添加稀釋物對甲烷/一氧化二氮反置擴散火焰之燃燒特性影響

Effects of Diluent Addition on Combustion Characteristics of Methane/Nitrous Oxide Inverse Diffusion Flame.

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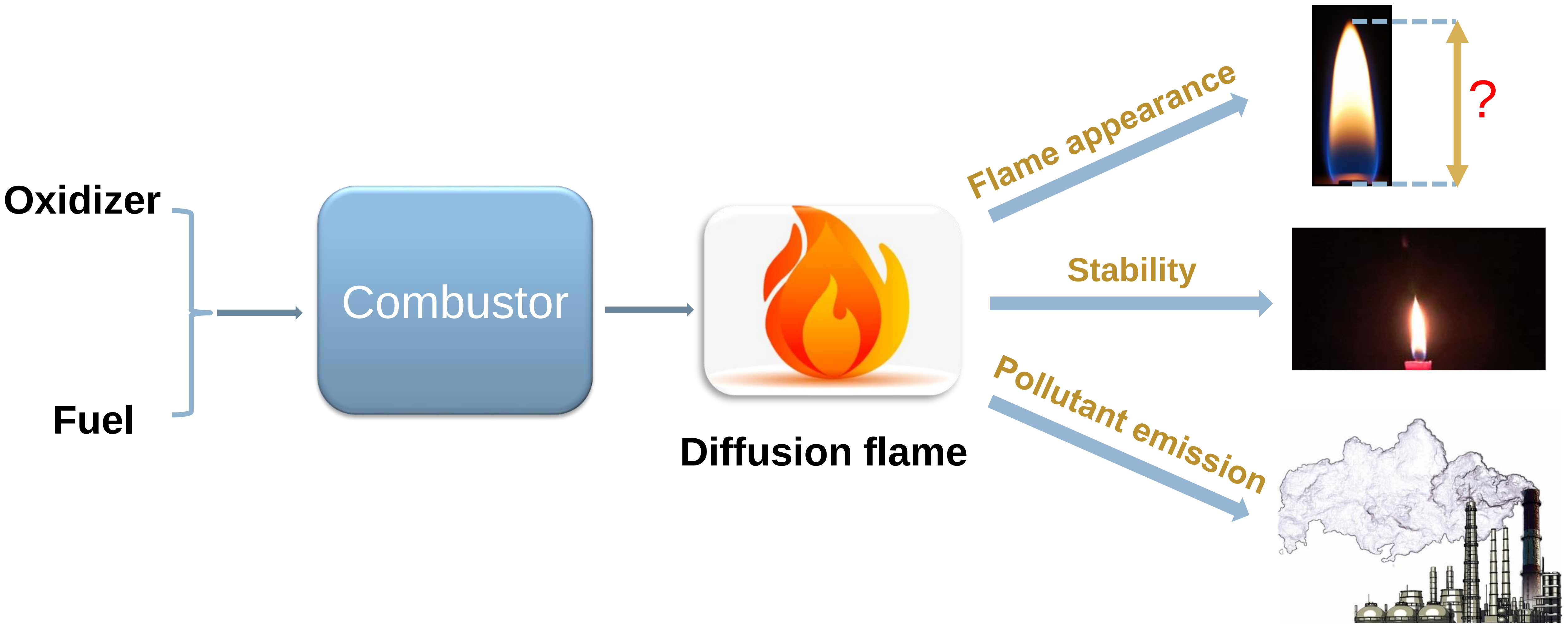
Background


Oxidizer
Fuel
Combustor
Diffusion flame
Flame appearance
?
Stability
Pollutant emission

Introduction-Diffusion flame

- *According to the different feeding pattern of the fuel and oxidizer*
  - Normal diffusion flame (NDF)-fuel jet is issued into oxidizer.
  - Inverse diffusion flame (IDF)-inner oxidizer jet is surrounded by an outer fuel jet.

[**Kumfer et al.**, *Combust. Flame* (2008)]

Introduction-Diffusion flame

- **Mikofski *et al. 2006* -** Inverse diffusion flame height predictions using modified Roper’s analysis for circular port burners agreed with measured reaction zone heights.

- **Johnson and Sobiesiak *2011* -** Numerical simulations suggest that when the mixture upstream of the partially premixed flame reaches $\Phi \approx 1$, it propagates upstream and stabilizes closer to the burner as an IDF.

Introduction - Oxy-enriched combustion

- **Leo *et al. 2007* -** The concentration of OH^* and CH^* increase as oxygen content increases. Besides, the excitation mechanism of OH and CH is thermal mechanism.

- **Yepes *et al. 2013* -** The radicals OH and O grow with oxygen content, the laminar burning velocity increase by approximately 25% for an enrichment level of 4%.

Introduction - Oxy-enriched combustion

If the oxygen concentration exceed 21%, it call oxy-enriched combustion. Moreover, the limit of the oxygen concentration is 100%, namely pure oxygen combustion.

- **Advantages**
  - ✓ Reduce the total amount of oxidizer
  - ✓ Increase the flame temperature
  - ✓ Save energy
  - ✓ Reduce pollutant emission
- **Disadvantages**
  - ✓ Higher cost
  - ✓ Low reliability of equipment system

Introduction - Nitrous oxide

- Nitrous oxide (N_2O) is often used as the oxidizer propellant for propulsion systems, it is so-called "green propellant"
- **Nitrous oxide (N_2O, so call laughing gas)**
  - ✓ Total reaction step：$N_2O \rightarrow N_2 + \frac{1}{2}O_2 + 82\left(\frac{MJ}{kmol}\right)$
  - ✓ Temperature of self-decomposition：520°C
  - ✓ Temperature of maintained self-decomposition ：1000°C
  - ✓ Adiabatic flame temperature ：1640°C

Introduction -Nitrous-oxide flame

- **Vanderhoff *et al. 1986* –** They studied the H_2/N_2O premixed flame, the equivalence ratio changed from lean to stoichiometric, temperature distribution and NO, O_2, N_2, and OH concentration distributions were obtained for preliminary results.

- **Newman-Lehman *et al. 2013* -** In CH_4/N_2O and C_2H_6/N_2O opposed diffusion flames, the flame suppression effect caused by N_2O addition is attributed to the relationship between reaction step $3H_2 + O_2 = 2H_2O + 2H$ and the reaction step $N_2O + H_2 = N_2 + H_2O$.

Introduction - The effect of diluent addition

- **McLintock *et al. 1968* -** When using carbon dioxide as the diluent, he reported a strong soot suppressing effect, but when using helium there was virtually no noticeable effect on soot suppression.

- **Kailasanathan *et al. 2013* -** The carbon dioxide-diluted flame is the coolest, with a peak temperature of 1760 K and the helium-diluted flame is the hottest, with a peak temperature of 2140 K.

Introduction - Pollutant emission

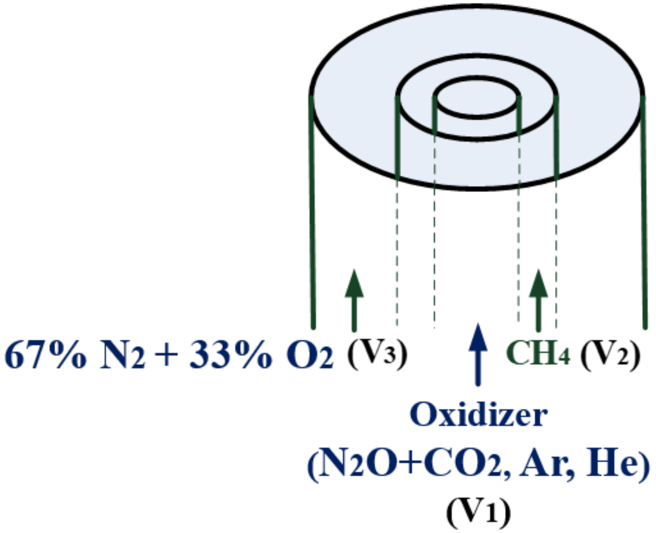
- **Rørtveit *et al. 2002* –** Adding diluent on the opposed hydrogen flame, the addition of He and CO_2 effectively reduced the NO content at similar temperatures condition.

- **Yamamoto *et al. 2011* –** They studied the effect of the lifted flame on pollutant emission. The NOx content decreases when the external flame lift.

Motivation and Objective

- Nitrous oxide is a strong oxidizer and widely used in industrial applications such as rocket propulsion, internal combustion engines.
- The addition of diluent is not only important for the combustion phenomenon, but also more likely to achieve pollution emission control.

Small-scale triple port burner



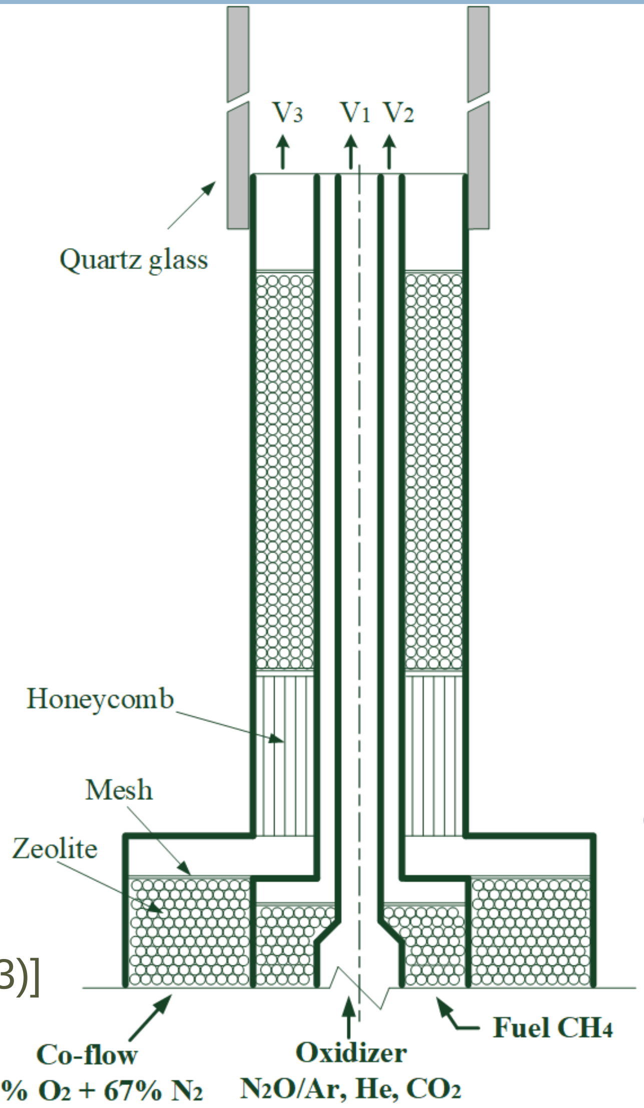
Dilute with Ar, He, CO_2

Flame behavior

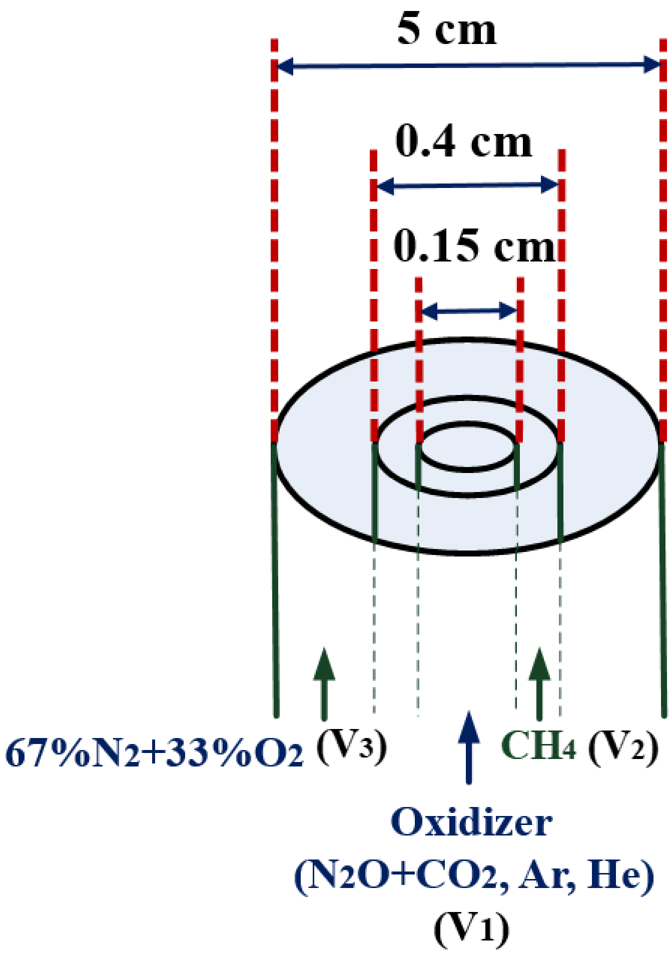
Investigation

- ✓ Flame configuration
- ✓ Pollutant emission
- ✓ The effect of diluent addition

Experimental Apparatus and Condition

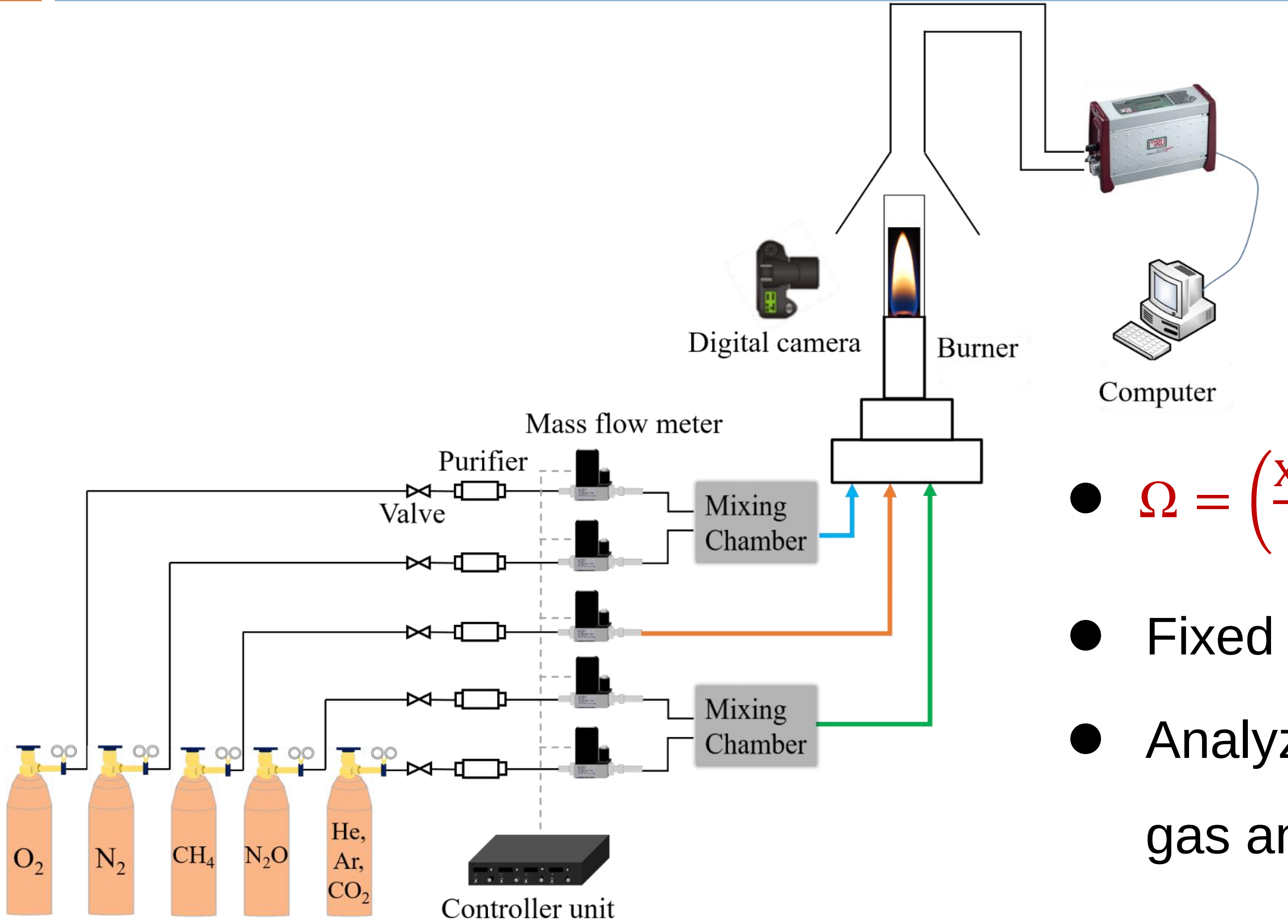


[**Lin**, *Master thesis. NCKU* (2013)]



ID = 0.15 ; 0.4 ; 5 cm
OD = 0.21 ; 0.5 ; 5.3 cm
L = 85 mm

Experimental Apparatus and Condition



- $\Omega = \left(\frac{X_{Diluent}}{X_{N_2O}}\right) \cdot 100\% \qquad R = \frac{V_1}{V_2}$
- Fixed $V_2 = 25$, $V_3 = 9.12$ (cm/s)
- Analyze pollutant emission by gas analyzer.

Theoretical Model

- ***Triaxial Burke-Schumann flame*** [**Ko et al.**, *Combust. Sci. Technol.* (2005)]
  - Including of the effect of axial diffusion and unequal stream velocities.
  - Reaction for the diffusion flame is one step overall.
  - Le=1, Fick's law for gas transport.
  - The buoyancy effect and shear force are neglected.
  - Steady, laminar, constant pressure and obey Ideal Gas equation for Jet.
- ***Governing equations***
  - In cylindrical coordinate system, coupling conservation of mass and energy equations.

$$\mathrm{Pe}\frac{\partial(\gamma_i)}{\partial\eta}-\left[\frac{1}{\xi}\frac{\partial}{\partial\xi}\left(\xi\frac{\partial(\gamma_i)}{\partial\xi}\right)+\frac{\partial^2(\gamma_i)}{\partial\eta^2}\right]=0\ ,\quad \gamma_{FO}=\tilde{Y}_F-\tilde{Y}_O\ ;\ \gamma_{FT}=\tilde{Y}_F+\tilde{T}\ ;\ \gamma_{OT}=\tilde{Y}_O+\tilde{T}$$

  - Using the method of separation of variables with boundary conditions, the general solution are obtained:

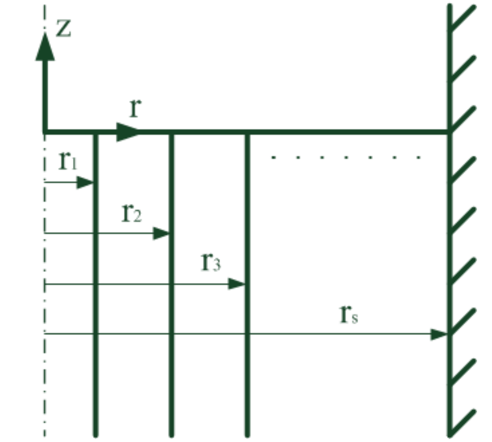
$$\boldsymbol{\xi}\equiv\frac{r}{r_s}\quad,\quad\boldsymbol{\eta}\equiv\frac{z}{r_s}$$

$$0=A'_{0,3}+\sum_{n=1}^{\infty}A'_{n,3}\frac{2}{\Phi_n J_0^2(\Phi_n)}J_0(\Phi_n\xi_f)\exp\left(\frac{Pe-\sqrt{Pe^2+4\Phi_n^2}}{2}\eta_f\right)$$

where

$$A'_{0,3}=B_{0,3}-A_{0,3}=-\tilde{Y}_{3,j}\cdot\left(\frac{u_3}{u_2}\right)-c_1^2\left[\tilde{Y}_{1,j}\cdot\left(\frac{u_1}{u_2}\right)+\tilde{Y}_{2,j}\right]+c_2^2\left[\tilde{Y}_{2,j}+\tilde{Y}_{3,j}\left(\frac{u_3}{u_2}\right)\right]$$

$$A'_{n,3}=B_{n,3}-A_{n,3}=c_2J_1(c_2\Phi_n)\left[\tilde{Y}_{2,j}+\tilde{Y}_{3,j}\left(\frac{u_3}{u_2}\right)\right]-c_1J_1(c_1\Phi_n)\left[\tilde{Y}_{1,j}\cdot\left(\frac{u_1}{u_2}\right)+\tilde{Y}_{2,j}\right]$$



Modified Theoretical Model

- **General solution**

$$\gamma_{FO} = \gamma_{FT} - \gamma_{OT} = \tilde{Y}_F - \tilde{Y}_O = B_{0,M} - A_{0,M}$$

$$+ \sum_{n=1}^{\infty} \left(B_{n,M} - A_{n,M}\right) \frac{2}{\Phi_n J_0^2(\Phi_n)} J_0(\Phi_n \xi)\, exp\left(\frac{Pe - \sqrt{Pe^2 + 4\Phi_n^2}}{2}\eta\right)$$

- **Definition of equivalence ratio**

$$\emptyset \equiv \frac{\left(O/F\right)_{stoic.}}{\left(O/F\right)}$$

$$\frac{Y_O}{Y_F} = \frac{\left(\frac{N_O M_O}{N_F M_F}\right)_{stoic.}}{\emptyset}$$

Modified Theoretical Model

- ***Definition of dimensionless mass fraction*** [**Ko et al.**, *Combust. Sci. Technol.* (2005)]

$$\tilde{Y}_j \equiv \frac{Y_j/M_j\left(\nu_j'' - \nu_j'\right)}{Y_F/M_F\left(\nu_F'' - \nu_F'\right)}$$

$$\times \frac{M_F(\upsilon_F'' - \upsilon_F')}{M_O(\upsilon_O'' - \upsilon_O')}$$

$$\frac{\dfrac{Y_O}{M_O(\upsilon_O'' - \upsilon_O')}}{\dfrac{Y_F}{M_F(\upsilon_F'' - \upsilon_F')}} = \tilde{Y}_O = \frac{\dfrac{\left(\dfrac{N_O M_O}{N_F M_F}\right)_{stoic.}}{M_O(\upsilon_O'' - \upsilon_O')}}{\dfrac{\emptyset}{M_F(\upsilon_F'' - \upsilon_F')}}$$

Modified Theoretical Model

- ***Modified general solution***

$$\mathrm{O} = \mathrm{N_2O}, \mathrm{F} = \mathrm{CH_4}$$

$$\tilde{Y}_O = \frac{1}{\emptyset}$$

$$\tilde{Y}_F = 1$$

$$1 - \frac{1}{\emptyset} = B_{0,M} - A_{0,M} + \sum_{n=1}^{\infty} \left(B_{n,M} - A_{n,M}\right) \frac{2}{\Phi_n J_0^2(\Phi_n)} J_0(\Phi_n \xi)\, exp\left(\frac{Pe - \sqrt{Pe^2 + 4\Phi_n^2}}{2}\eta\right)$$

$$\emptyset = 0.8 \text{、} 1 \text{、} 1.2$$

Numerical Simulation Method

- The CHEMKIN PRO, OPPDIF program, is used for numerical simulation.
- In order to understand the effects of diluent addition, three kinds of artificial species are created for simulation, namely N_2O(A), Ar(A), CO_2(A).

