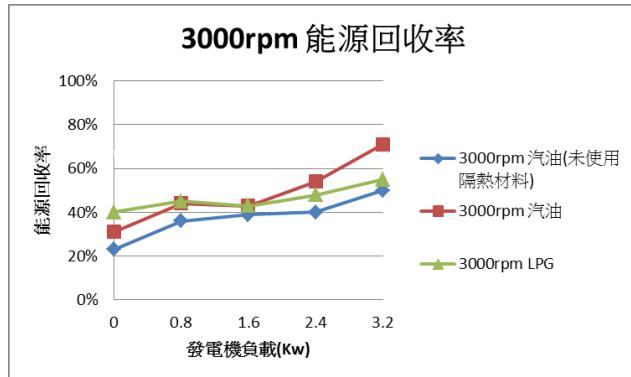


圖 15、3000 rpm 能源回收率。

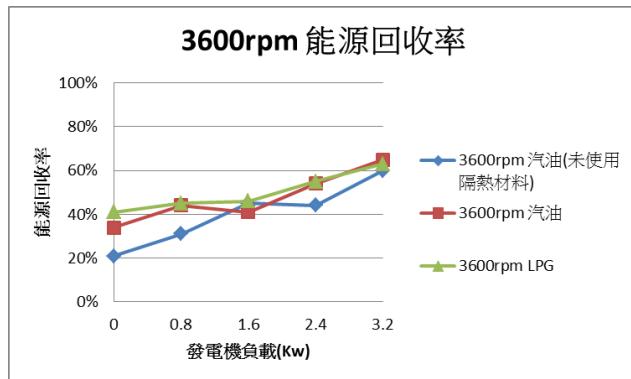


圖 16、3600 rpm 能源回收率。

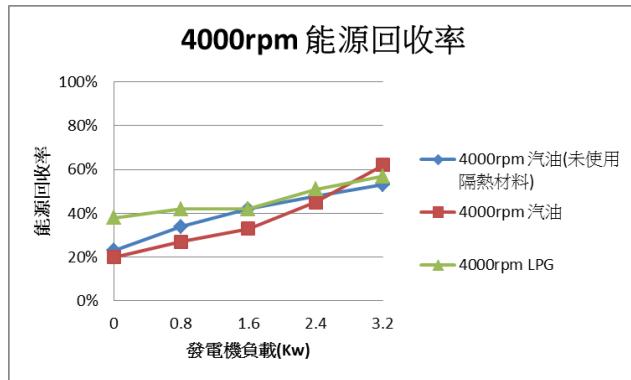


圖 17、4000 rpm 能源回收率。

#### 四、結論

廢氣排放 CO 會隨著引擎轉速及發電負載的增加而減少，在高發電負載的狀況下 CO 排放更趨近於零；HC 排放，轉速對 HC 排放影響較小，而在高發電負載下排放量會有明顯的下降，在發電負載 3.2 kW 的狀況下，HC 排放都能 10 ppm 以下；NO<sub>x</sub> 排放，在無發電負載的狀況下都在 10 ppm 以下，但隨著發電負載的增加，讓引擎效率有所提升，NO<sub>x</sub> 排放量也跟著增加，液化石油氣增加幅度更為明顯；從上面說明得知 CO 及 HC 因 LPG 燃燒較完全，排放量較少；而 NO<sub>x</sub> 因 LPG 燃燒溫度較高，使得排放量較汽油系統高。

能源回收率再提高發電負載後也跟著提升，使用隔熱材料後讓能源效率提升了約 10%左右的能源效率，液化石油氣則隨著引擎轉速的提升，而逐漸趨近於汽油的回收率，而引擎轉速增加，燃料消耗也跟著增加，對能源回收率不一定會有效果；在未使用隔熱材料最高回收率達到 59.8%，使用隔熱材料後最高達到 71.2%，液化石油氣回收率達 63.1%。

## 五、致謝

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# 中鋼公司真空精煉製程冶煉超低碳鋼液的技術

## The Technology of RH Vacuum Refining Process for Producing Super-low C Steels in China Steel Corporation

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### 摘要

碳氮成份的控制能力是先進鋼廠的重要指標，西元 2004 年以前，中鋼生產的極低碳氮鋼種，碳氮含量最嚴苛規格為  $\leq 25\text{ppm}$ 。近來，為提昇市場對極低碳氮鋼材的「深沖成形性」需求，西元 2004 年中旬開始積極開發規格碳氮含量  $\leq 18\text{ppm}$  的鋼種。因此需提昇煉鋼的技術水準，積極開發超低碳氮之冶煉連鑄技術，以穩定生產深沖成形的高品級冷軋超低碳氮鋼材。研究藉由開發試煉相關鋼種，建立 RH 真空精煉製程冶煉鋼液  $[C] \leq 10\text{ppm}$  的製程技術，以滿足超低碳鋼種的生產需求。

關鍵字：極低碳鋼、IF 鋼、極低氮鋼

### Abstract

The control ability of carbon and nitrogen of molten steel is an important indicator of advanced steelmaking plants. The lowest specifications of carbon and nitrogen content was less than 25ppm for the ultra-low carbon and nitrogen steels in China Steel Corporation (CSC) before 2004. Recently, in order to enhance the market demand for the high-deep drawing formability of ultra-low carbon and nitrogen steel, the specifications of carbon and nitrogen being less than 18ppm of steels were to be developed in mid 2004. Therefore, the steelmaking and continuous casting technology for the super-low [C], [N] steels should be actively developed. The refining technology of RH vacuum process making molten steel with  $[C] \leq 10\text{ppm}$  was established by experimenting on related steels. So, the high-grade and high-deep drawing steels are stably produced to meet the customer demand.