With ITC effect

↕ Chemical effect

With IT effect

↕ Thermal/Diffusion effect

With I effect

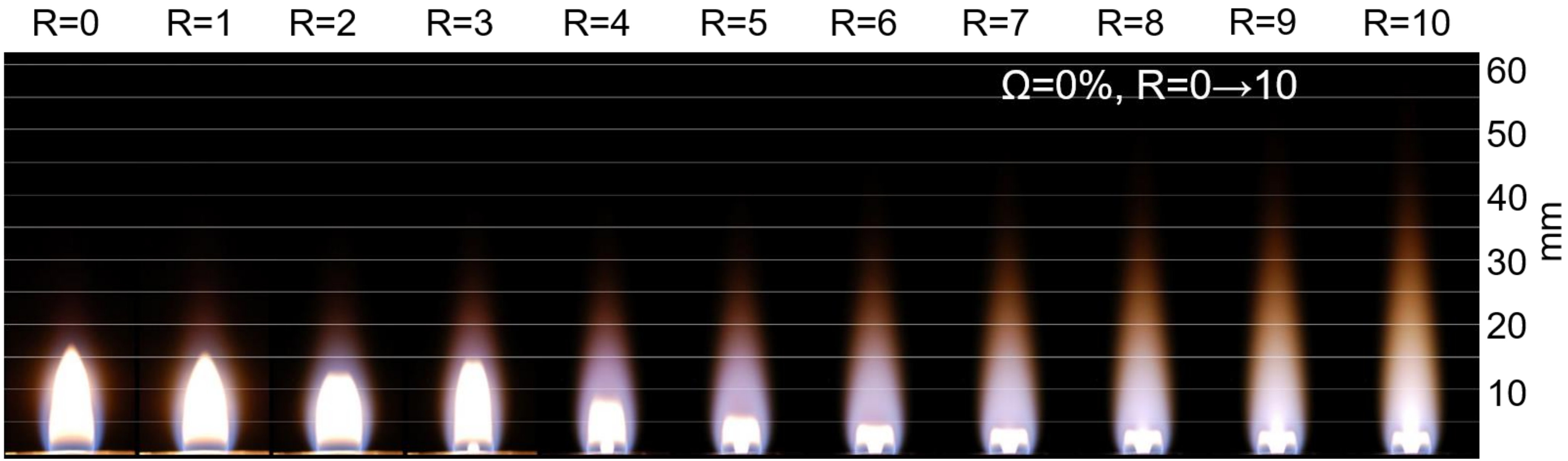
↕ Inert effect

No dilution

Case (ITC)	80% N_2O +20% Ar
Case (IT)	80% N_2O +20% Ar(A)
Case (I)	80% N_2O +20% N_2O(A)
Base case	100% N_2O

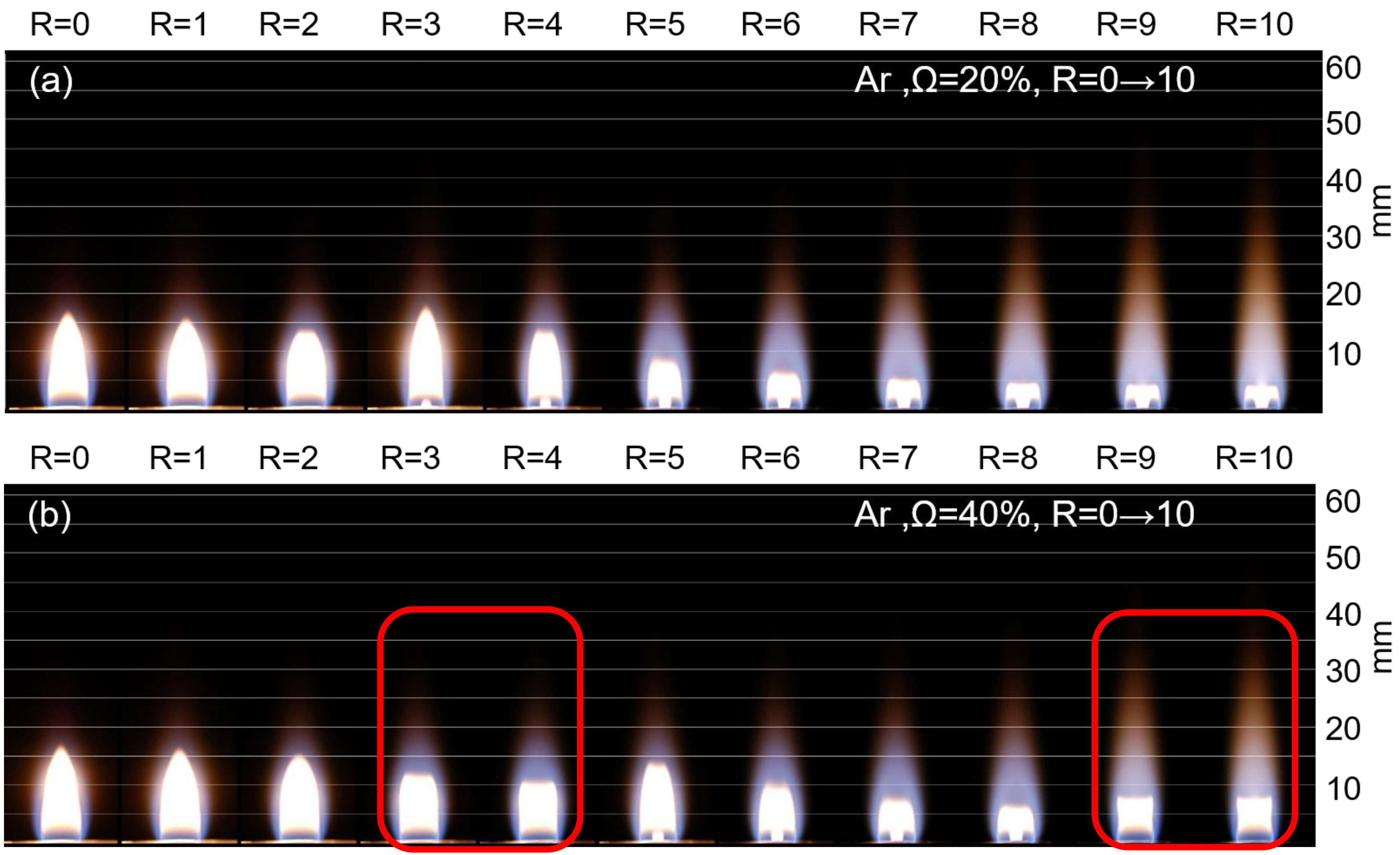
[**Shih**, *Combust.T and M* (2013)]

Results and Discussion - Flame Configuration

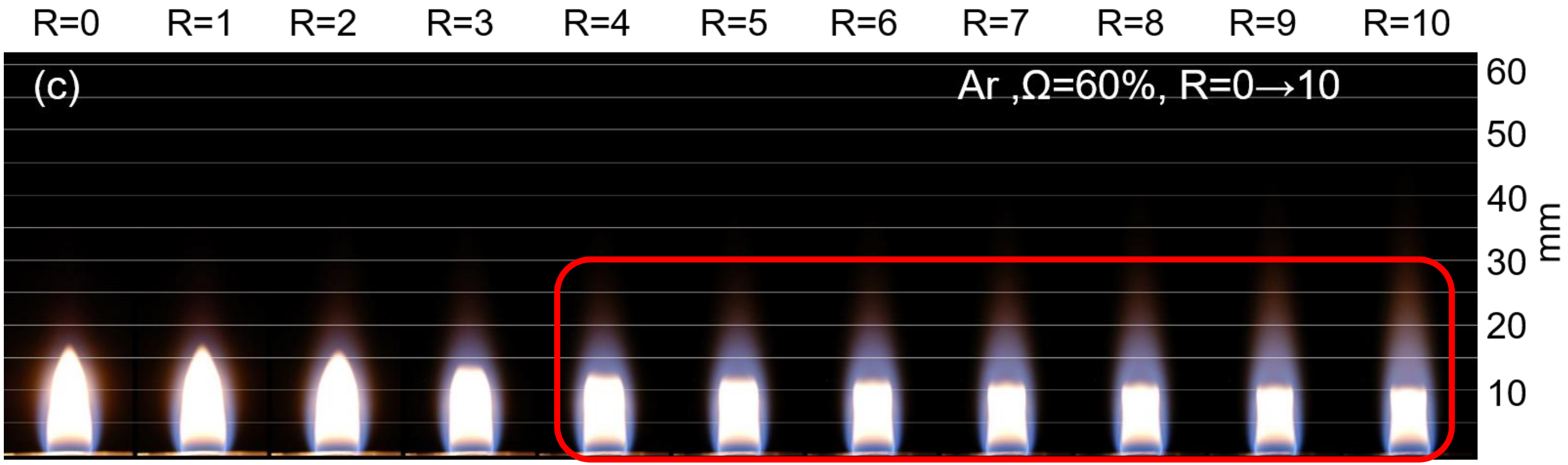


- The IDFs form at some certain Ω and R.
  - Because the flow field velocity of partially-premixed flame (PPF) and flame propagation speed can not approach to balance.

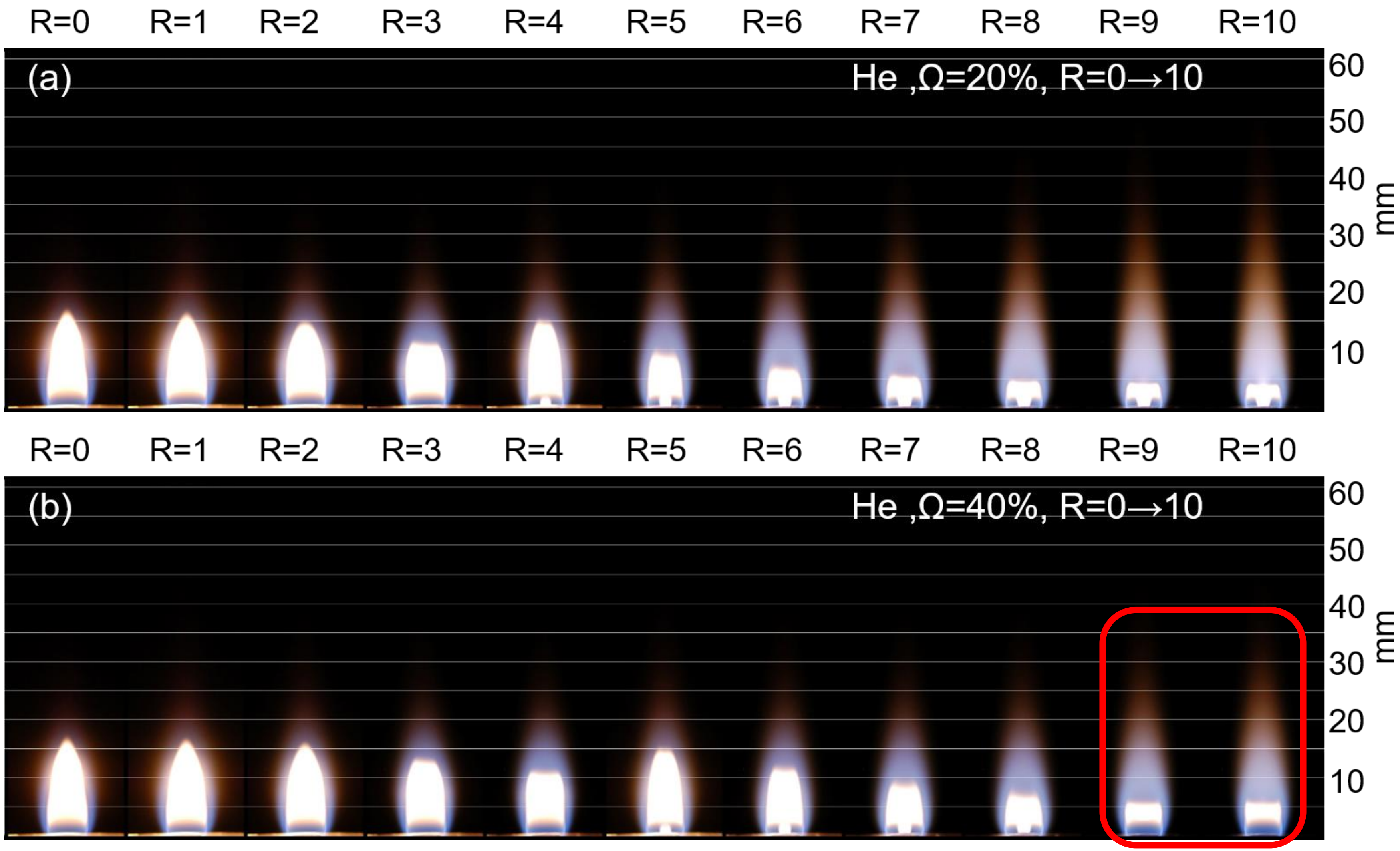
Results and Discussion - Flame Configuration (Ar)



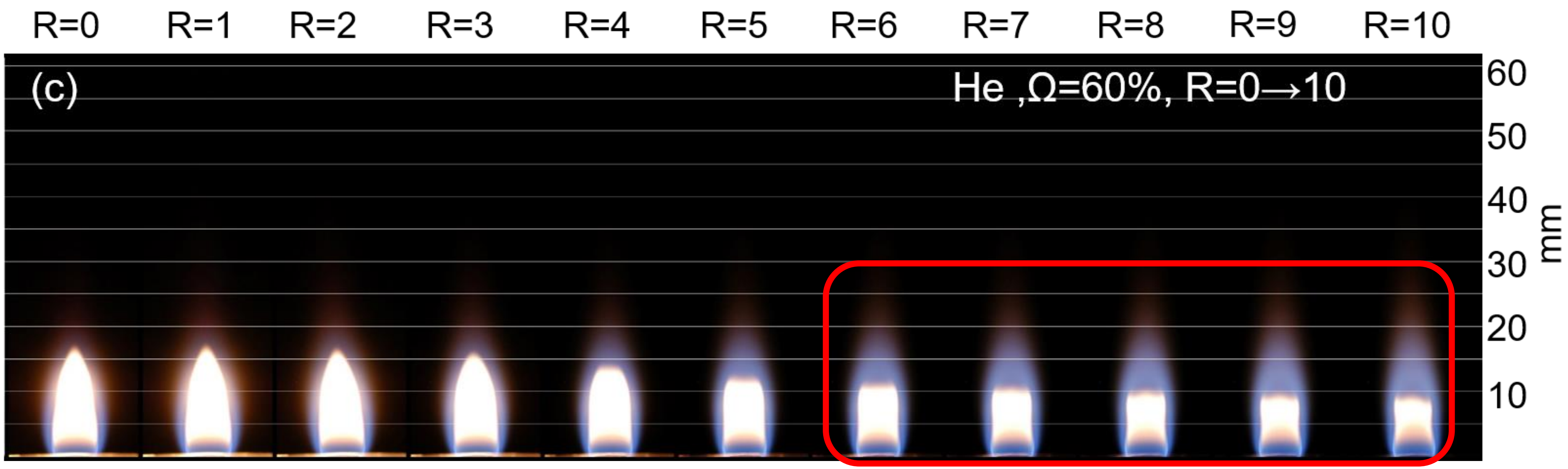
Results and Discussion - Flame Configuration (Ar)



Results and Discussion - Flame Configuration (He)

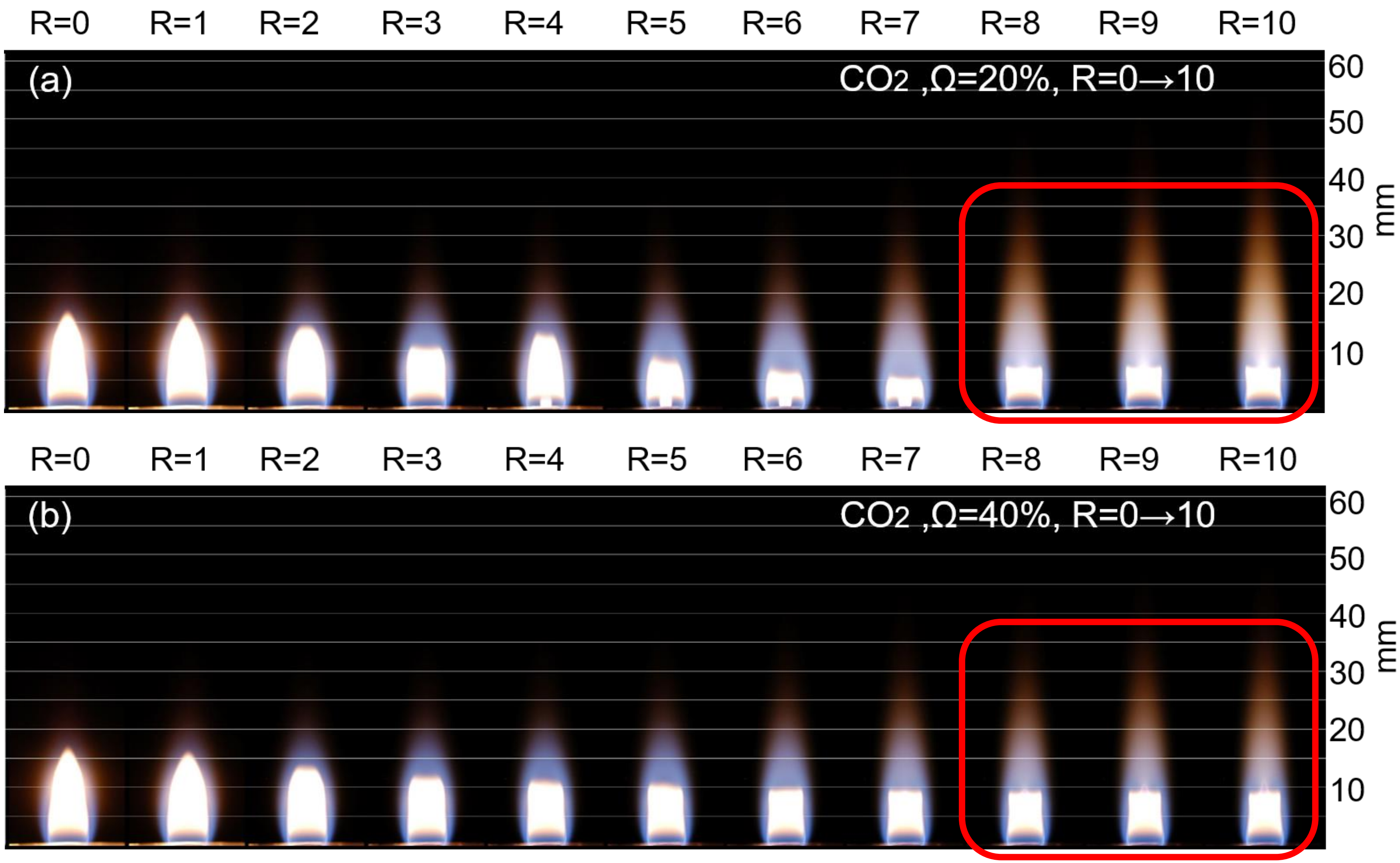


Results and Discussion - Flame Configuration (He)