Keywords: ultra-low carbon steel, IF steel, ultra-low nitrogen steel

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## 一、前言

碳氮成份的控制能力是先進鋼廠的重要指標，中鋼煉鋼廠對於生產全系列之電磁鋼、IF 鋼及熱浸鍍鋅汽車用鋼等極低碳氮鋼種，已累積了相當豐富的經驗，前述高品級鋼種碳氮含量在 2004 年以前最嚴苛之規格為小於 25 ppm。進一步降低鋼材的碳氮含量是必然的趨勢，提昇中鋼的煉鋼技術，開發碳氮含量規格小於 18 ppm 最高品級 IF 鋼之煉製條件，使能順利生產此類型鋼種，應屬必行之任務。

依據  $C \leq 18\text{ppm}$ - $N \leq 18\text{ppm}$  高純度冷軋 IF 鋼種的煉鋼和連鑄製程設計，如表 1 所示：鋼液煉製過程中，氮含量降低主要依賴轉爐製程吹煉脫氮，轉爐冶煉的目標在於煉製氮含量  $\leq 12\text{ppm}$  的鋼液；RH 真空脫氣製程目標定於煉製碳含量  $\leq 10\text{ppm}$  之鋼液，同時要全力防止復氮發生，以確保 RH 製程處理完畢之鋼液碳、氮含量皆小於 10、12 ppm；連鑄製程嚴格控制鋼液復氮量  $\leq 5\text{ppm}$ 、復碳量  $\leq 5\text{ppm}$ ，以穩定產製碳氮含量規格皆小於 18 ppm 之高品級 IF 鋼種。

因此本研究主要目的：是針對極低碳氮鋼種於 RH 製程冶煉時，研究其各種製程參數與脫碳速率的關係，開發 RH 製程最佳的脫碳參數，使穩定煉製碳含量小於 10 ppm 之鋼液，以順利生產碳氮含量小於 18 ppm 之高品級 IF 鋼種。

表 1、 $C \leq 18\text{ppm}$ - $N \leq 18\text{ppm}$  高純度冷軋鋼種煉鋼和連鑄製程設計

成 分 製 程	轉 爐	RH 真空精煉	鋼液連鑄製程
碳 [C]	脫碳	脫碳 $[C] \leq 10\text{ppm}$	復碳量 $\leq 5\text{ppm}$
氮 [N]	脫氮 $[N] \leq 12\text{ppm}$	復氮量 $\leq 0\text{ppm}$ 目標： $[N] \leq 12\text{ppm}$	復氮量 $\leq 5\text{ppm}$

## 二、文獻蒐集與分析

### 2.1 RH 脫碳反應的熱力學原理

RH 真空精煉製程的脫碳反應原理，簡單而言，是在上昇管之下部位吹進氮氣，造成鋼液流動之壓差動力源，將鋼液環流於真空反應槽與盛鋼桶之間，藉真空反應槽內的真空壓力條件，驅使鋼液內的溶質元素 C、O 產生反應、生成 CO 氣體被脫除。依熱力學觀點，RH 製程一般操作鋼液溫度約 1600 °C，反應平衡常數視為定值，故鋼液內 C、O 的平衡含量，因 CO 之分壓而變，亦即受制於真空反應槽內 CO 分壓( $P_{CO}$ ) 之值。而 RH 製程真空槽鋼液中 C、O 之脫除，是以下列反應形式進行：



而反應平衡常數  $K_{CO} = \frac{P_{CO}}{[O^e] \cdot [C^e]}$

$$\log K_{CO} = \frac{1860}{T(^\circ K)} + 1.643$$

當反應溫度  $T=1600^\circ C$  時， $[C^e(\%)] \cdot [O^e(\%)] = 0.0023 (P_{CO})_{atm}$ ，此氣-液異相反應平衡，依勒沙特列原理，若氣相中  $P_{CO}$  值越低，反應越有利於向右進行，即  $[C^e(\%)] \cdot [O^e(\%)]$  乘積值越低。

## 2.2 RH 脫碳反應的動力學原理

根據文獻闡明：RH 製程脫碳反應的模型[1]概要如圖 1 所示，此模型認定 CO 反應發生的地點在下列 3 處：

- (1) 真空槽內曝露於真空氣氛下的鋼液自由表面（表面脫碳）；
- (2) 吹入氬氣泡的表面（氣泡脫碳）；
- (3) 真空槽內鋼液內部過飽和 CO 氣體的逸出（內部脫碳）。

而在 RH 真空槽內 CO 反應速率的控制步驟有 4 個：(1) C 在鋼液側的質傳；(2) O 在鋼液側的質傳；(3) 在鋼液和氣相之界面 CO 化學反應速率；(4) CO 在氣相側的質傳。此模型再考慮真空槽和盛鋼桶環流層、滯留層之間的鋼液對流，係循既定的「環流速度」和實測的混合時間作為依據，由文獻推定 C、O、CO 之質傳係數和氬氣泡的表面積，進行脫碳反應與碳含量變化之間的計算。