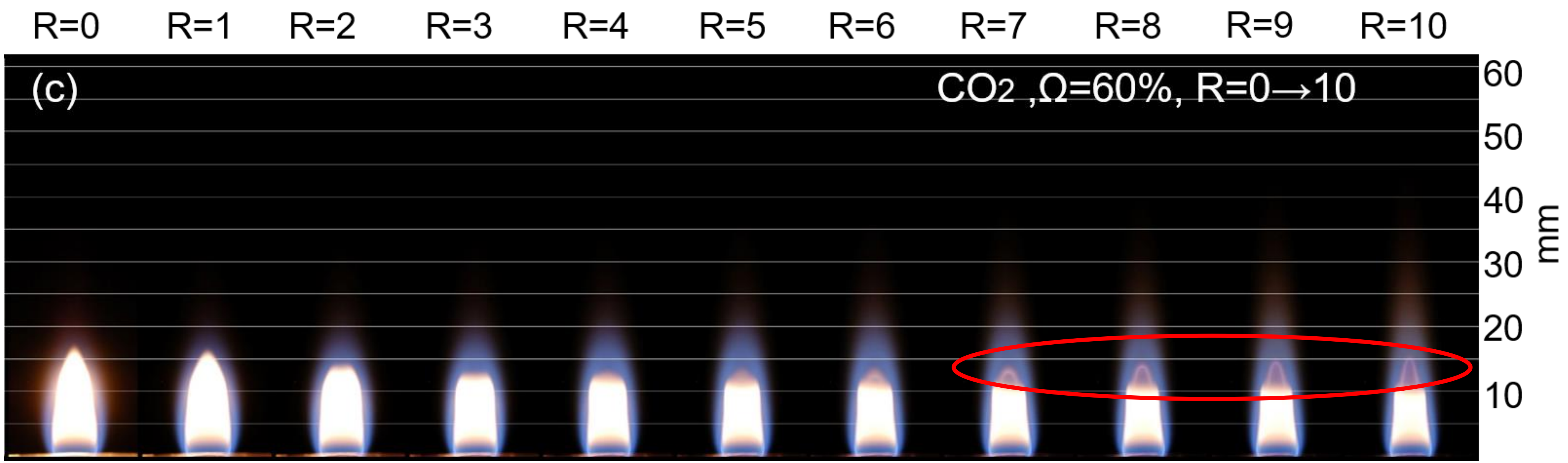


- Similar to Ar condition, it has a square-soot zone but it narrow down with increasing R.

Results and Discussion - Flame Configuration (CO_2)

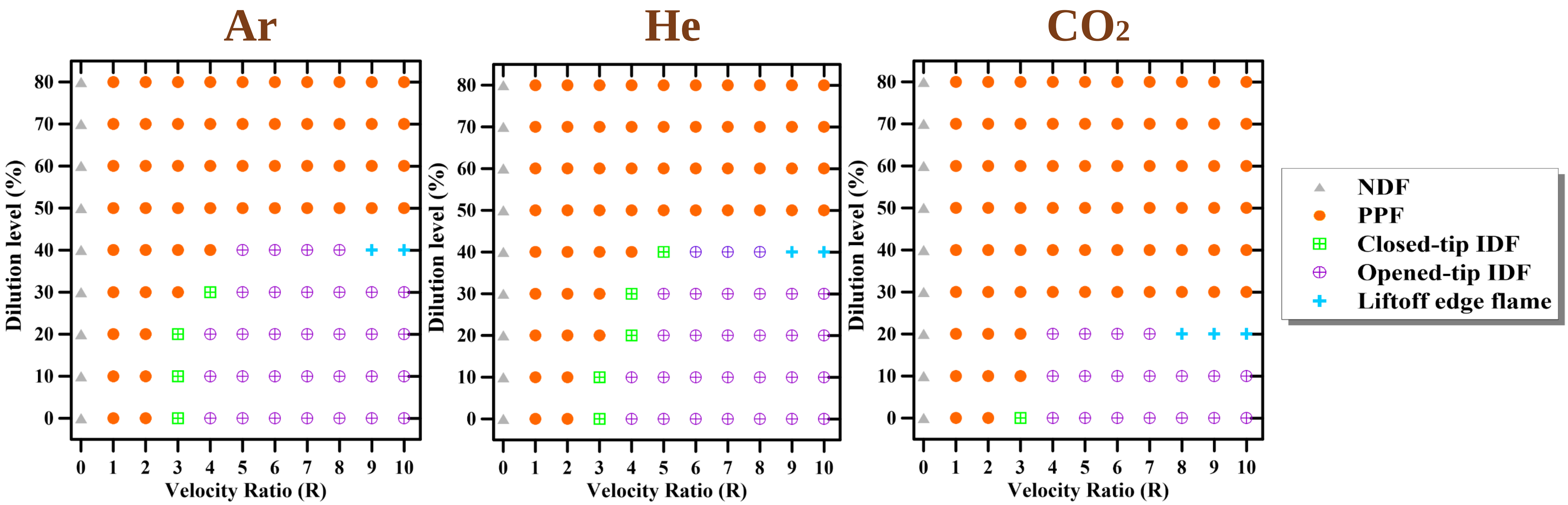


Results and Discussion - Flame Configuration (CO_2)



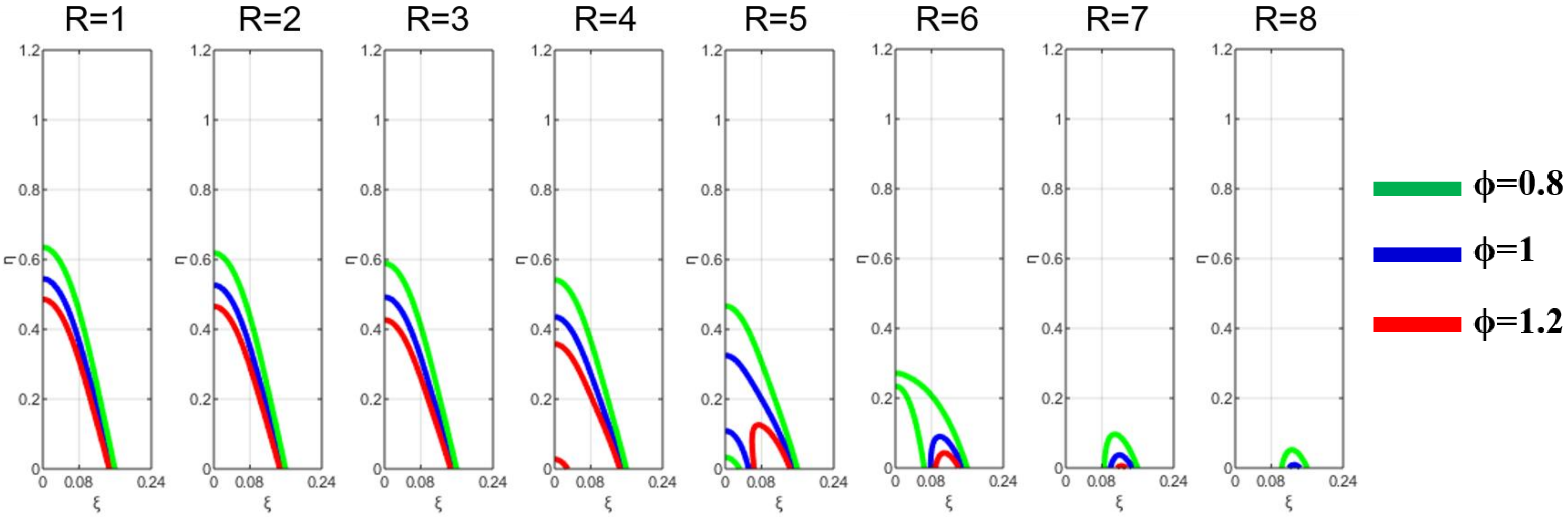
- Increasing the CO_2 concentration, the secondary flame becomes apparently. It is conjectured that the CO from pyrolysis of CO_2 re-react with N_2O, resulting in a CO flame.

Results and Discussion - Flame Configuration



Results and Discussion - Flame Shape Prediction

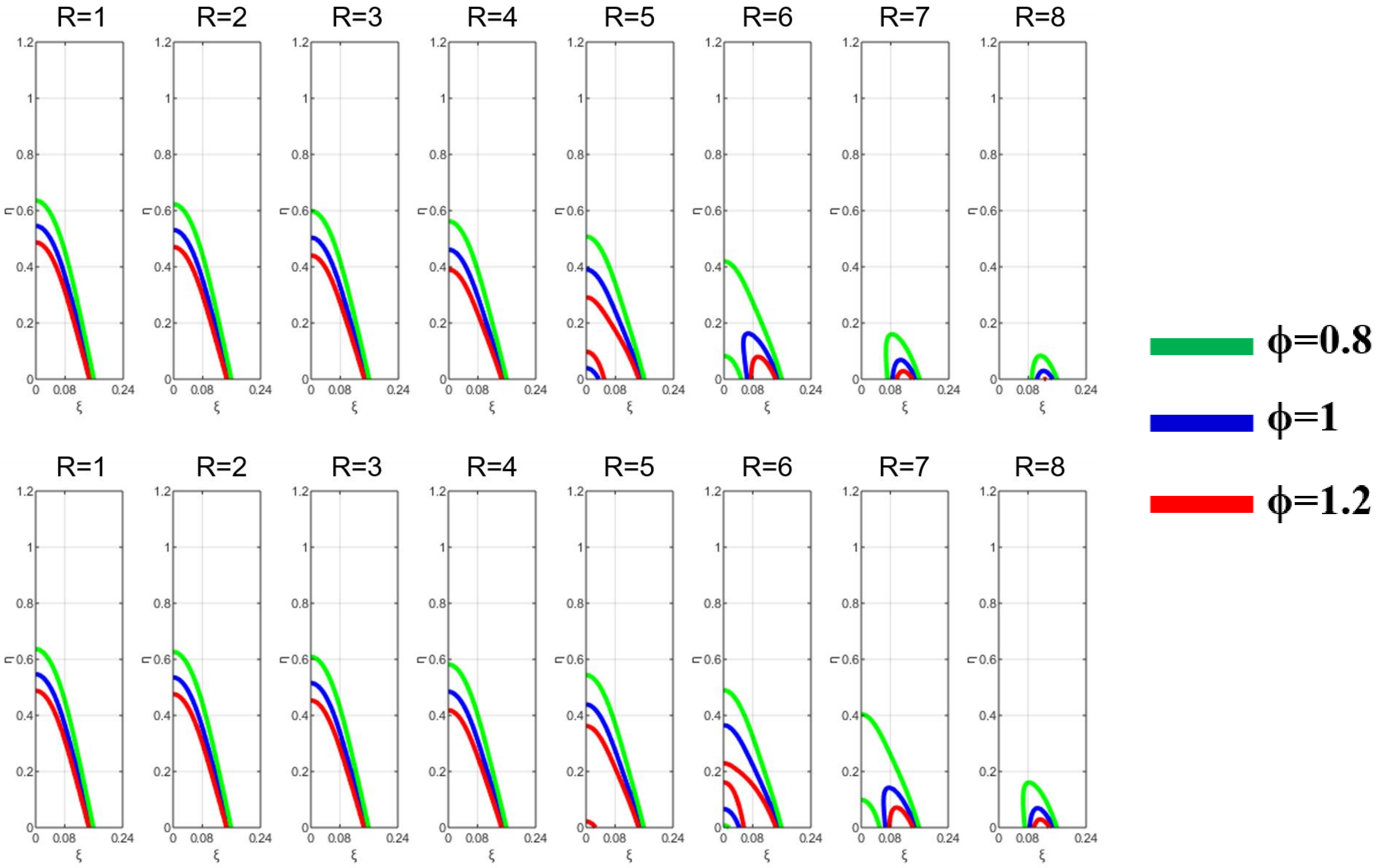
$\Omega = 0\%$



Results and Discussion - Flame Shape Prediction (Ar)

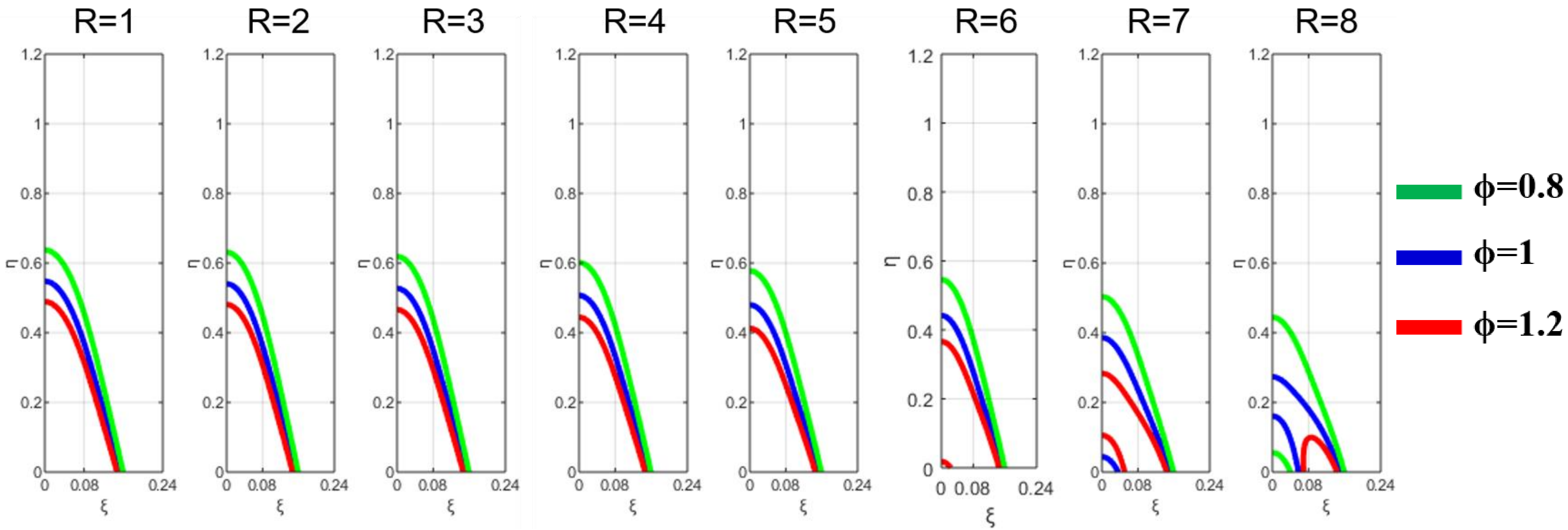
$\Omega = 20\%$

$\Omega = 40\%$



Results and Discussion - Flame Shape Prediction (Ar)

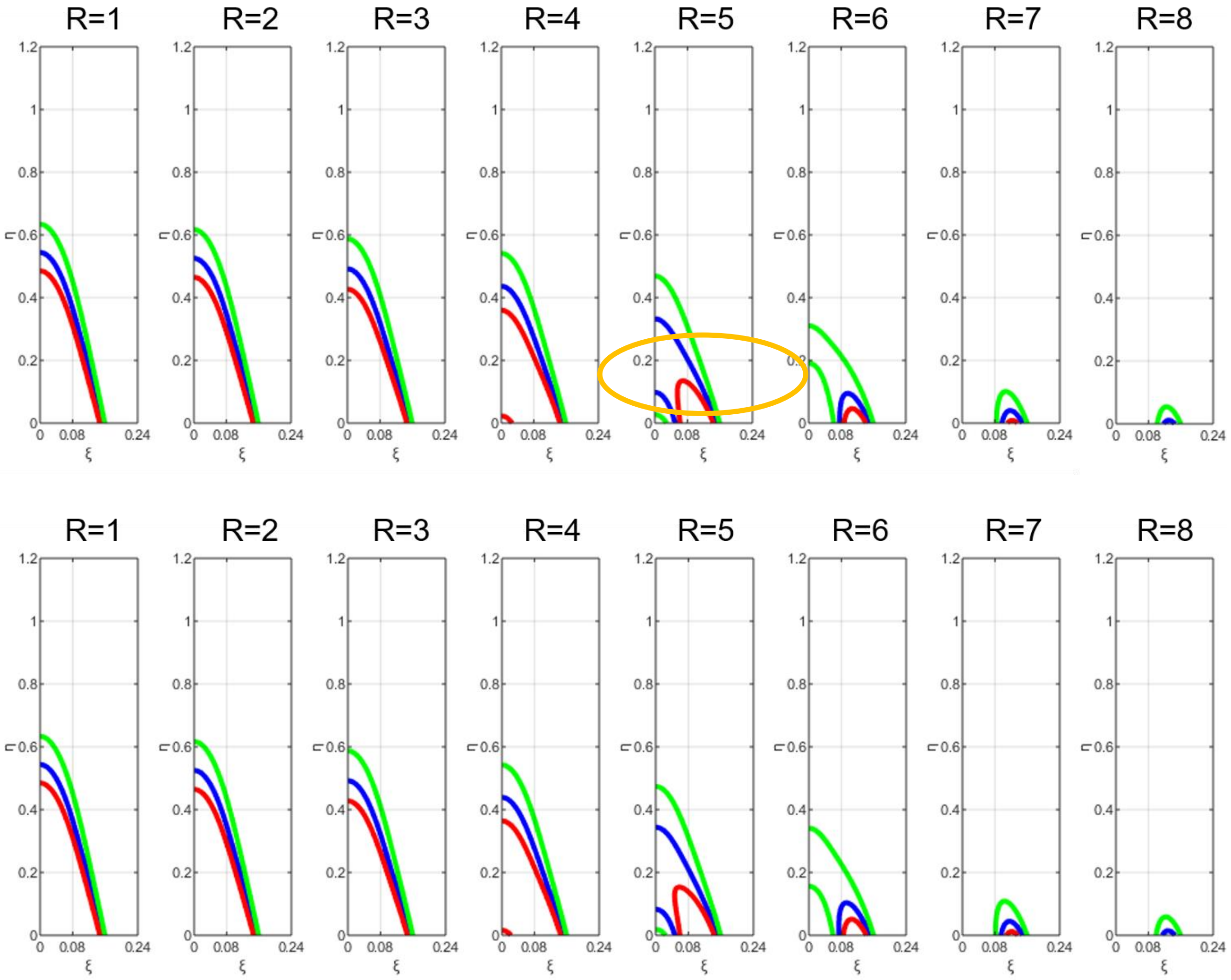
$\Omega = 60\%$



Results and Discussion - Flame Shape Prediction (He)