關於 RH 脫碳反應的行為機構可分為三階段：(I)第一階段、當鋼液碳含量  $[C] > 50ppm$  時，為在鋼液內部 CO 生成為主的內部脫碳反應機構；(II)第二階段、當  $50ppm > [C] > 30ppm$  時，為鋼液內部脫碳、表面脫碳、氣泡脫碳三者兼具的機構；(III)第三階段、當  $[C] < 30ppm$  時，為鋼液表面脫碳為主的機構，以及鋼液被污染復碳。

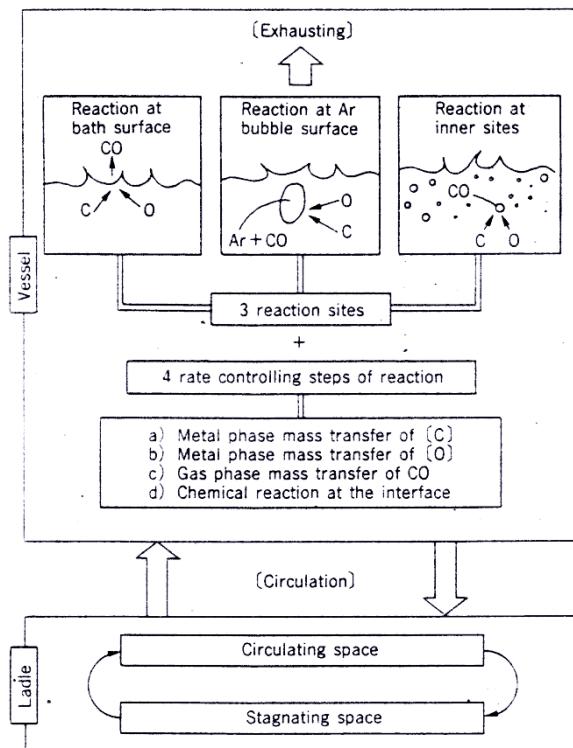


圖 1、RH 製程脫碳反應模型<sup>1</sup>

一般描述 RH 脫碳反應速率的動力學方程式如下[2]：

$$(1) \text{ 脫碳反應速率方程式: } -\frac{d[C]}{dt} = k_1 \left\{ [C] - \frac{P + P_{CO}^*}{K[O]} \right\} + k_2 \left\{ [C] - \frac{P}{K[O]} \right\}$$

$$(2) \text{ 真空壓力 } P \text{ 下降速率方程式: } -\frac{dP}{dt} = k_P P$$

$k_1 = \frac{Q}{Vl} \frac{akb}{(Q+akb)}$  : 內部脫碳反應(bulk reaction)之速率常數；

$k_2 = \frac{Q}{Vl} \frac{aks}{(Q+aks)}$  : 表面脫碳反應(surface reaction)之速率常數；

$k_P$  : 真空壓力下降速率常數；

$Q$  : 鋼液環流速率；

$Vl$  : 盛鋼桶內之鋼液容積；

$akb$  : 真空槽內鋼液內部脫碳反應之容積質傳係數；

$aks$  : 真空槽內鋼液表面脫碳反應之容積質傳係數；

$P_{CO}^*$  : CO 氣體成核之過飽和壓力(約 15 ~ 18 mmHg)；

$K$  : CO 反應之平衡常數；

RH 脫碳反應的第一階段：當鋼液[C]>50ppm 時，RH 處理過程之碳含量變化方程式可寫成：

$$[C] = [C]_0 \exp(-k_C t)$$

$[C]_0$  為起始之碳含量和  $k_C$  為脫碳反應速率常數，此時可定義： $k_C=k_1$ ；此式即為一般 RH 處理過程之碳含量變化方程式。所以，RH 第一階段脫碳技術主要在調整各種製程參數，藉以增加反應速率常數  $k_C$  為首要目標。

當鋼液  $[C]<30\text{ppm}$  時，RH 脫碳反應的第三階段，此階段的特點是脫碳反應停滯，主要原因是脫碳反應逐漸接近平衡，因此，脫碳過程中氣、液界面 CO 化學反應為控制反應速率的主要瓶頸。本研究需將鋼液在 RH 製程脫碳至 10ppm 以下，所以如何增強第三階段脫碳反應，為本研究亟待開發建立之技術所在。

RH 脫碳的第三階段中，脫碳反應幾乎只發生在真空槽內鋼液表面之氣液界面上，所以，第三階段促進脫碳的技術思路，只由從增加真空槽內反應界面的面積，以及脫碳反應之容積質傳係數著手。先進鋼廠的一些做法和措施如下所述：

#### (A) 提高環流強度

環流強度為鋼液環流量與盛鋼桶鋼液量的比值( $Q/V_1$ )，圖 2 說明[3]：第三階段中的脫碳速率和終點碳主要取決於環流強度，而不是單由環流量決定的。圖 2 中之 #1RH 的環流量 90(t/min)、環流強度達  $1(\text{min}^{-1})$ ；#2RH 的環流量達 135(t/min)、但環流強度只有  $0.73(\text{min}^{-1})$ ；這就是#1RH 的脫碳速率較佳和終點碳較低的原因。按圖 3 [4]，第三階段中增加循環氫氣量，直接增加鋼液環流強度，對降低終點碳很有幫助。實際上若循環氫氣量不變，隨著鋼液碳含量的下降，鋼液內部 CO 逸出量就會減少，所以無法維持原先的環流強度。