$\Omega = 20\%$

$\Omega = 40\%$



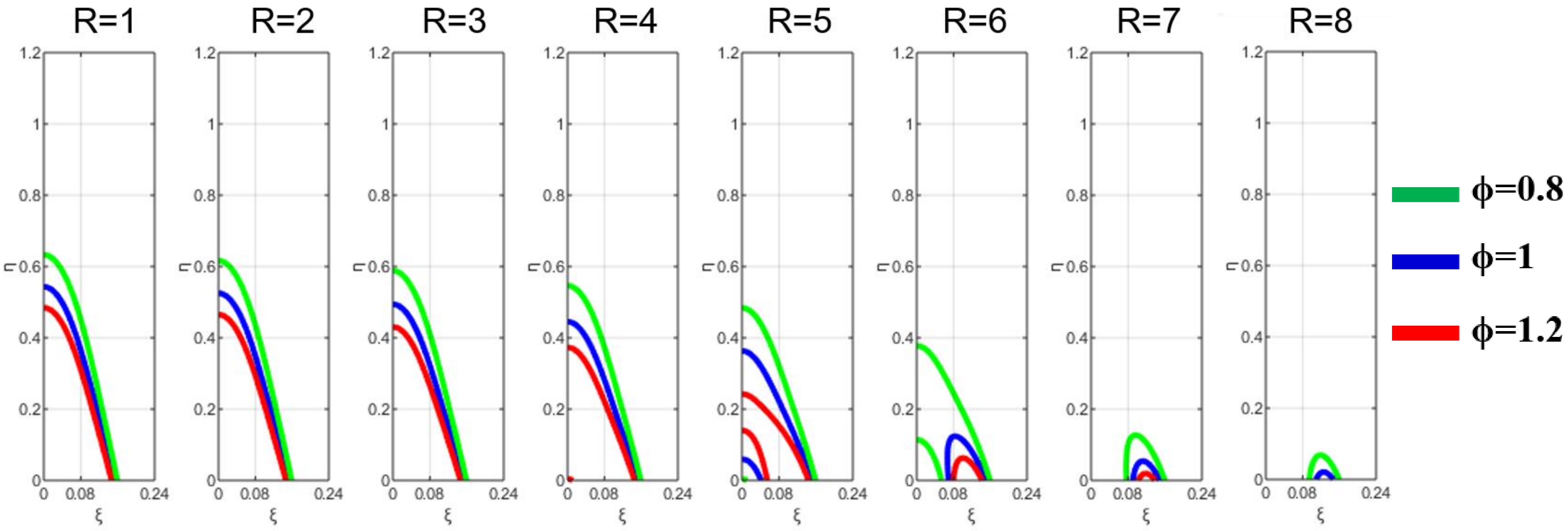
ϕ=0.8

ϕ=1

ϕ=1.2

Results and Discussion - Flame Shape Prediction (He)

$\Omega = 60\%$

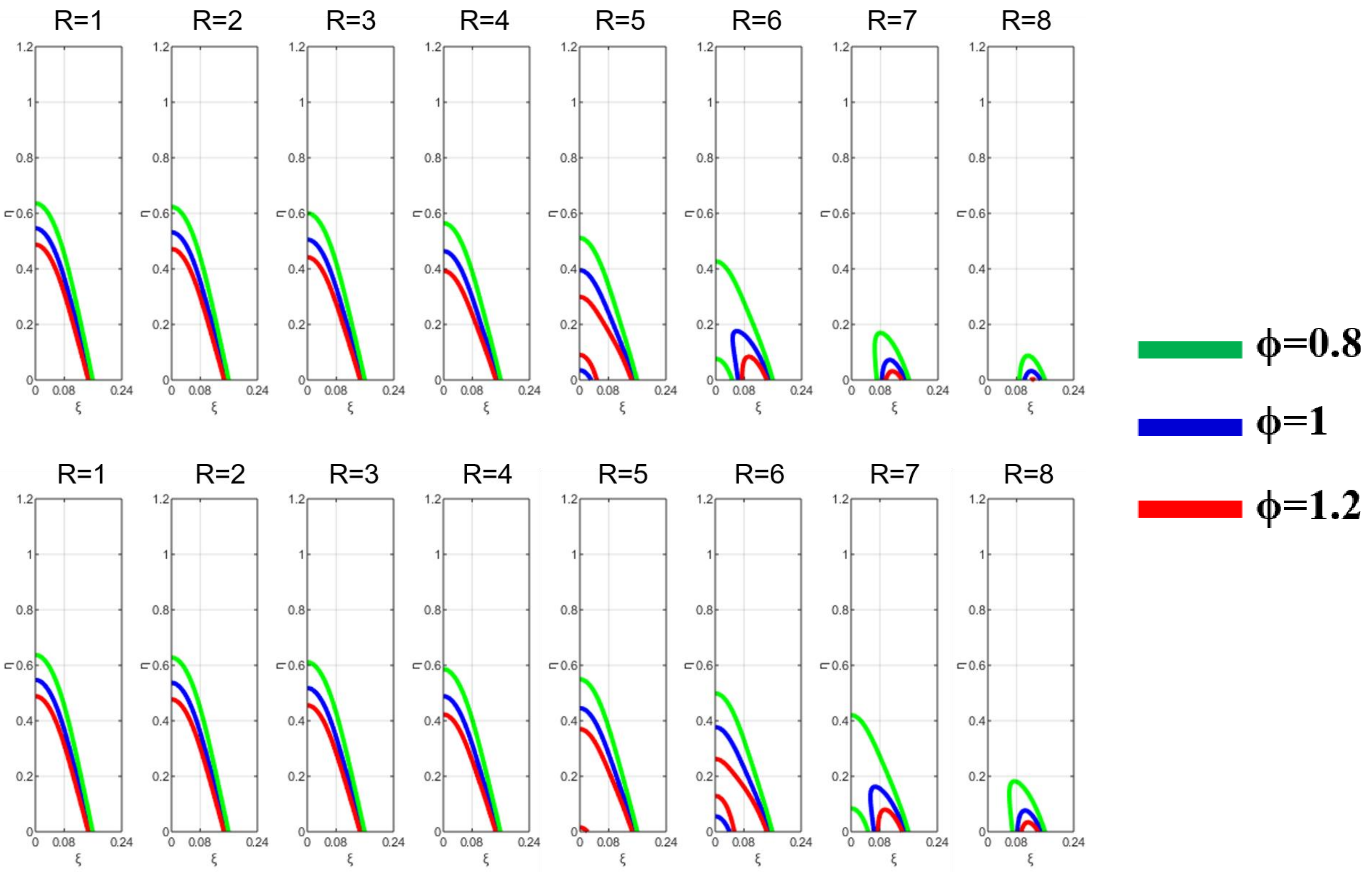


- Helium addition do not affect the velocity ratio of IDF formation.

Results and Discussion - Flame Shape Prediction (CO_2)

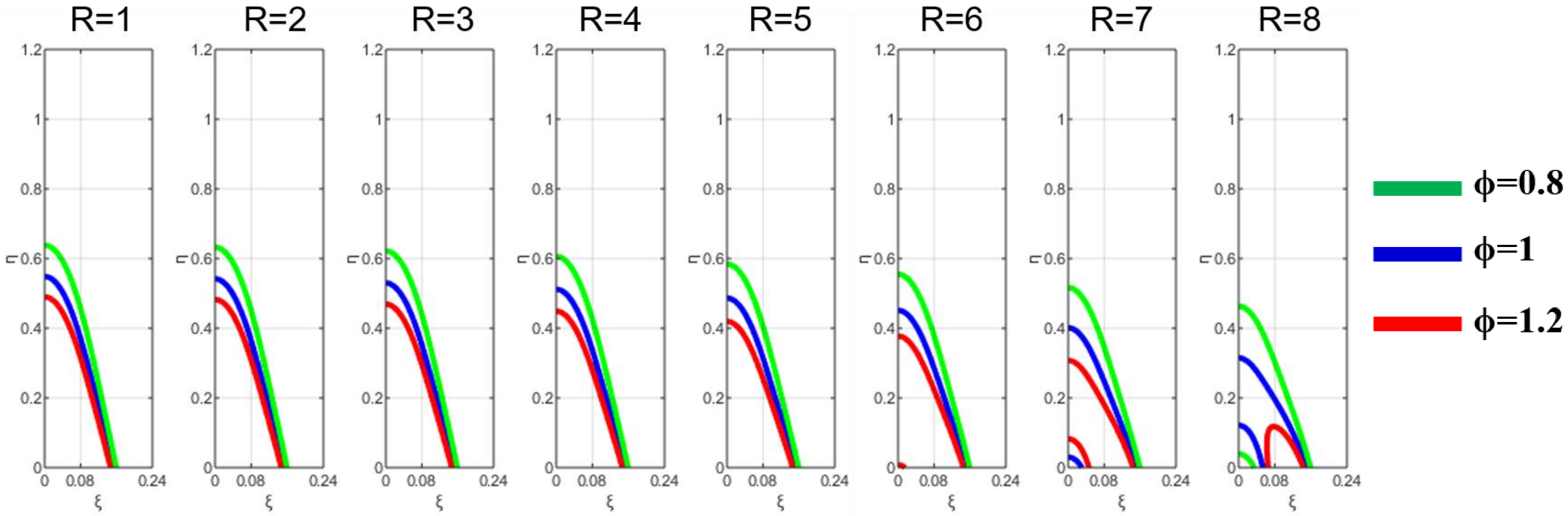
Ω = 20%

Ω = 40%



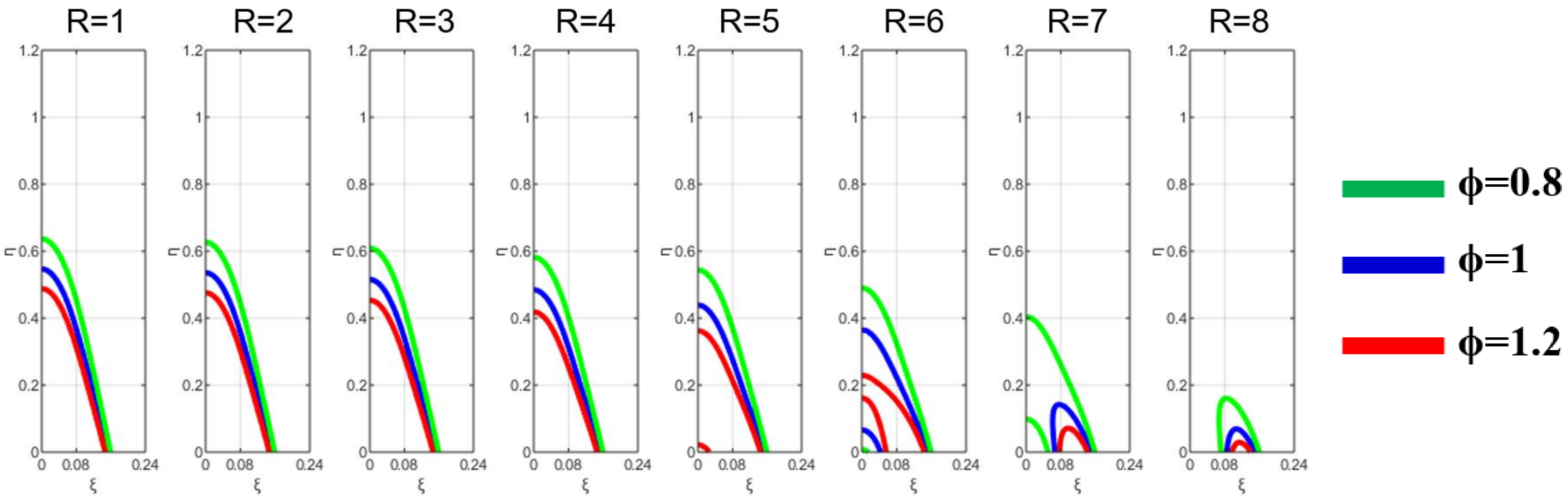
Results and Discussion - Flame Shape Prediction (CO_2)

$\Omega = 60\%$



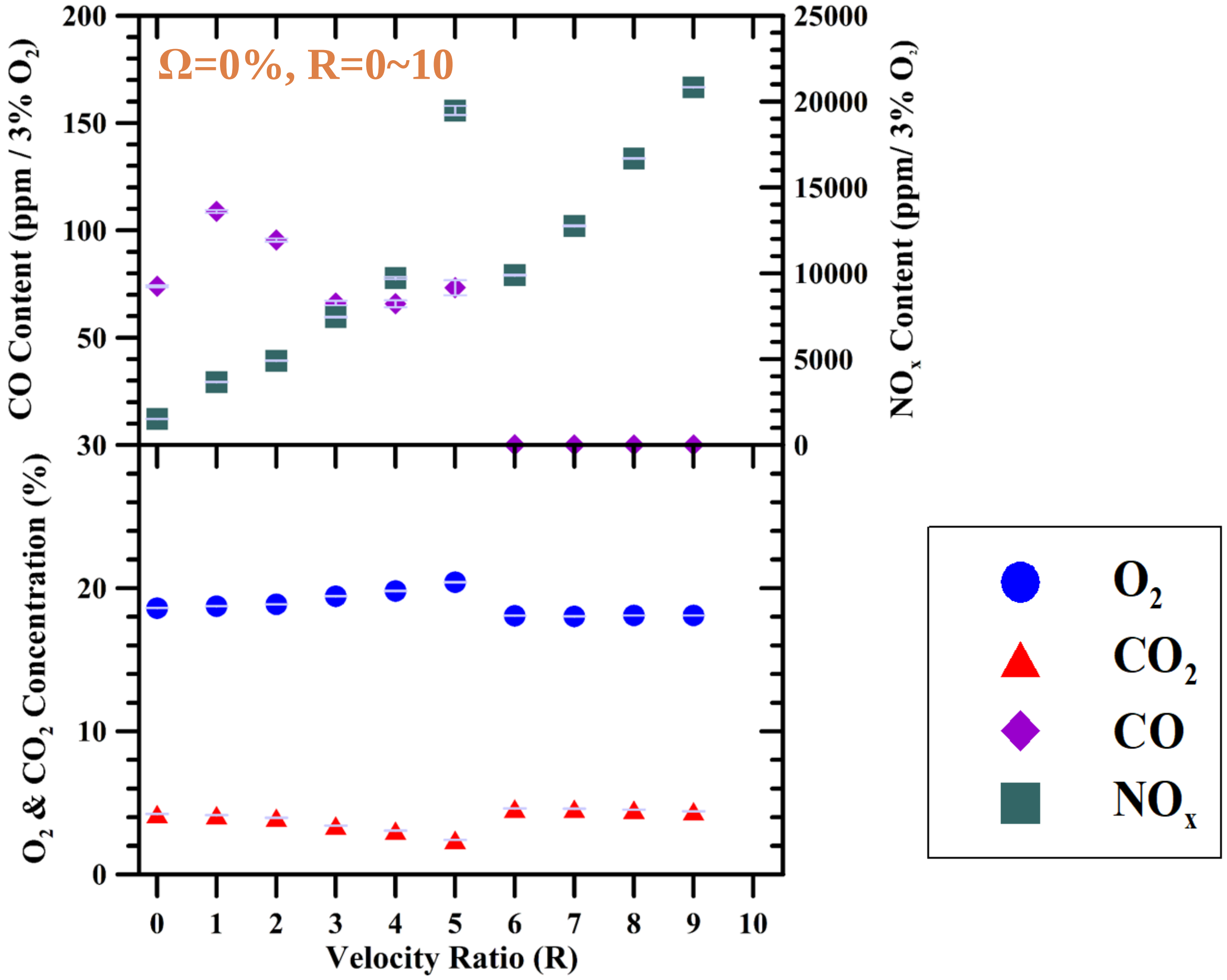
Results and Discussion - Flame Shape Prediction

40% Ar

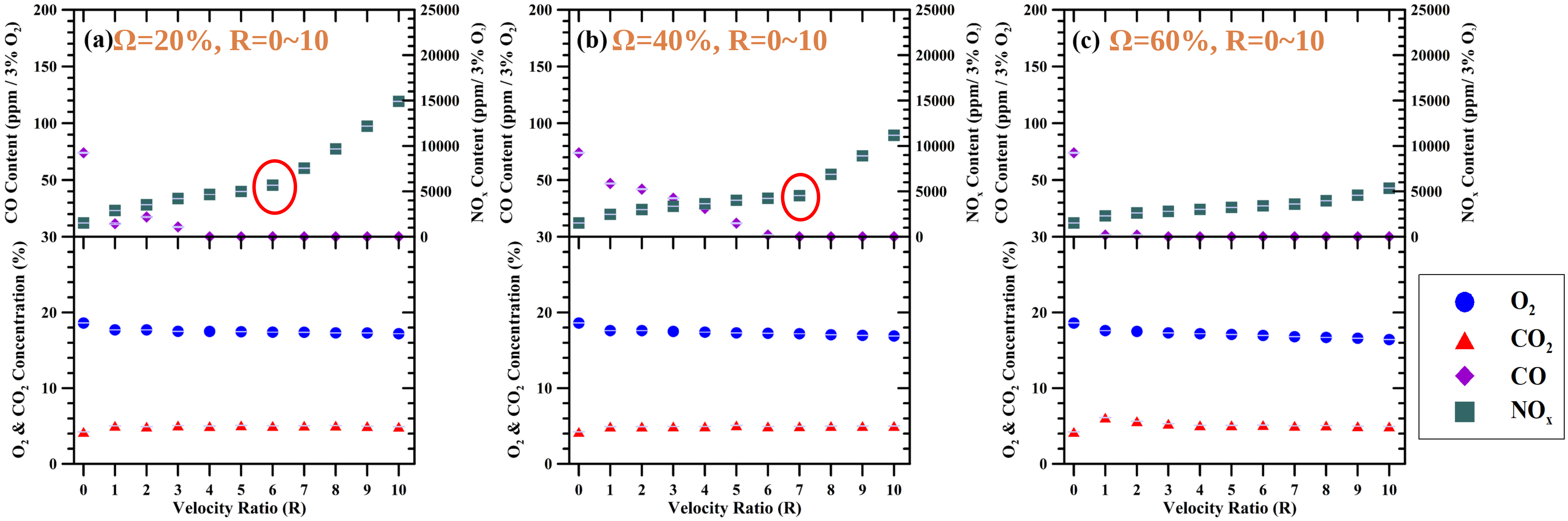


- The step of IDF formation :
  - ✓ Normal diffusion formation.
  - ✓ The branch of fuel-rich, stoichiometric and fuel-lean for inner flame form sequentially.
  - ✓ The branch of fuel-rich, stoichiometric and fuel-lean for inner and outer flame form an envelope sequentially and then form an opened-tip IDF.