	No. 1RH	No. 2RH	test
Heat size (t/ch)	90	185	185
Gas exhaust capacity at 0.5Torr (kg/hr)	600	950	950
Minimum pressure (Torr)	0.1	0.3	0.3
Vessel inside diameter (mm)	1852	1854	1854
Snorkel inside diameter (mm)	400	450	510
Circulation gas flow rate (Nl/min)	1800	1600	3000
Steel circulation rate : Q (t/min)	90	100	135
O <sub>2</sub> flow rate (Nl/min)	13000	20000	20000

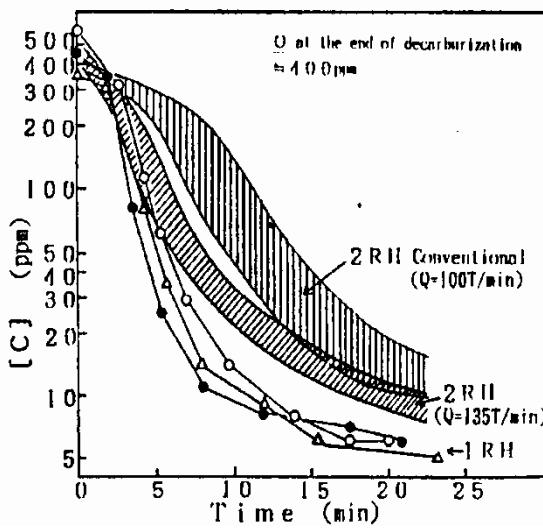


圖 2、不同設備條件下，RH 製程處理時[C]之變化[3]

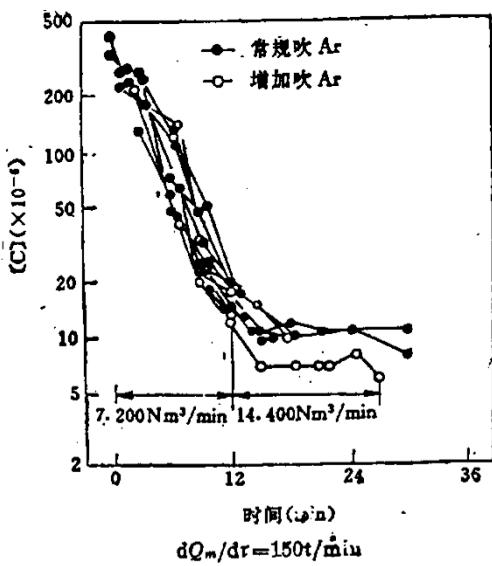


圖 3、RH 製程脫碳處理過程，鋼液碳含量[C]之變化[4]

#### (B) 增加脫碳反應之容積質傳係數

根據松野等人[5]所發表之真空脫氣槽內總體鋼液脫碳反應之容積質傳係數  $ak$  之關係式為：

$$ak = 2.27 \times 10^{-7} \cdot Q_g^{0.67} \cdot [C]_v^{1.76} \cdot [O]_v^{0.75} \cdot \left[ \ln\left(\frac{101.3}{P_2}\right) \right]^{0.67} \cdot A$$

式中  $Q_g$ ：循環氫氣流量

$[C]_v$ 、 $[O]_v$ ：真空槽內之 C、O 含量

$P_2$ ：真空槽內之壓力

A：真空槽內之截面積

RH 操作中真空壓力迅速抽低，不但有利於脫碳反應之熱力學條件，亦可使脫碳反應之總體質傳係數迅速增加，循環氫氣流量  $Q_g$  之大量化，促進上昇管內脫碳反應的進行，增加真空槽內底部截面積 A，皆可促進真空槽內鋼液脫碳反應之容積質傳係數，亦可增加真空槽內反應界面的面積。在脫碳反應的第三階段中因真空槽內之壓力和 C、O 含量已趨近定值，增加循環氫氣流量對總體質傳係數的助益較大，由圖 3 之結果亦得到驗證。

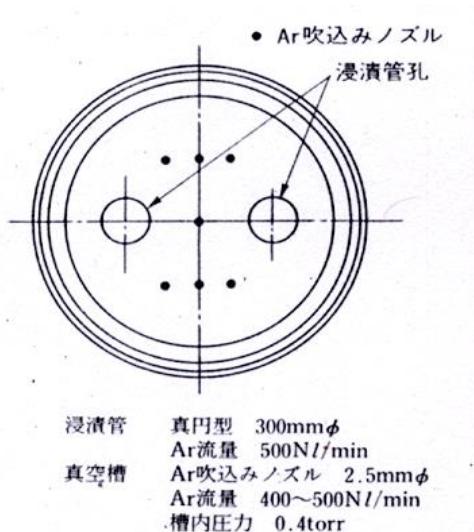
#### (C) 增加真空槽內反應界面的面積

理論上，在相同的真空環境下，單位時間的脫碳速率將因鋼液反應面積的增加而增大。為了煉製極低碳鋼，針對增加鋼液 CO 反應界面的面積，在 RH 製程所採用的技術為在真空槽裡吹入氫氣，作法如下：

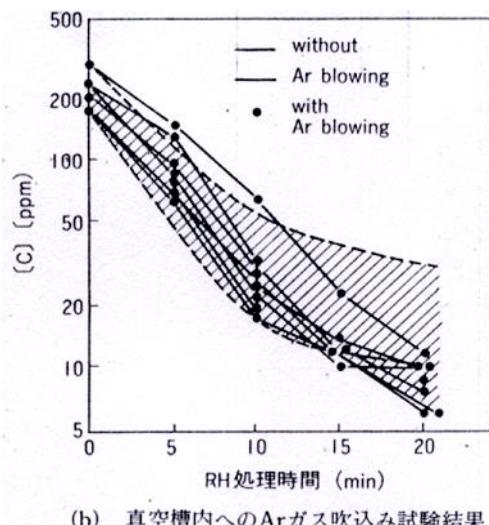
- (1) 在真空槽底部增加通氣孔，圖 4(a) [6] 所示，為藉強化攪拌來促進脫碳，其實績則如圖 4(b) 所示，值得注意的是，這不但提高在  $[C] > 50\text{ppm}$  時的脫碳速率，在  $[C] < 50\text{ppm}$  以下的極低碳區仍然維持相當高的脫碳速率，並無脫碳停滯的現象發生。RH 經過 20 分鐘

的脫碳操作後，[C]可穩定在 10ppm 以下。

- (2) 在真空槽下部側壁配置通氣管，如圖 5(a)[7]所示，亦大幅增加了脫碳反應面積，增加脫碳的速率，日本鋼管(NKK)開發了此技術後，最低碳含量可達 10ppm 以下，實績如圖 5(b) 所示。

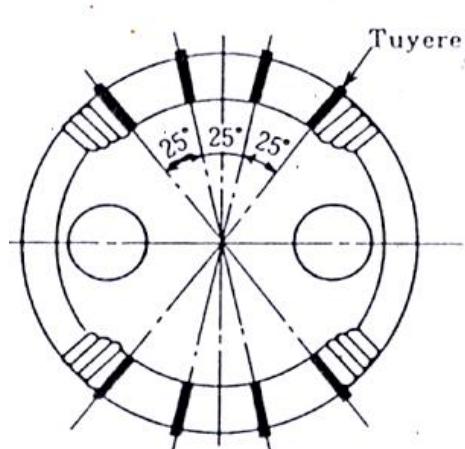


(a) 真空槽内へのArガス吹込み試験条件

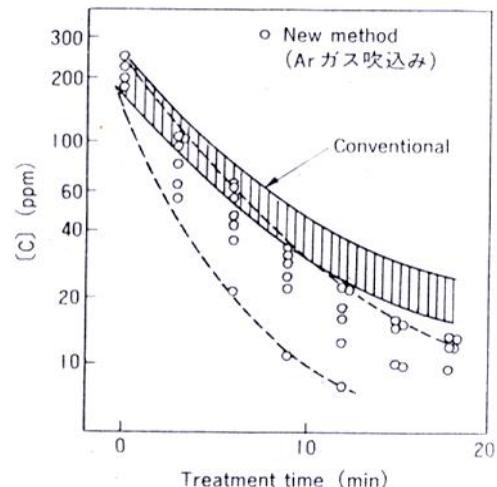


(b) 真空槽内へのArガス吹込み試験結果

圖 4、RH 製程真空槽底部吹入氮氣時的脫碳情況[6]



(a) 下部槽の羽口設置位置



(b) 底部吹込みによる脱炭速度の向上

圖 5、RH 製程真空槽下部側壁吹入氮氣時的脫碳情況[7]

## 2.3 防止 RH 脫碳反應第三階段之污染復碳

附在 RH 製程真空槽槽壁上的地金，在極低碳處理過程中再熔化，成為鋼液復碳的主因，為防止地金污染復碳，必須實施無地金操作，主要的方法如下[2, 8]：

- 使用乾淨度良好之 RH 真空槽，如 10 回以內之真空槽。
- 利用預熱保持真空槽內部溫度  $1400^{\circ}\text{C}$  以上，使噴濺鋼液不會在槽壁上黏結凝固。
- 使用頂吹氧氣清除槽壁上的地金。

另外，在處理超低碳鋼之前，RH 真空槽不要處理碳含量太高之鋼種，而且鋼液接觸的位置必須使用無碳之鎂鉻耐火磚，特別是真空槽底部的築磚，如此，鋼液方能防止污染復碳。

## 三、結果與討論

### 3.1 真空精煉爐頂吹氬氣促進脫碳實驗

當鋼液碳含量  $[C] < 30\text{ppm}$  時，為 RH 製程真空脫碳反應的第三階段，特點是脫碳反應趨向停滯，主要原因是碳氧反應逐漸接近平衡，脫碳反應幾乎只發生在真空槽內鋼液表面之氣、液界面上，因此，脫碳過程中氣、液界面 CO 化學反應為控制反應速率的主要瓶頸。所以，第三階段促進脫碳的技術思路，只能由增加真空槽內反應界面的面積，以及脫碳反應之容積質傳係數著手。

本實驗利用鋼鐵研發處的真空精煉爐設備，在脫碳末期(鋼液碳含量  $[C] < 30\text{ppm}$ )頂吹氬氣，如圖 6 所示，加強鋼液表面攪拌，增加鋼液的表面積，試驗增加真空槽內鋼液表面的面積是否會促進脫碳反應。