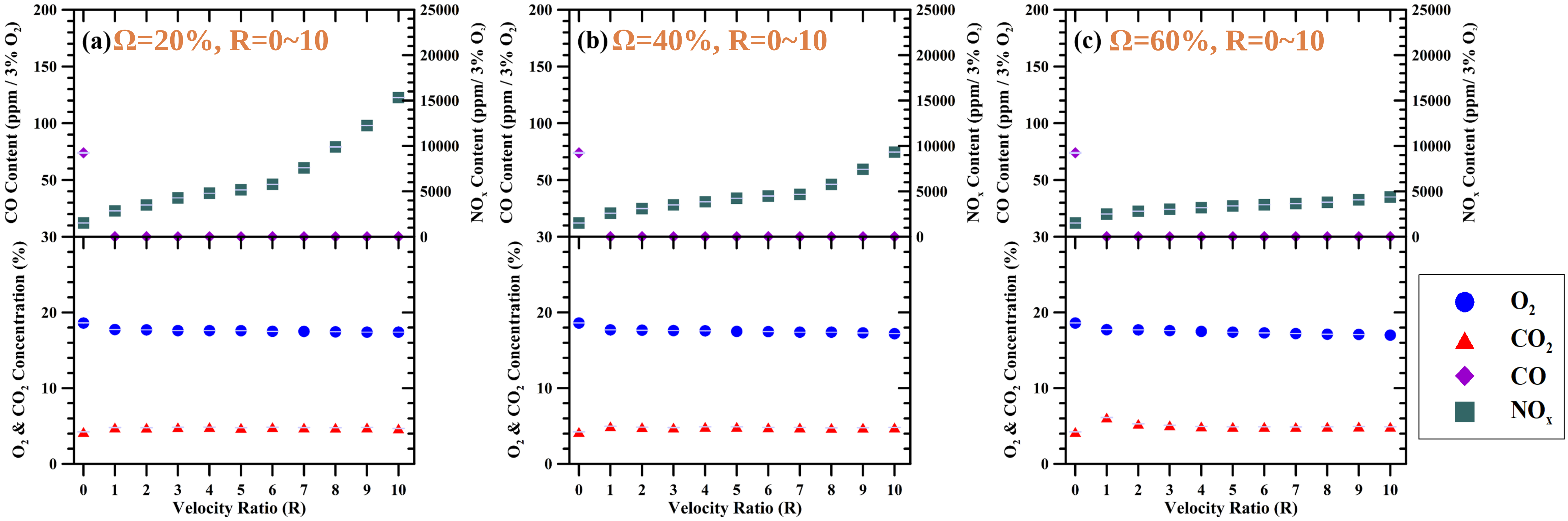
Results and Discussion - Pollutant Emission



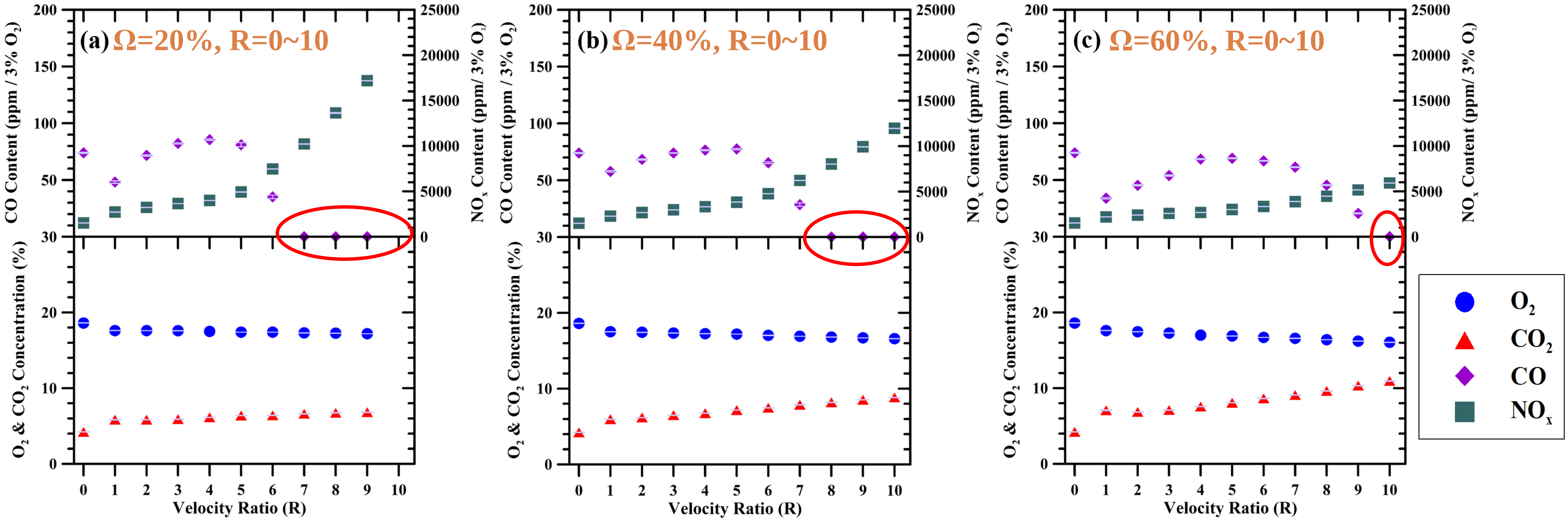
Results and Discussion - Pollutant Emission (Ar)



Results and Discussion - Pollutant Emission (He)



Results and Discussion - Pollutant Emission (CO_2)

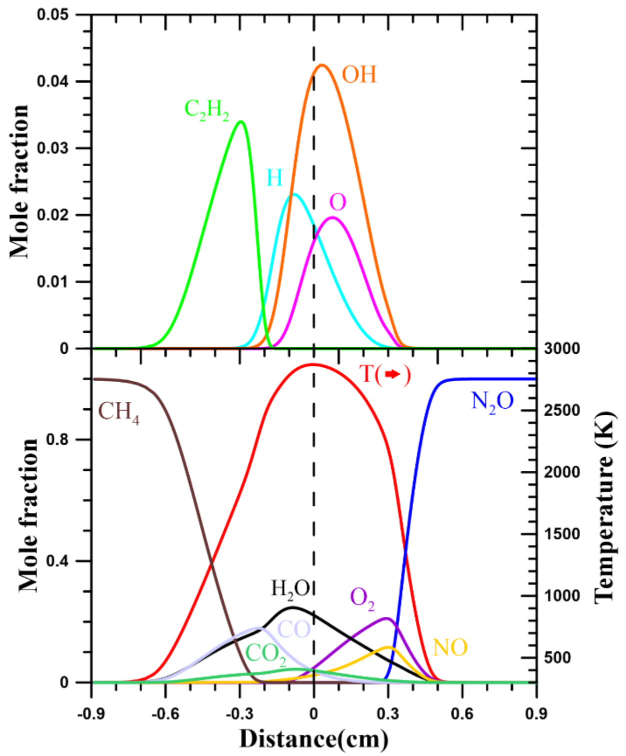


- The formation of CO:
  - ✓ Temperature decrease result in incomplete combustion reaction
  - ✓ CO_2 convert to CO

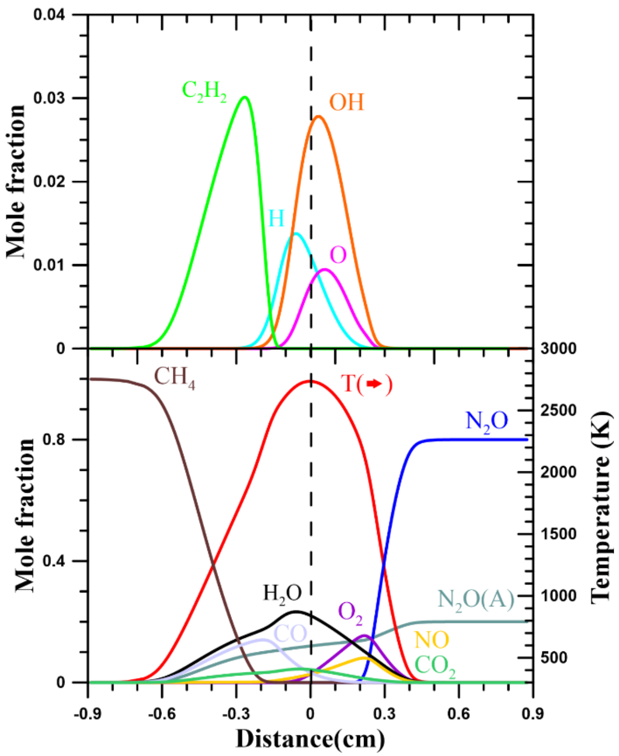
Results and Discussion – Flame structure (Ar)

Ω = 20%

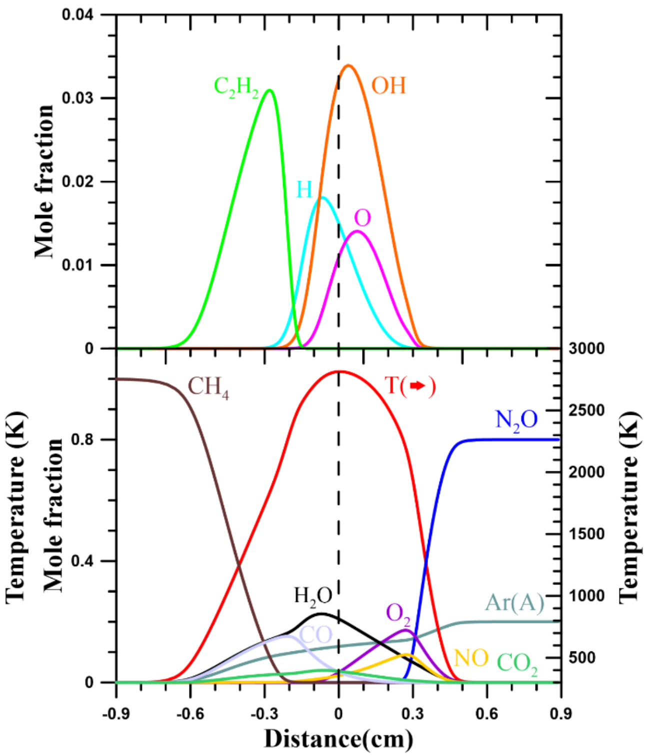
Base case



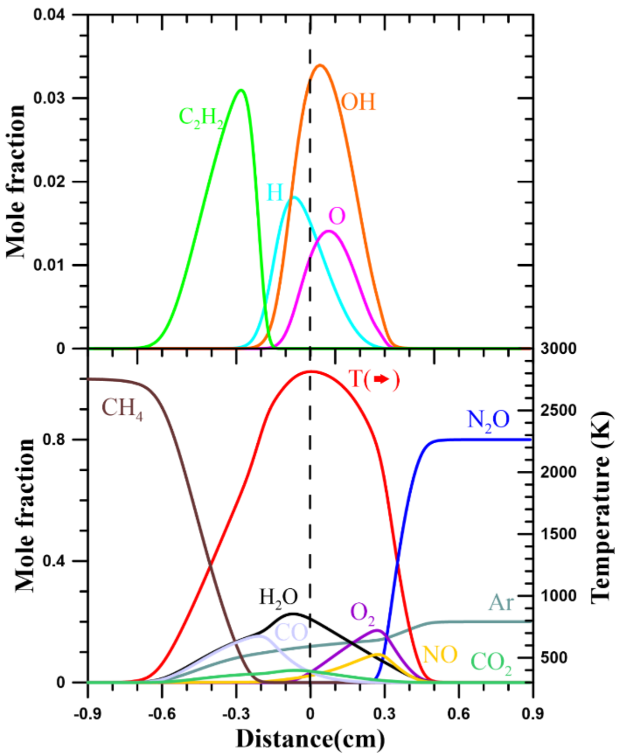
Case I



Case IT



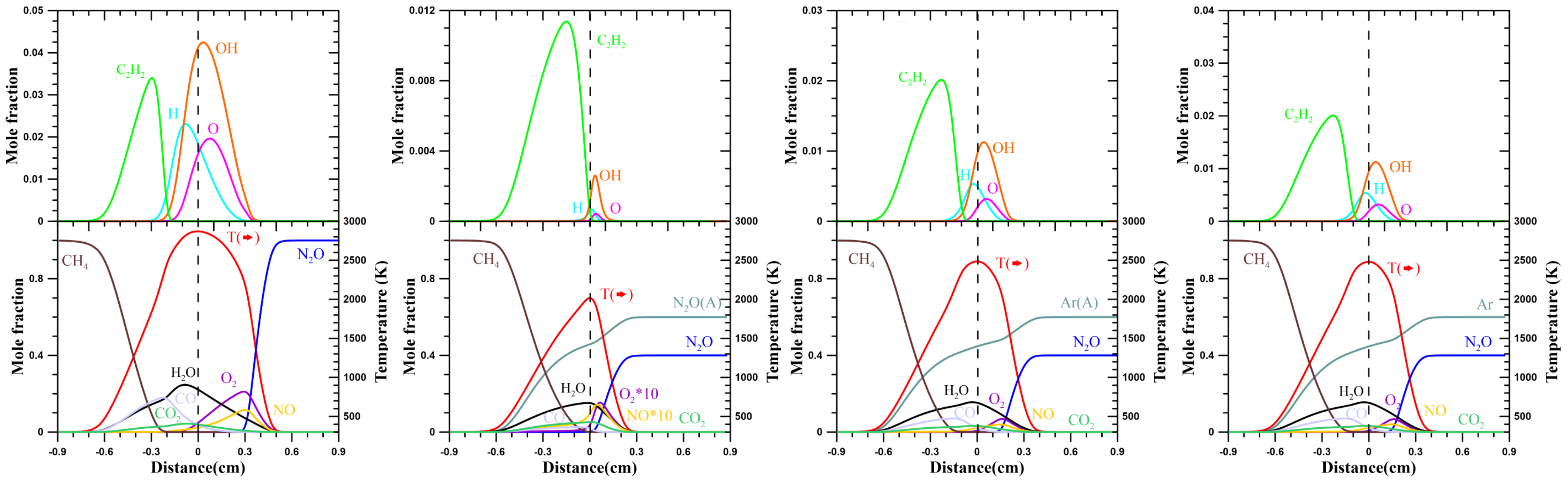
Case ITC



Results and Discussion – Flame structure (Ar)

Ω = 60%

Base case   Case I   Case IT   Case ITC



Results and Discussion – Flame structure (Ar)

- **The effects of Ar addition on flame structure**

  - ✓ Inert effect mainly affects the decrease of intermediate products and flame temperature.

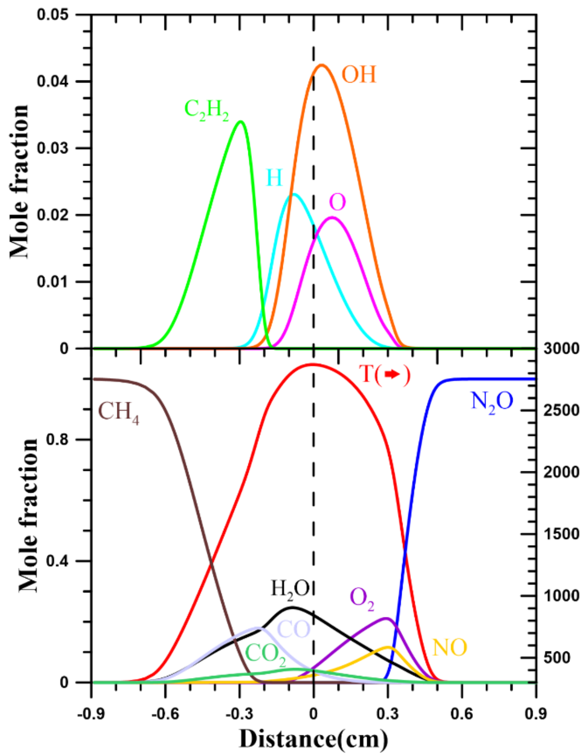
  - ✓ Thermal/diffusion effect affects the increase of intermediate products and flame temperature.

  - ✓ Chemical effect dominate inhibition of C_2H_2 at high Ar concentration.

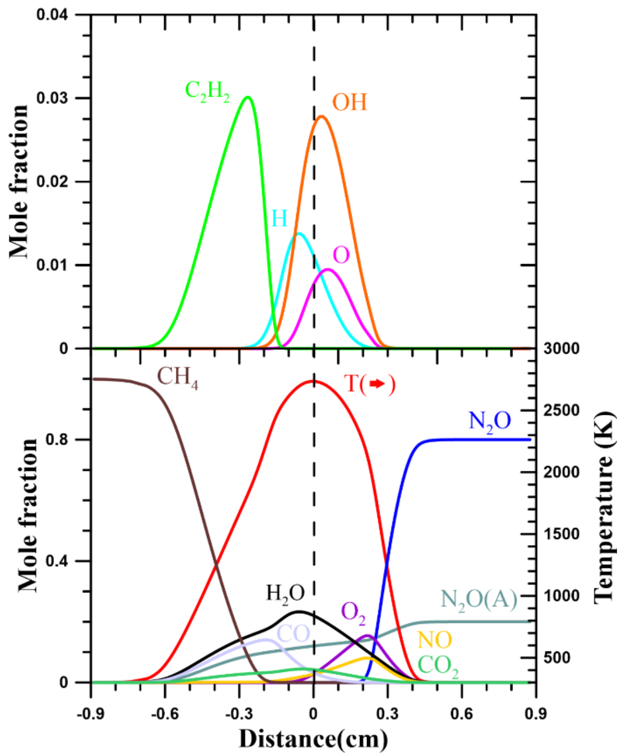
Results and Discussion – Flame structure (CO2)

$\Omega = 20\%$

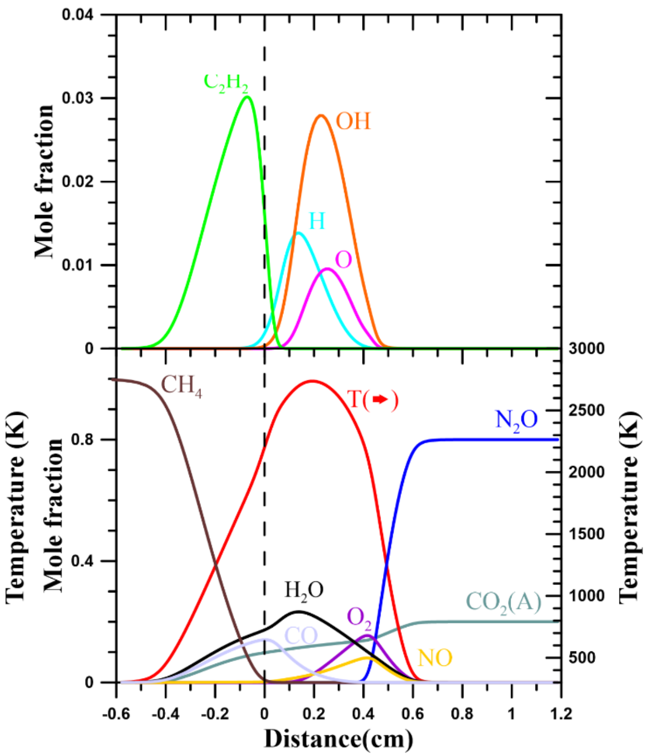
Base case



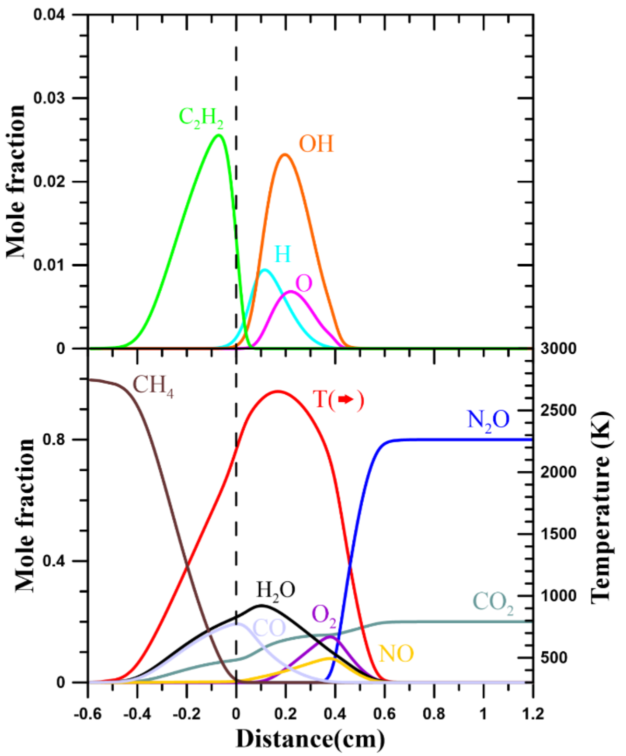
Case I



Case IT



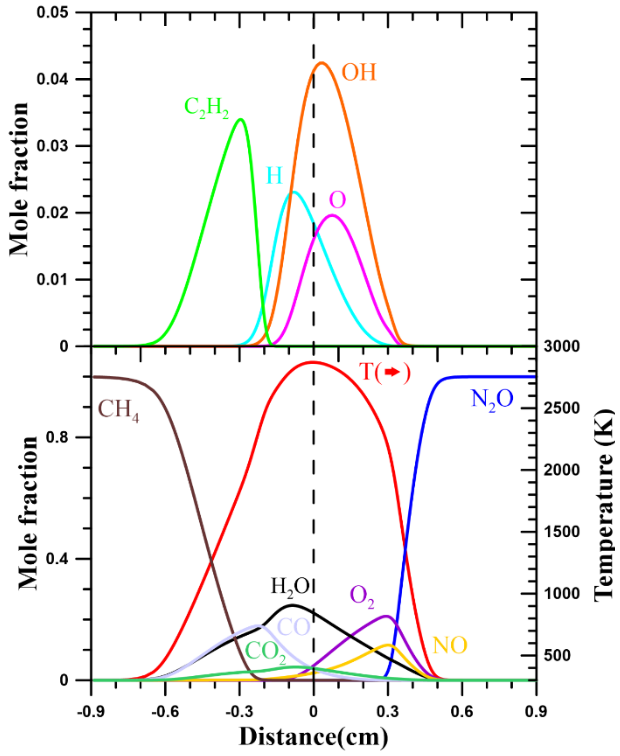
Case ITC



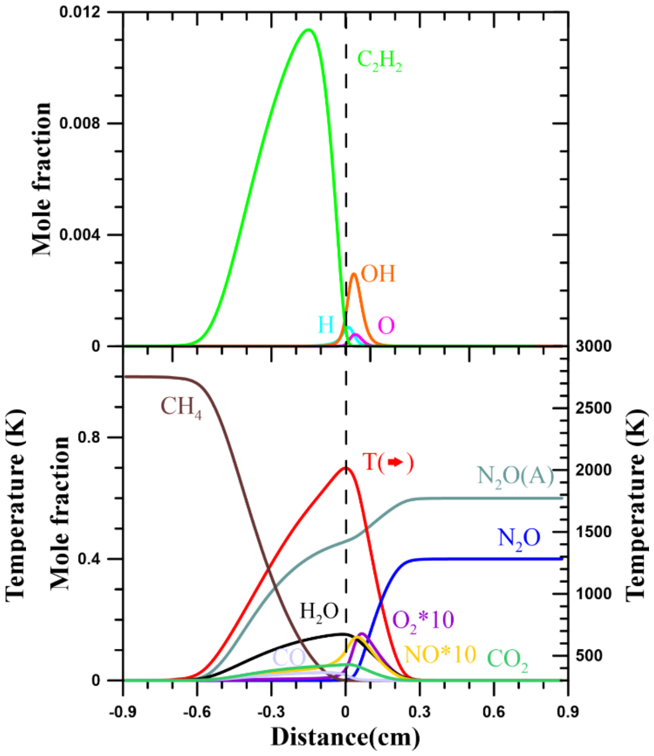
Results and Discussion – Flame structure (CO2)