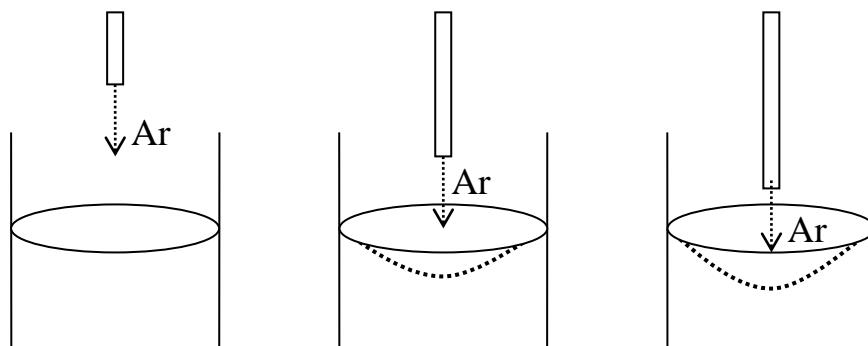


圖 6、真空精煉爐頂吹氬氣脫碳實驗示意圖

真空精煉爐頂吹氬氣脫碳實驗步驟為：

1. 熔煉鋼材、調整鋼液成份 C, Mn, P, S, Al 如下所示：

C (ppm)	Mn (%)	P (%)	S (ppm)	N (ppm)	Sol Al (%)
30	0.10	0.015	50	~15	0.01~0.03

2. 鋼液溫度調整至 1600°C，鋼液取樣確認成份。
3. 鋼液加氧化鎊皮後，測氧、溫度。維持溫度 1600°C。
4. 開始計時，抽真空使壓力至 1mbar 以下。
5. 持續抽真空，使壓力維持在最低狀態，頂吹氬氣(Ar) 10 分鐘後，鋼液加鋁脫氧後取樣分析成份。

實驗結果如表 2 所示：

表 2、真空精煉爐頂吹氬氣脫碳的實驗結果

爐號序	頂吹管高度	頂吹 Ar 流量	起始 $[C]_0$	10 min 後 $[C]_{10}$	脫碳速率常數
R638	200mm	25 NL/min	35	20	0.056
R639	400mm	25 NL/min	33	25	0.028
R639	150mm	25 NL/min	25	14	0.058
R640	150mm	40 NL/min	33	15	0.079
R085	--	0	35	21	0.051

一般真空脫碳反應過程的鋼液碳含量  $[C]_t$  變化方程式可寫成：

$$[C]_t = [C]_0 \exp(-k_C t)$$

$[C]_0$  為起始之碳含量， $k_C$  為脫碳速率常數， $k_C$  可由上述方程式移項整理後得到：

$$k_C = -\frac{1}{t} \ln([C]_t / [C]_0)$$

$k_C$  值越大、脫碳反應速率越快，圖 7 和圖 8 顯示出真空精煉爐脫碳反應速率常數的比較圖。圖 7 顯示：當真空精煉爐頂吹氬氣流量為 25 NL/min 時，頂吹氬氣管高度為 400mm、200mm、150mm，脫碳速率常數  $k_C$  值分別為 0.028、0.056、0.058 1/min，顯示頂吹氬氣管的自由距(free board) 越小，氬氣在鋼液表面擾動的表面積越大，脫碳反應表面積的增加，因此脫碳速率常數  $k_C$  值越大；但不吹氣的爐次 R085 之脫碳速率常數  $k_C$  值為 0.051 1/min，顯然在真空中吹氬

氣會破壞真空度，使真空壓力由 1mbar 上升至 4~5mbar，對脫碳反應造成負面影響，因此抵銷了增加 CO 反應表面積的效果。

圖 8 顯示：當真空精煉爐頂吹氬氣管的自由距相同為 150mm 時，頂吹氬氣流量分別為 0(不吹氣)、25 和 40 NL/min 時，脫碳速率常數  $k_C$  值分別為 0.051、0.058 和 0.079 1/min，顯然頂吹管在較接近鋼液表面位置時，頂吹氬氣流量越大，脫碳速率常數  $k_C$  值亦越大，此時頂吹氬氣有效擾動鋼液表面，增加 CO 反應表面積的效果，超過吹氬氣使真空壓力上升的負面影響。

實驗證實：真空脫碳末期，若在鋼液表面適當位置頂吹氬氣，有效造成鋼液表面攪動、增加鋼液的表面積，可有效促進脫碳反應速率。但若頂吹管的位置太高，在脫碳末期真空吹氬氣會破壞真空度，使真空壓力上升，對脫碳反應造成負面影響，抵銷增加 CO 反應表面積的效果。

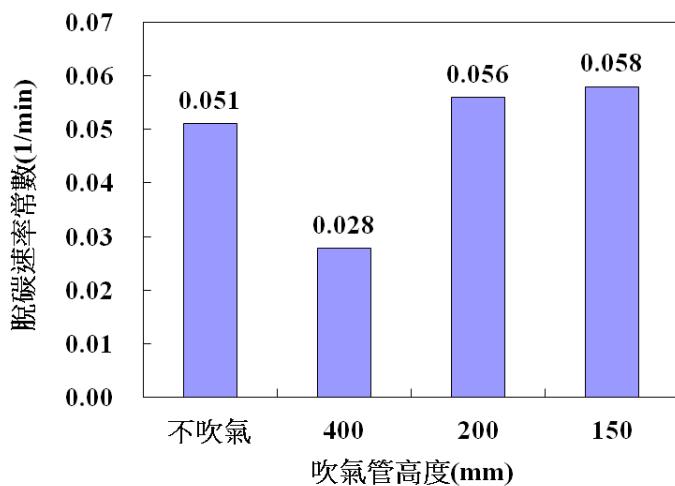


圖 7、頂吹氬氣流量 25 NL/min 時，鋼液脫碳反應速率常數的比較圖

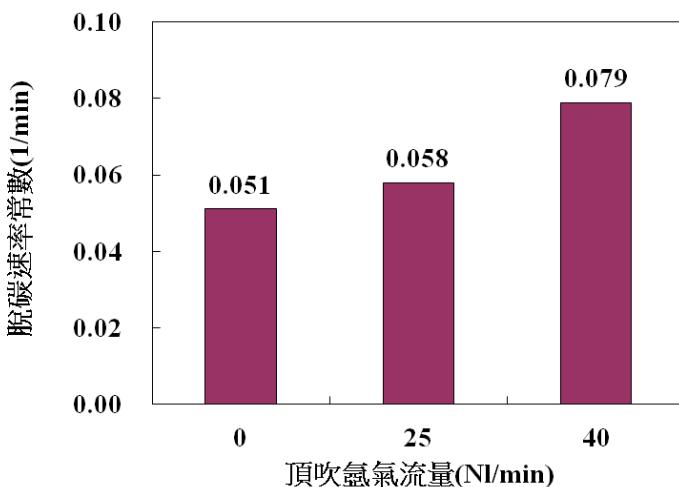


圖 8、頂吹氬氣管的自由距為 150mm，脫碳反應速率常數的比較圖

### 3.2 RH 處理極低碳鋼種脫碳數據分析

圖 9 顯示 RH 製程處理極低碳鋼脫碳結束後鋼液殘留氧含量 RD[O]對處理結束後 RA[C]之關係，鋼液脫碳後之殘留氧  $RD[O] \geq 400\text{ppm}$  的爐數有 21 爐， $RA[C] \leq 10\text{ppm}$  的爐數有 13 爐， $RA[C] \leq 10\text{ppm}$  的比例有  $13/21=61.9\%$ ；而當  $RD[O] \geq 450\text{ppm}$  的爐數有 13 爐， $RA[C] \leq 10\text{ppm}$  的爐次有 9 爐， $RA[C] \leq 10\text{ppm}$  的比例提升至  $9/13=69.2\%$ ；在未延長 VCD 脫碳時間且連續多爐次處理之下，提高脫碳後之殘留氧  $RD[O]$  值將有助於  $RA[C] \leq 10\text{ppm}$  的合格率。

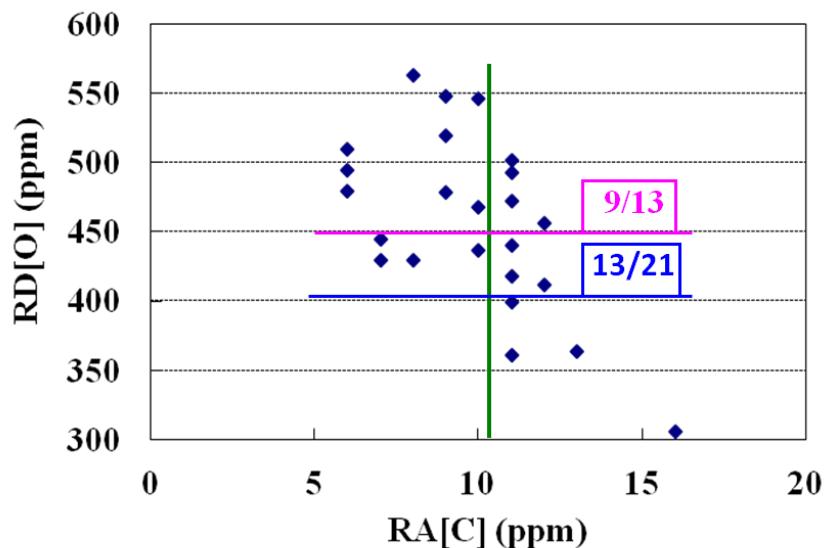


圖 9、脫碳結束後鋼液殘留氧含量  $RD[O]$  對處理結束後  $RA[C]$  之關係分佈圖

在 RH 製程冶金功能的研究，已建議煉鋼現場注意 RH 製程真空抽氣系統與原先設計能力有落差，經操作與維護單位同仁共同努力檢修改善後，極低碳鋼脫碳盛期速率常數  $k_C$  值的變化，如圖 10 所示，由檢修前的  $0.20/\text{min}$ ，經第一階段初次維修後， $k_C$  提高到  $0.28/\text{min}$ ，再經第二階段維修後， $k_C$  再提高到  $0.36/\text{min}$ ，已恢復至原先的設計能力。提高脫碳速率最直接的好處為煉製碳含量更低的鋼液，且可縮短 RH 處理時間，節省成本，確保極低碳鋼的煉製成功率。