$\Omega = 60\%$

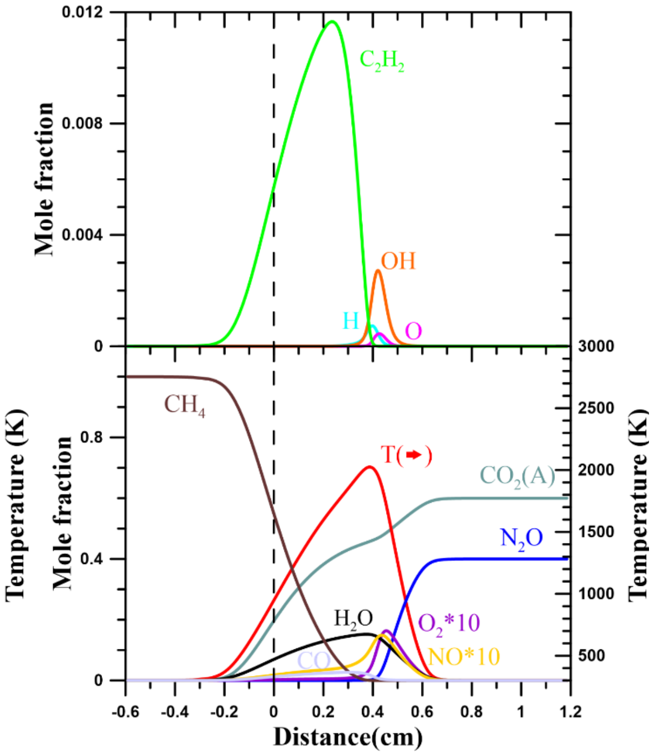
Base case



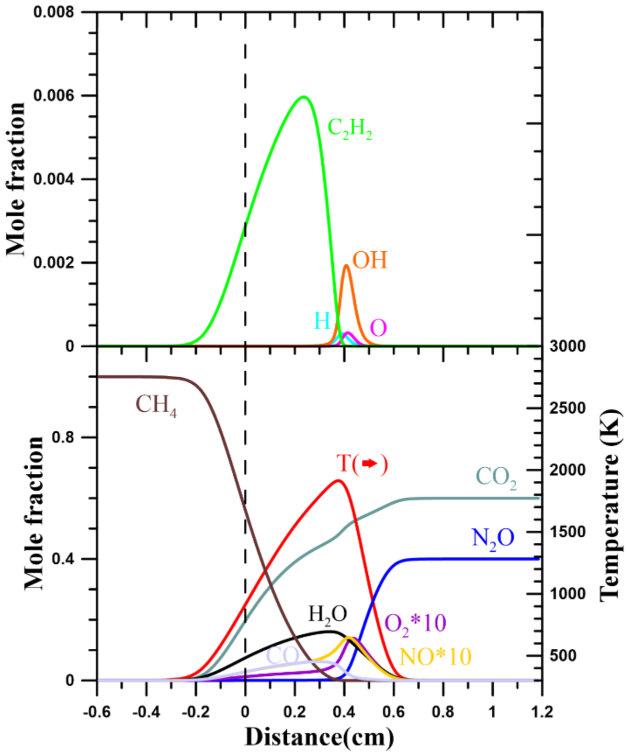
Case I



Case IT



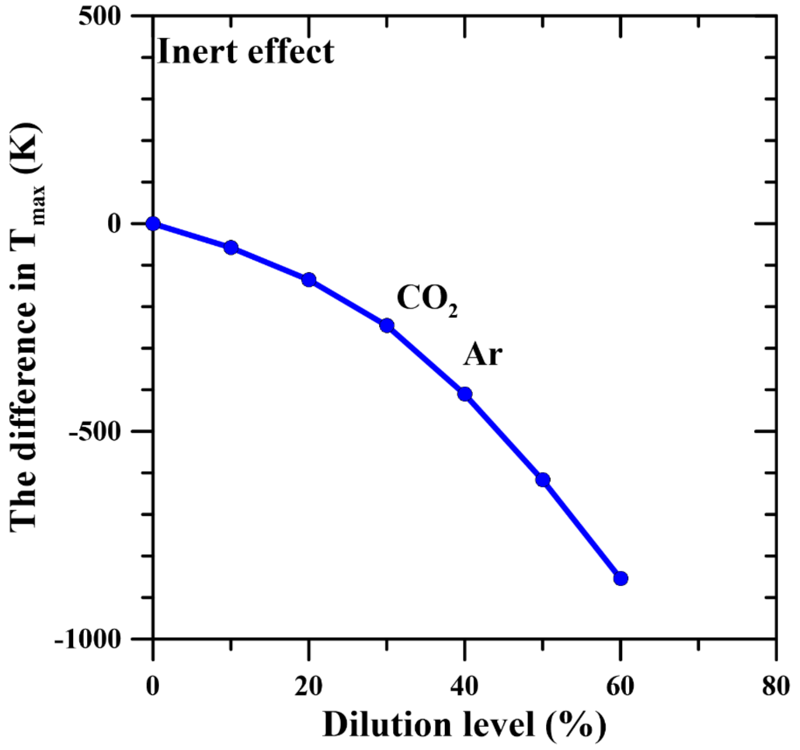
Case ITC

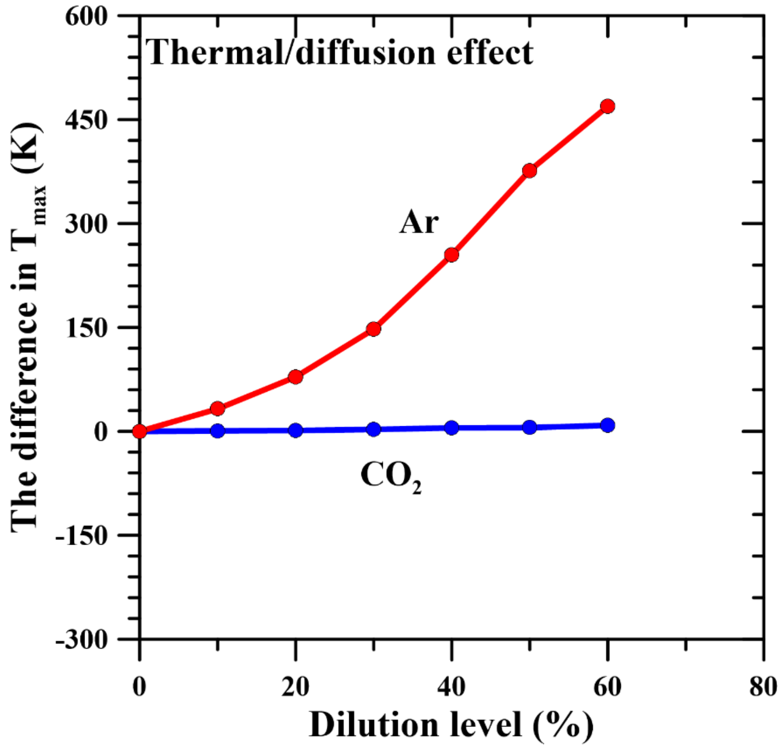


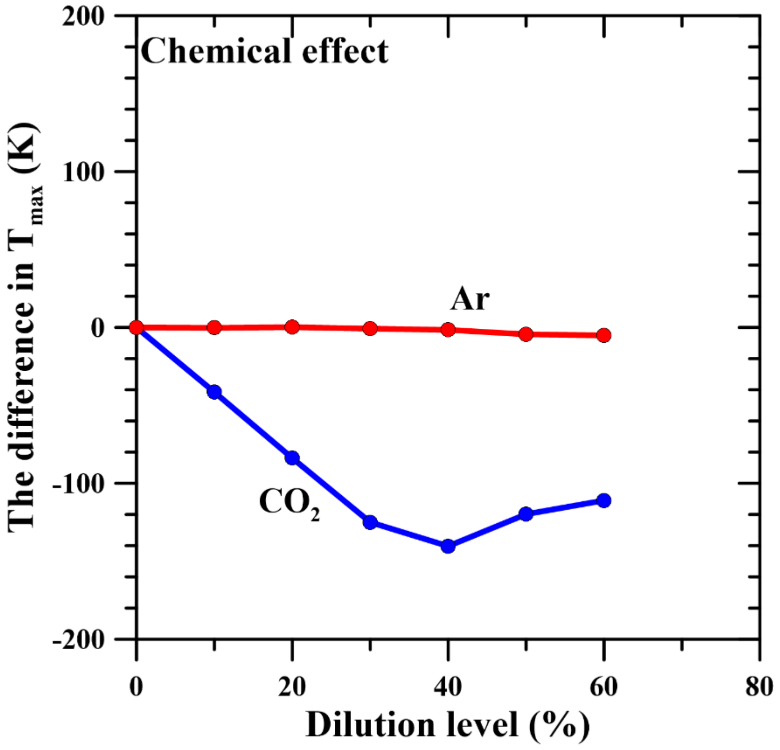
Results and Discussion – Flame structure (CO_2)

- **The effects of CO_2 addition on flame structure**
  - ✓ Thermal/diffusion effect does not have much effect on the flame structure.
  - ✓ Chemical effect mainly affects the decrease of intermediate products and flame temperature.

Results and Discussion – Flame Temperature

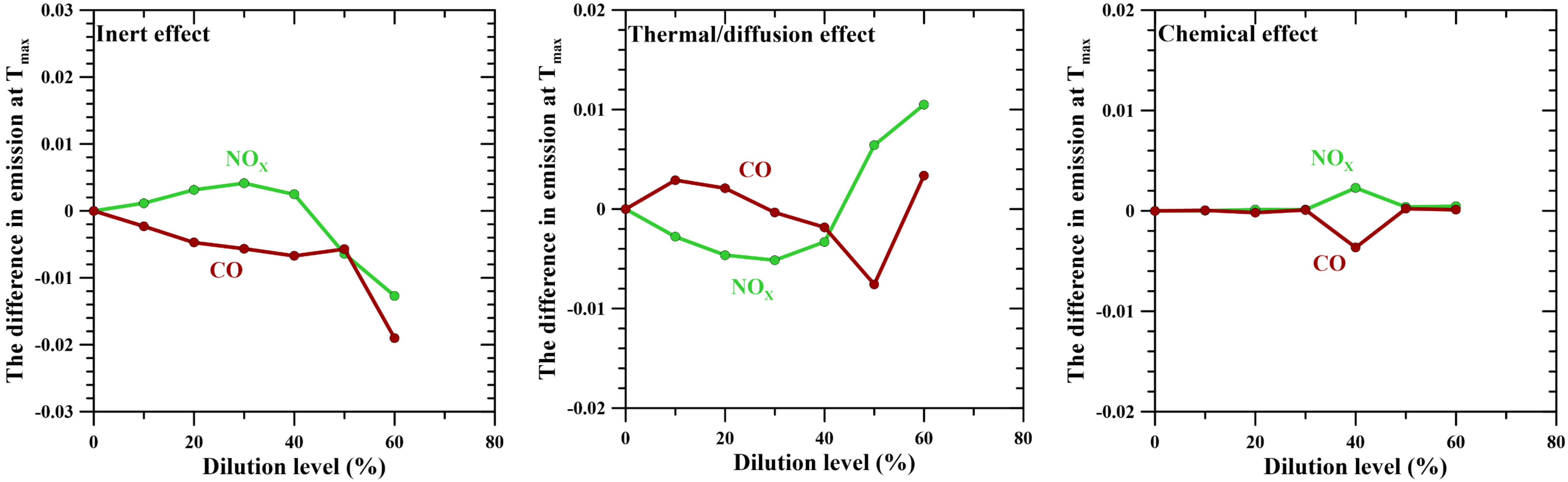




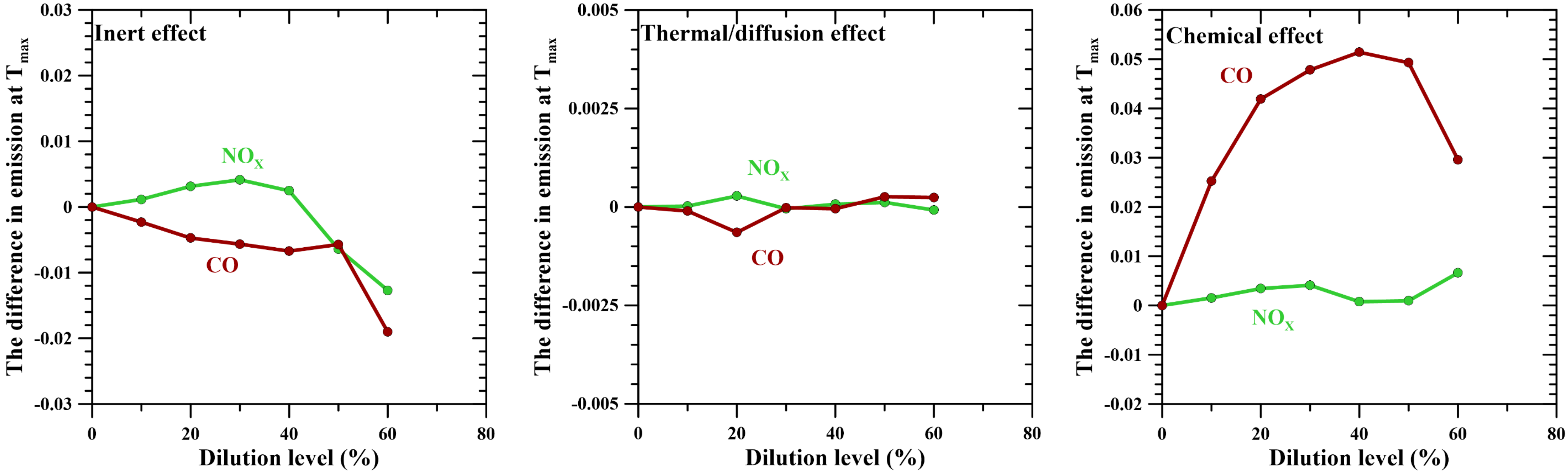


Ar	CO_2
20.8 kJ/kmol·K	37.2 kJ/kmol·K

Results and Discussion – Pollutant Emission (Ar)

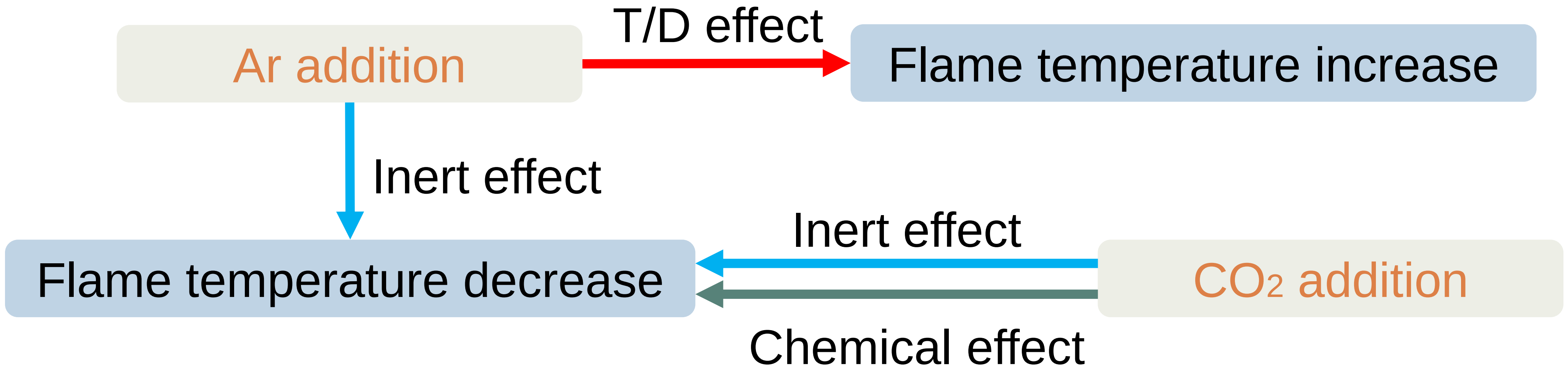


Results and Discussion – Pollutant Emission (CO_2)

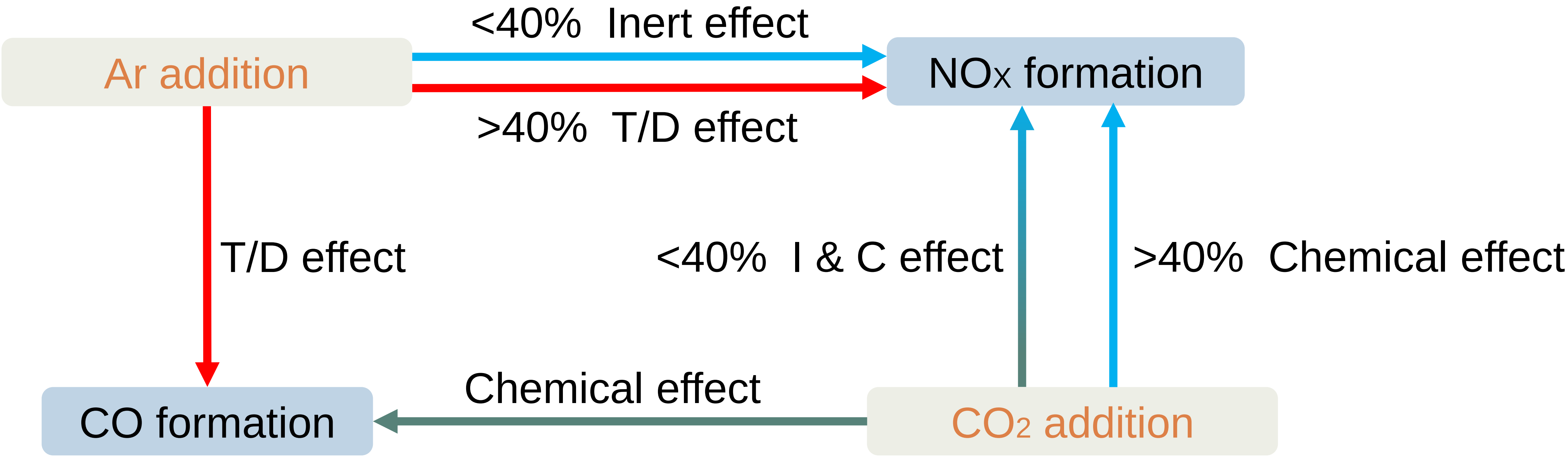


Results and Discussion – Flame Temperature & Pollutant Emission

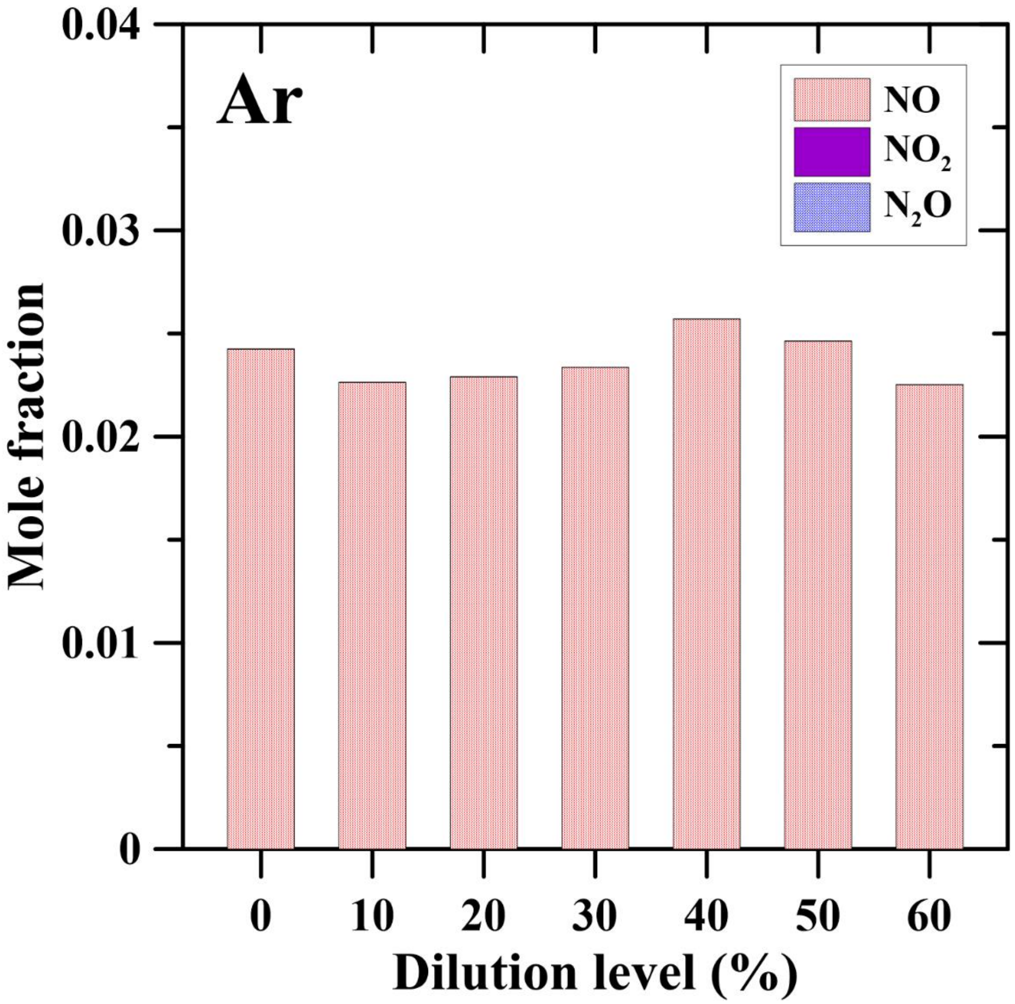
- **The effects of diluent addition on flame temperature**

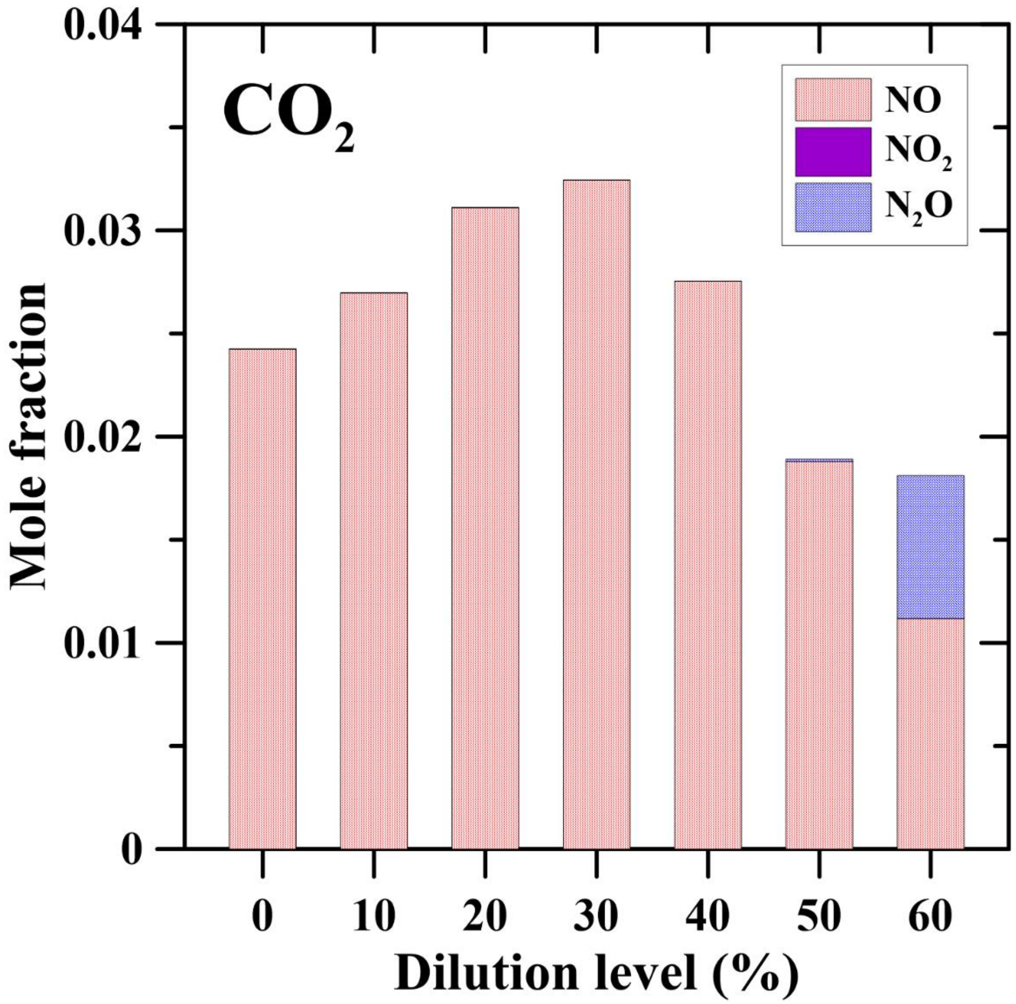


- **The effects of diluent addition on pollutant emission**



Results and Discussion – Mechanism of NO Formation



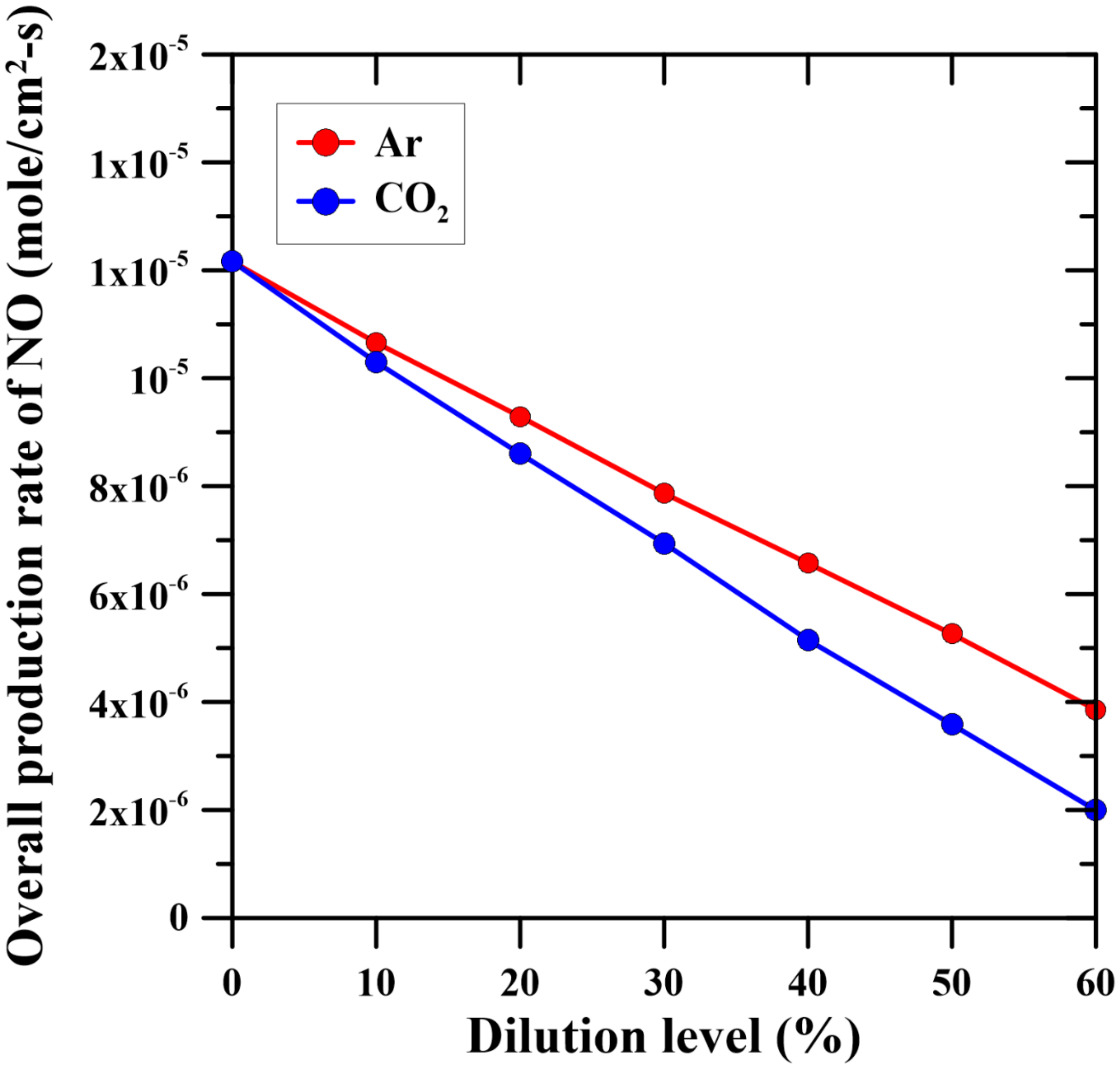


Results and Discussion – Mechanism of NO Formation

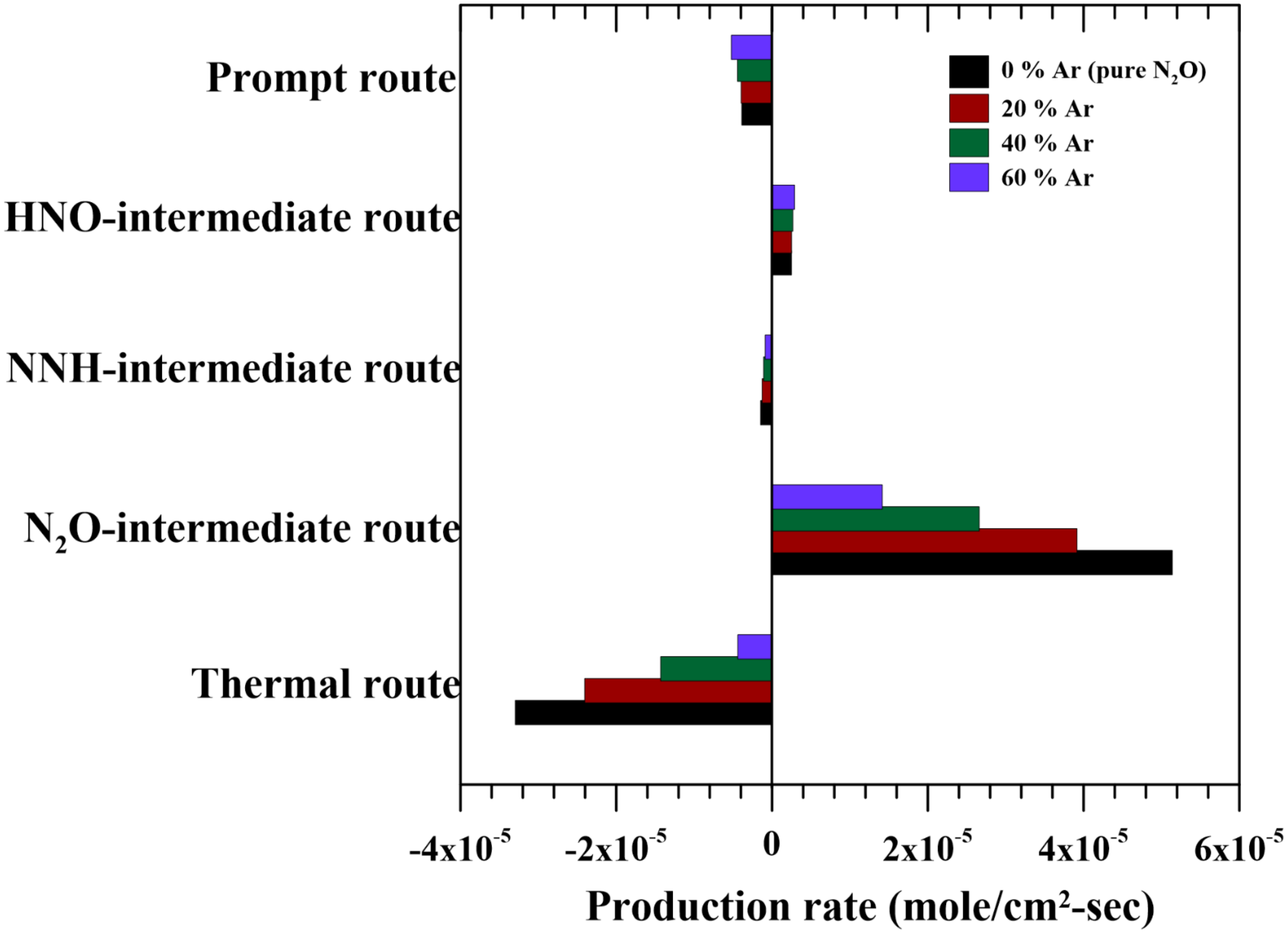
Reaction number and reaction steps.	
Thermal route	
R178	$N + NO = N_2 + O$
R179	$N + O_2 = NO + O$
R180	$N + OH = NO + H$
N_2O-intermediate route	
R182	$N_2O + O = 2NO$
R183	$N_2O + H = N_2 + OH$
R185	$N_2O + M = N_2 + O + M$
R199	$NH + NO = N_2O + H$
R228	$NCO + NO = N_2O + CO$
NNH-intermediate route	
R204	$NNH = H + N_2$
R205	$NNH + M = N_2 + H + M$
R208	$NNH + O = NH + NO$
R209	$NNH + H = H_2 + N_2$
HNO-intermediate route	
R212	$H + NO + M = HNO + M$
R213	$HNO + O = NO + OH$
R214	$HNO + H = H_2 + NO$
R215	$HNO + OH = NO + H_2O$
Prompt route	
R245	$C + NO = CO + N$
R246	$CH + NO = HCN + O$
R249	$CH_2 + NO = H + HNCO$
R255	$CH_3 + NO = HCN + H_2O$
R274	$HCCO + NO = HCNO + CO$
R283	$N + CO_2 = NO + CO$

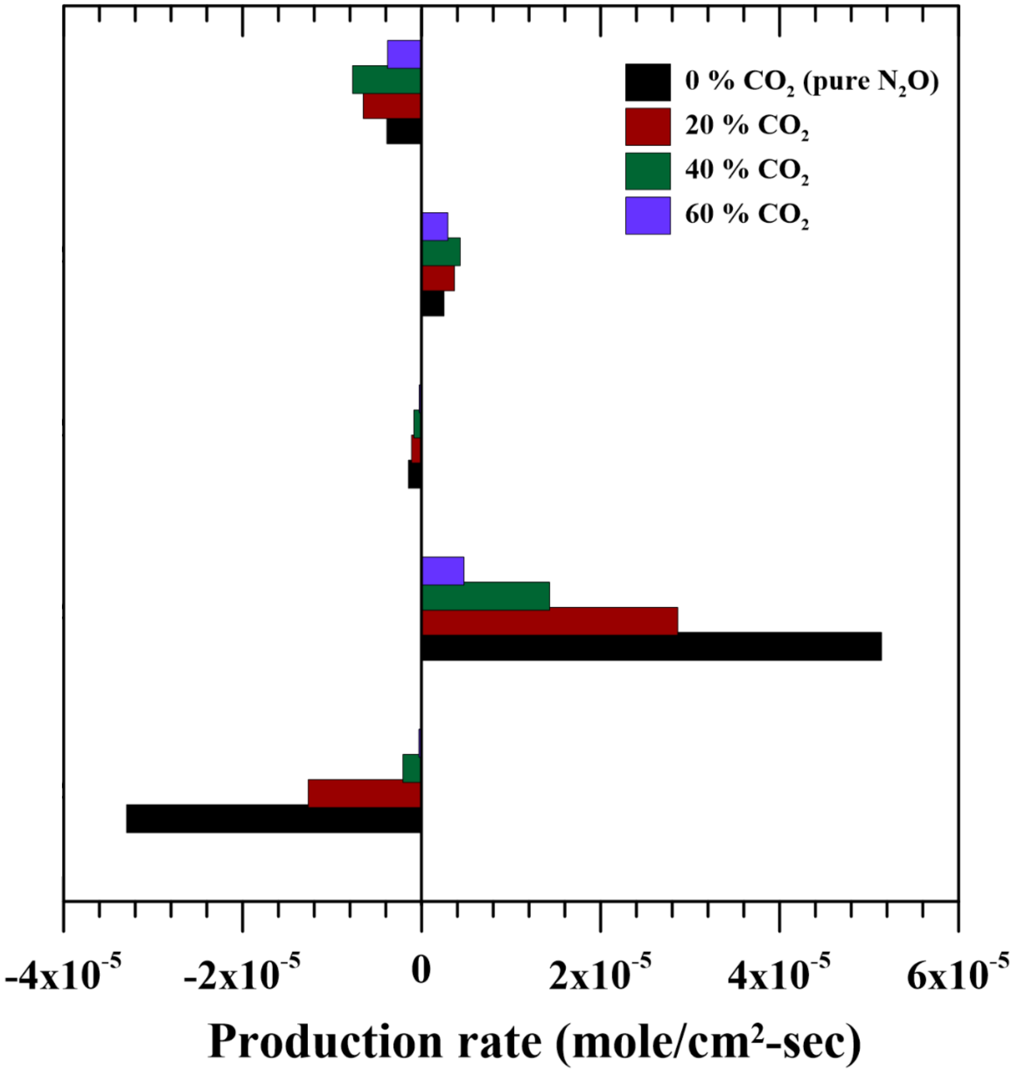
- **Thermal Route**
- **N_2O-intermediate route**
- **NNH-intermediate route**
- **HNO-intermediate route**
- **Prompt route**