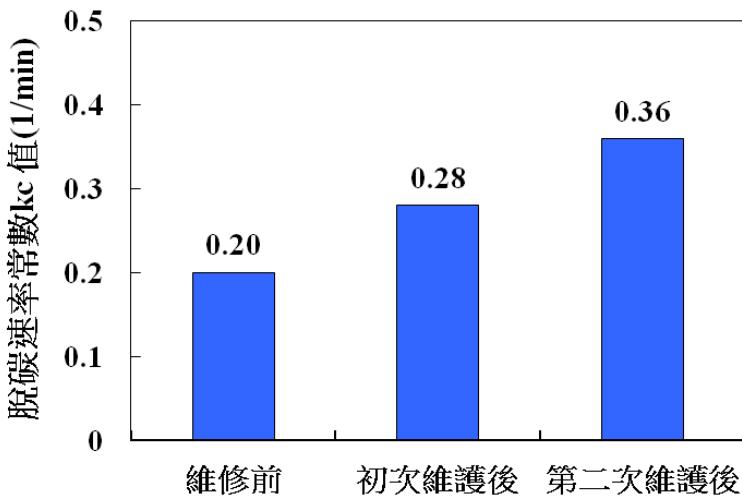


圖 10、RH 製程真空抽氣系統維修前後，脫碳速率常數  $k_C$  值演變圖

當鋼液  $[C] < 30\text{ppm}$  時，為 RH 脫碳反應末期階段，此階段為是否達到  $[C] < 10\text{ppm}$  的關鍵階段。當 RH 製程真空抽氣系統的能力正常時，真空壓力最低可達到 1mbar 以下，利用現場連續生產 12 爐次普通 IF 鋼種的結果，如圖 11 所示，此 12 爐次爐與爐的間隔時間極短，前 8 爐次上昇管循環氬氣流量為 1600Nl/min，後 4 爐次循環氬氣流量提高為 1800Nl/min；由圖 11 顯示：後 6 爐次的真空脫碳後(VCD)鋼液殘留  $[O]$  水準類似時，提高循環氬氣流量，可提升脫碳末期鋼液表面反應速率，因而使 RH 處理後之鋼液碳含量 RA[C] 可達到小於 10ppm 的世界一流水準。

一般 RH 處理極低碳鋼真空脫碳 VCD 後  $[O]$  與處理結束 RA[C] 之關係如圖 12 之深藍色點所示，此次研究的 12 爐次以紅點表示，分佈於底部雙曲線上，此顯現的意義為當 RH 製程真空抽氣系統能力正常，脫碳盛期速率常數  $k_C$  值為 0.36 時，VCD 後  $[O]$  與 RA[C] 的乘積約為  $3000 \text{ ppm}^2$ 。

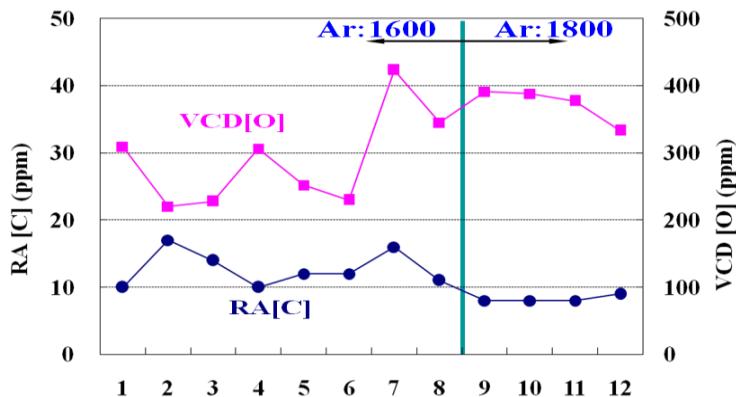


圖 11、RH 連續處理 12 爐次 IF 鋼種，處理結束後 RA[C] 和真空脫碳後 VCD [O] 的結果。

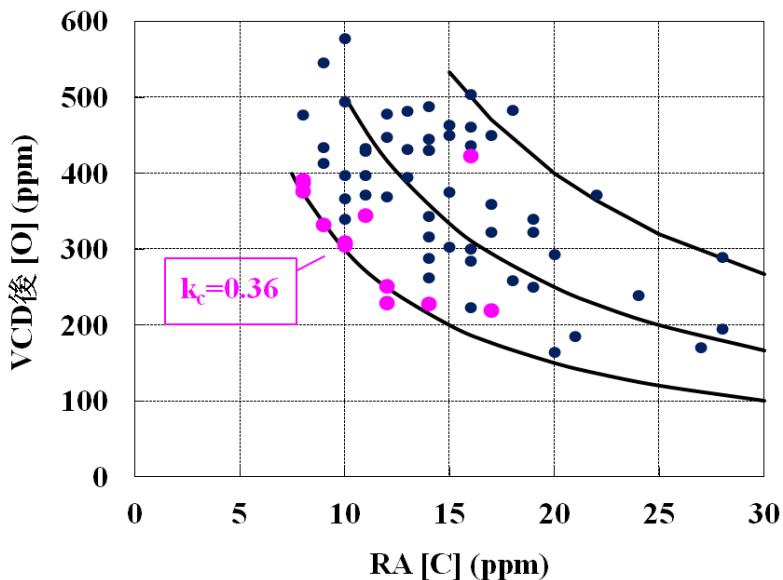


圖 12、RH 製程真空脫碳 VCD 後[O]與處理結束 RA[C]之關係分佈圖

#### 四、結論

綜合以上的結果和討論，可確立中鋼煉鋼廠 RH 欲穩定冶煉碳含量小於 10ppm 之鋼液製程條件如下：

- (1) 真空槽連續處理極低碳鋼、槽壁無地金附著。
- (2) 設備真空氣密良好，最低真空壓力可達 1 mbar 以下。
- (3) 提高 RH 製程循環的 Ar 流量  $\geq 1800 \text{ NL/min}$ 。
- (4) 利用 RH 製程頂吹氧氣功能，補足 VCD 後的鋼液殘留氧含量[O]  $\geq 400\text{ppm}$ 。
- (5) 延長脫碳的高真空時間大於 12 min 以上。

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- (1) 書籍：作者，書名，出版社，出版地，頁數，年。  
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