Results and Discussion – Mechanism of NO Formation

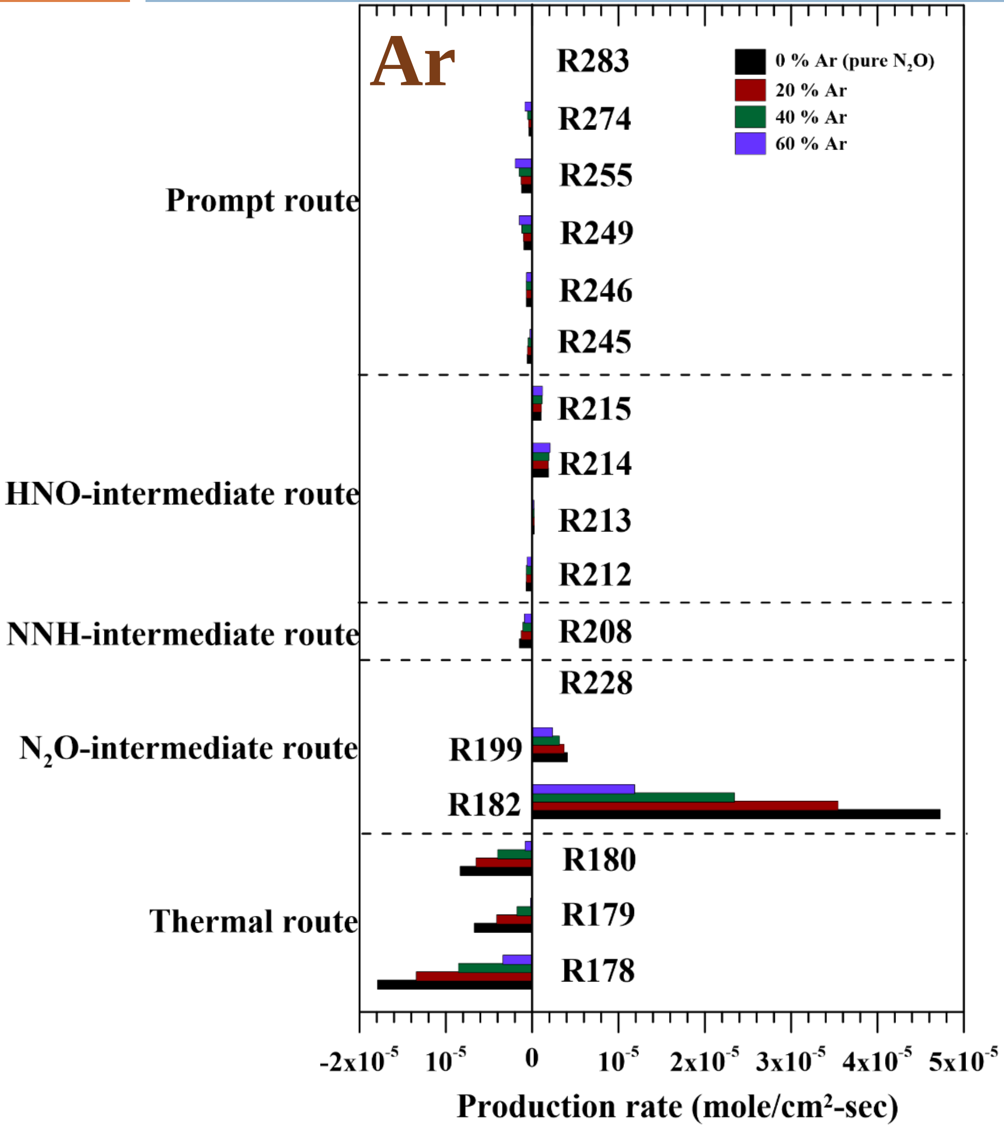


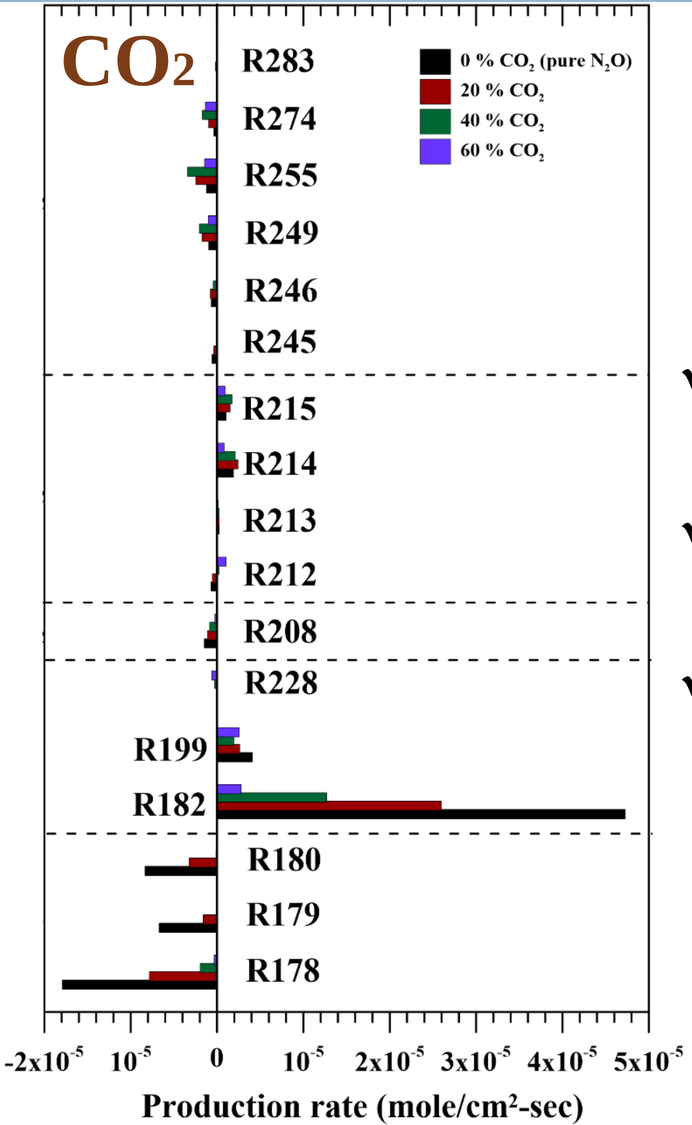
Results and Discussion – Mechanism of NO Formation





Results and Discussion – Mechanism of NO Formation

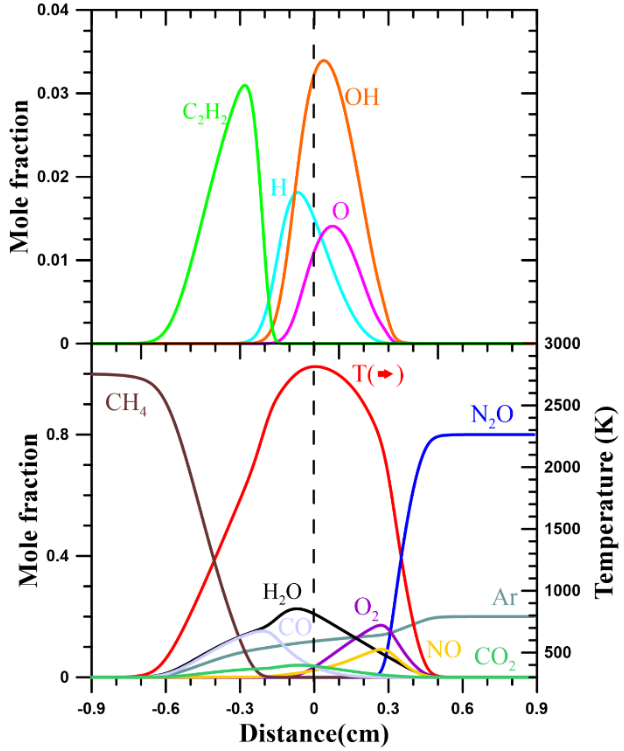




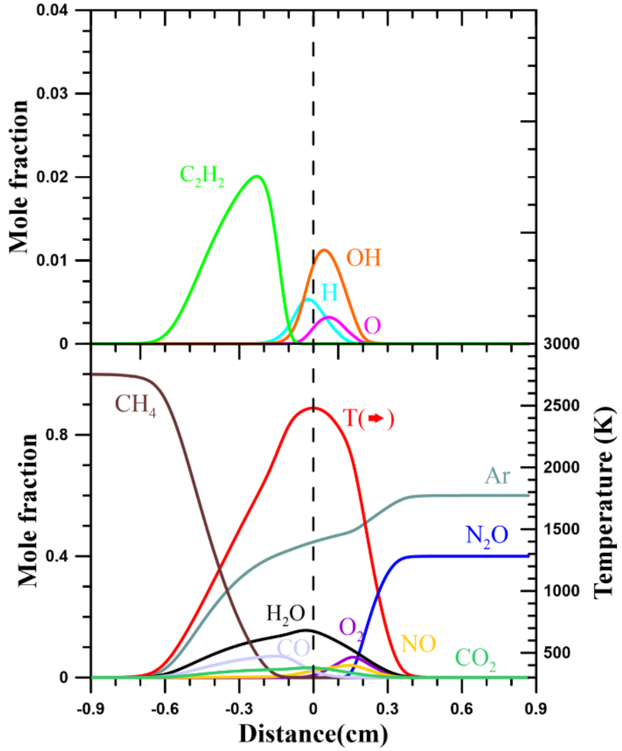
- ✓ **R182:** $N_2O + O = 2NO$
- ✓ **R199:** $NH + NO = N_2O + H$
- ✓ **R178:** $N + NO = N_2 + O$

Results and Discussion – Mechanism of NO Formation

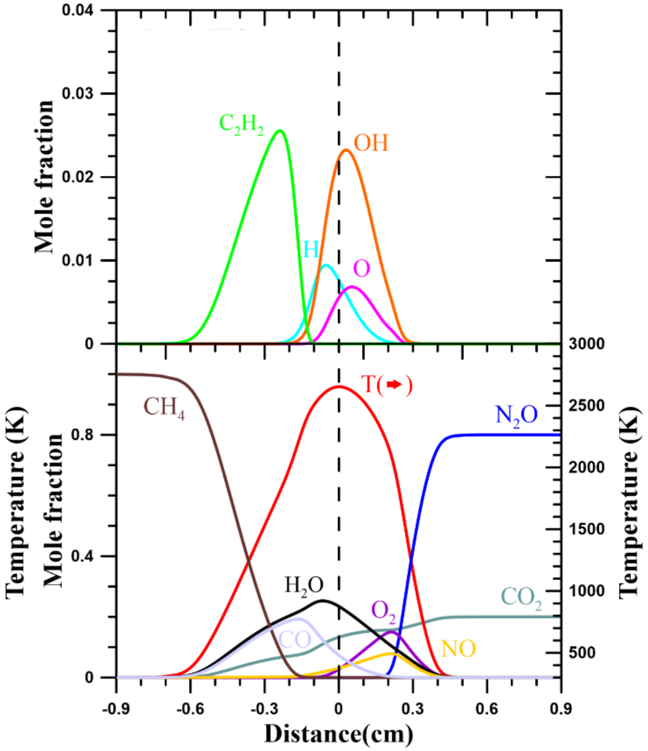
20% Ar



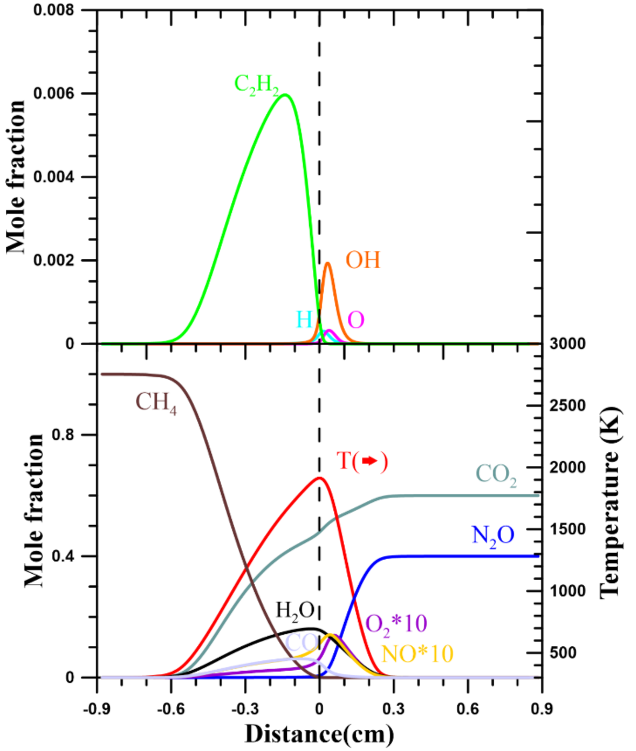
60% Ar



20% CO2



60% CO2



- The O radical decrement rate of CO_2 addition is larger than Ar addition.

R182: $N_2O + O = 2NO$

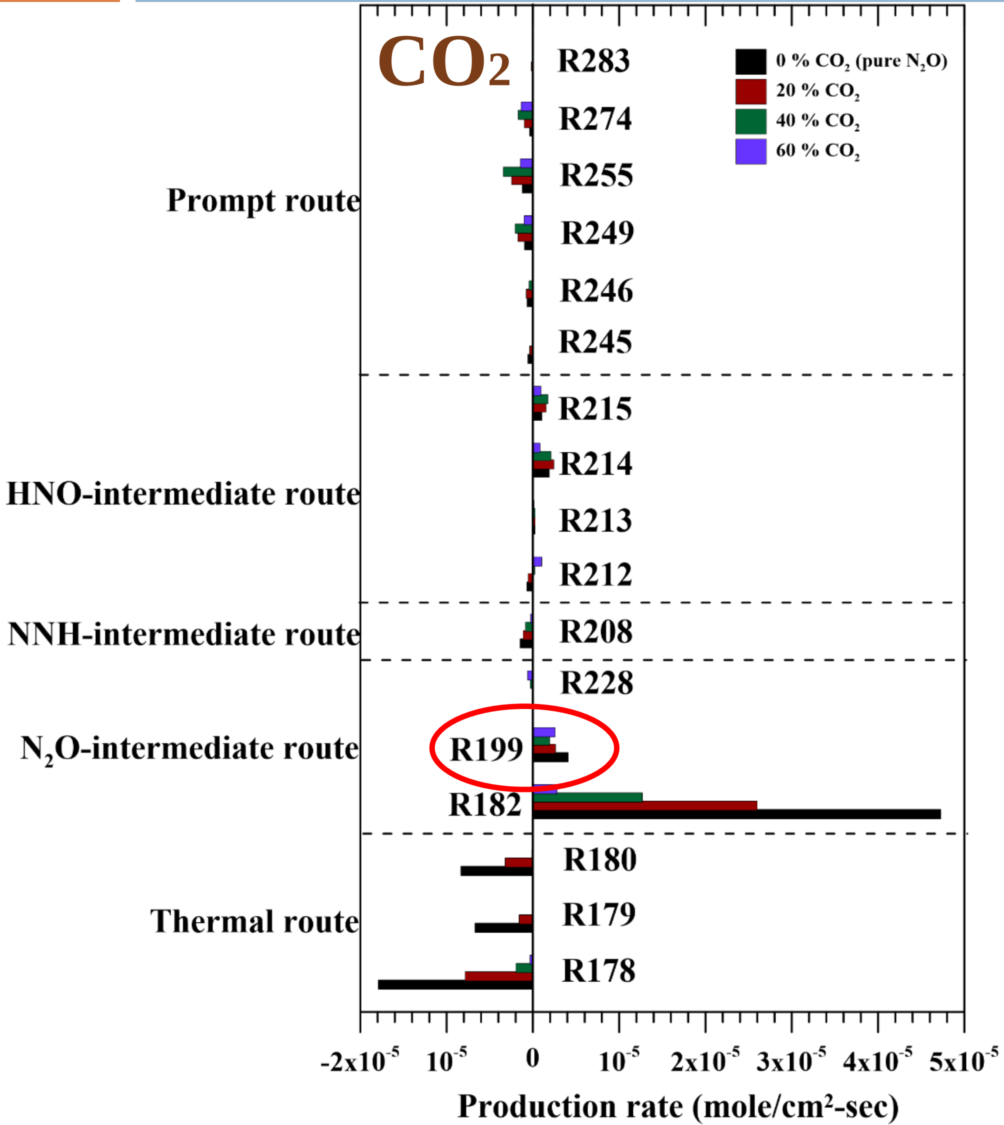
Results and Discussion – Mechanism of NO Formation

Total concentration of species for different diluent			
	O	N	N_2
20% CO_2	0.0014	0.0000007	0.2979
20% Ar	0.0035	0.0000016	0.344

$$\textbf{R178: } N + NO = N_2 + O$$

- N radical concentration under Ar addition is higher than CO_2 addition. It's the reason why the decrement rate of R178 production rate for CO_2 is higher than Ar .

Results and Discussion – Mechanism of NO Formation



Total concentration of species for different diluent			
	NH	H	N_2O
20% CO_2	0.00000074	0.0018	0.8
40% CO_2	0.00000051	0.00034	0.6
60% CO_2	0.00000035	0.000022	0.4

R199: NH + NO = N2O + H

1. **20% ~ 40%**
   - ✓ H radical concentration drops sharply.
2. **40% ~ 60%**
   - ✓ NH radical concentration decrease.

Conclusions

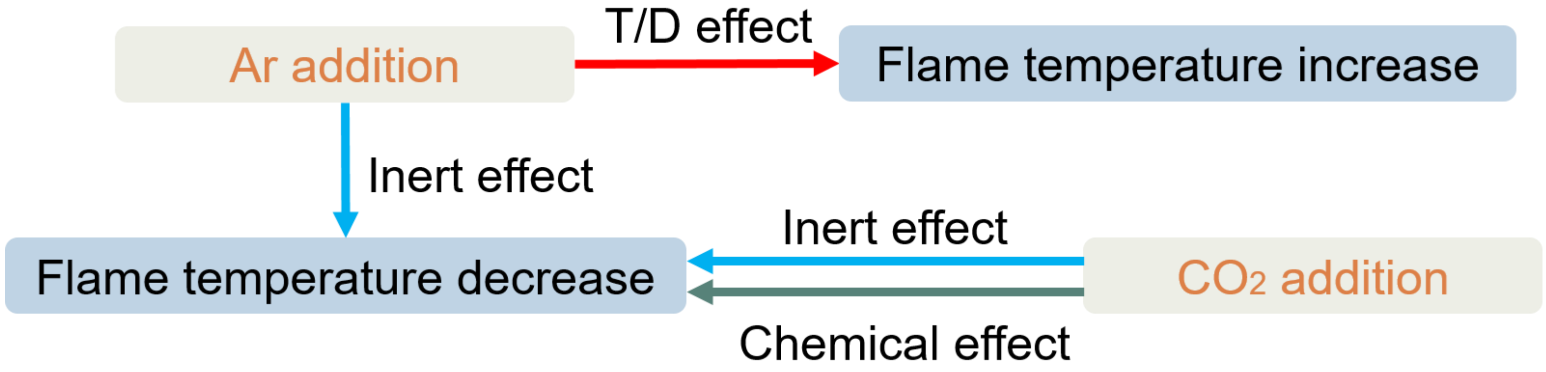
- In flame configuration, the flame types can be divided into five types regardless of diluent addition, namely NDF, PPF, closed-tip IDF, opened-tip IDF and liftoff edge flame.

- The step of IDF formation :
  - ✓ Normal diffusion formation.
  - ✓ The branch of fuel-rich, stoichiometric and fuel-lean for inner flame form sequentially.
  - ✓ The branch of fuel-rich, stoichiometric and fuel-lean for inner and outer flame form an envelope sequentially and then form an opened-tip IDF

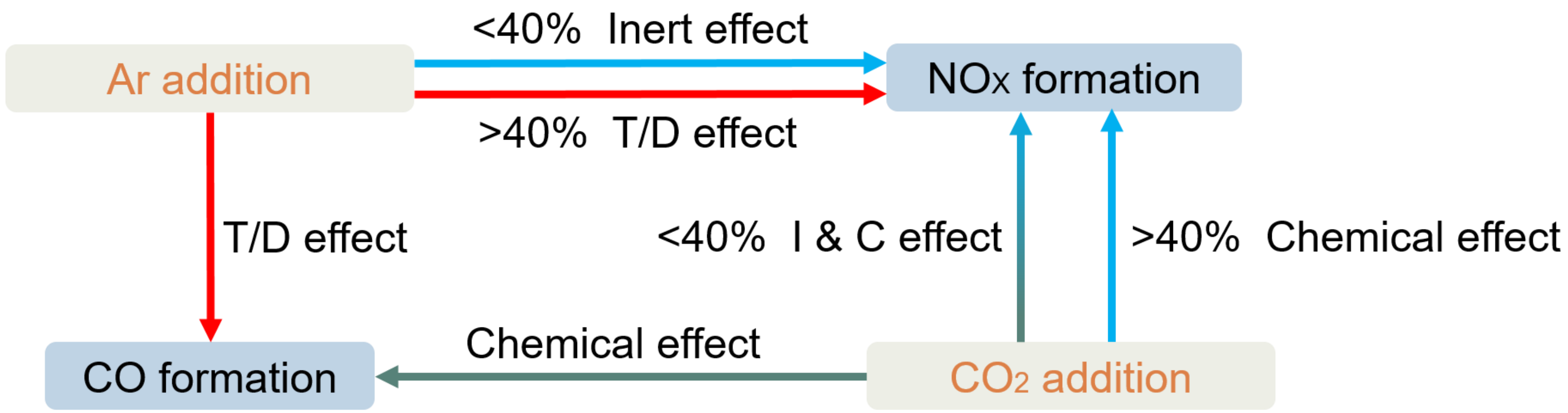
- It could effectively reduce the production of CO with the addition of He. In addition, the CO_2 addition enhance NO_x formation. It is may because CO and N_2O reacts again.

Conclusions

- **The effects of diluent addition on flame temperature**



- **The effects of diluent addition on pollutant emission**



Conclusions

- ***R182 : $N_2O + O = 2NO$***
  - ✓ The O radical decrement rate of CO2 addition is larger than Ar addition.
- ***R178: N + NO = N2 + O***
  - ✓ N radical concentration under Ar addition is higher than CO2 addition. It's the reason why the decrement rate of R178 production rate for CO2 is higher than Ar .
- ***R199: NH + NO = N2O + H***
  1. 20% ~ 40%
     - ✓ H radical concentration drops sharply.
  2. 40% ~ 60%
     - ✓ NH radical concentration decrease.

Acknowledgements

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Thank you for attention

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