Effect of dilution of fuel in CO_2 on the conversion of NH_3 to NO_x during oxy-fuel combustion

Key words: CO_2, Oxy-fuel combustion, NO, Fuel dilution

Cite this as: Kun-zan Qiu, Ye Yang, Zhuo You, Zhi-hua Wang, Zhi-jun Zhou, Jun-hu Zhou, Ke-fa Cen, 2015. Effect of dilution of fuel in CO_2 on the conversion of NH_3 to NO_x during oxy-fuel combustion. *Journal of Zhejiang University-SCIENCE A (Applied Physics & Engineering)*, 16(10):820-829. [doi:10.1631/jzus.A1300231]

Method

Table 1 Gas composition at the inlet of the reactor

Case name	CH_4 (v/v)%	NH_3 ppm	O_2 (v/v)%	CO_2 (v/v)%	Equivalence ratio (Φ)	Mechanism calculated
A1	5.88	1,176	19.74	74.26	0.6	GRI30 IRGRI
A2	9.84	1,969	18.89	71.07	1.05	
A3	13.48	2,697	18.11	68.14	1.5	
B1	0.59	118	1.97	97.43	0.6	GRI30 IRGRI
B2	0.98	197	1.89	97.11	1.05	
B3	1.35	270	1.81	96.81	1.5	
C1	0.06	12	0.2	99.74	0.6	M09 IRM09
C2	0.098	20	0.19	99.71	1.05	
C3	0.13	27	0.18	99.68	1.5	

· Apart from the equivalence ratio under high levels of CO_2, the formation of NO_x may be affected by the level of fuel in the mixture. But little is known about how variation in the level of fuel affects NO_x formation in an O_2/CO_2 atmosphere. The fuel level limits the speed of the branched chain reaction.

· Combustion of CO_2 diluted CH_4/NH_3 fuel with O_2/CO_2 as an oxidizer was simulated using a Chemkin Pro plug flow reactor.

· Variation in the chemical effects of CO_2 on NO_x formation from NH_3 due to changes in the fuel level in mixtures with high CO_2 concentrations was investigated via numerical simulation.

· The GRI-Mech mechanism 3.0 (GRI30), and the mechanism developed by Mendiara and Glarborg (2009), referred to as M09 in this paper. To study the effect of reaction $CO_2 + H \rightarrow CO + OH$ (1), the cases in Table 1 were also simulated with mechanisms without Reaction 1, namely the reverse reaction of CO+OH to form H+CO_2. These mechanisms are referred to as IRGRI and IRM09 in this article.

Main result

1. An unbranched chain reaction mechanism was proposed to illustrate the chemical effect of CO_2 on the radical pool and NO_x formation and reduction.

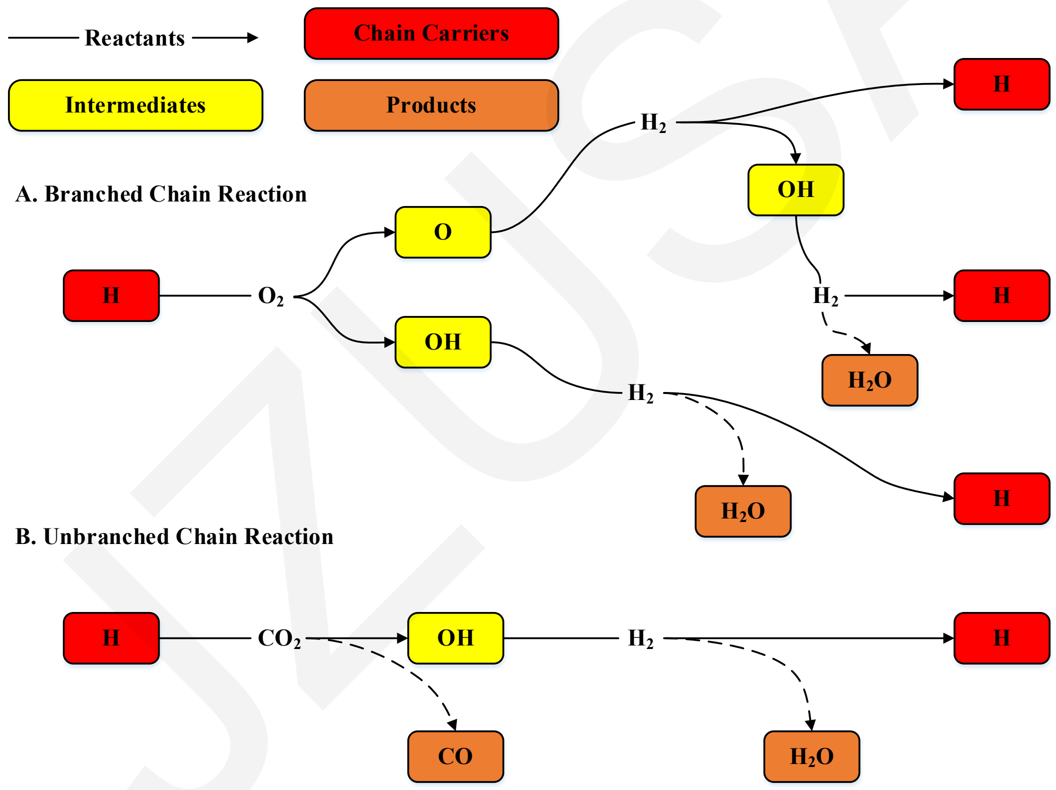

Fig. 1 The branched/unbranched chain reactions in oxy-fuel combustion

Main result

2. As the CO_2 concentration increases, the mutual promotion of the unbranched chain reaction and the branched reaction gradually changes to mutual competition for H, which gradually weakens the promotion of NO formation. Under conditions with CO_2 levels higher than 90%, however, the formation of NO is inhibited.

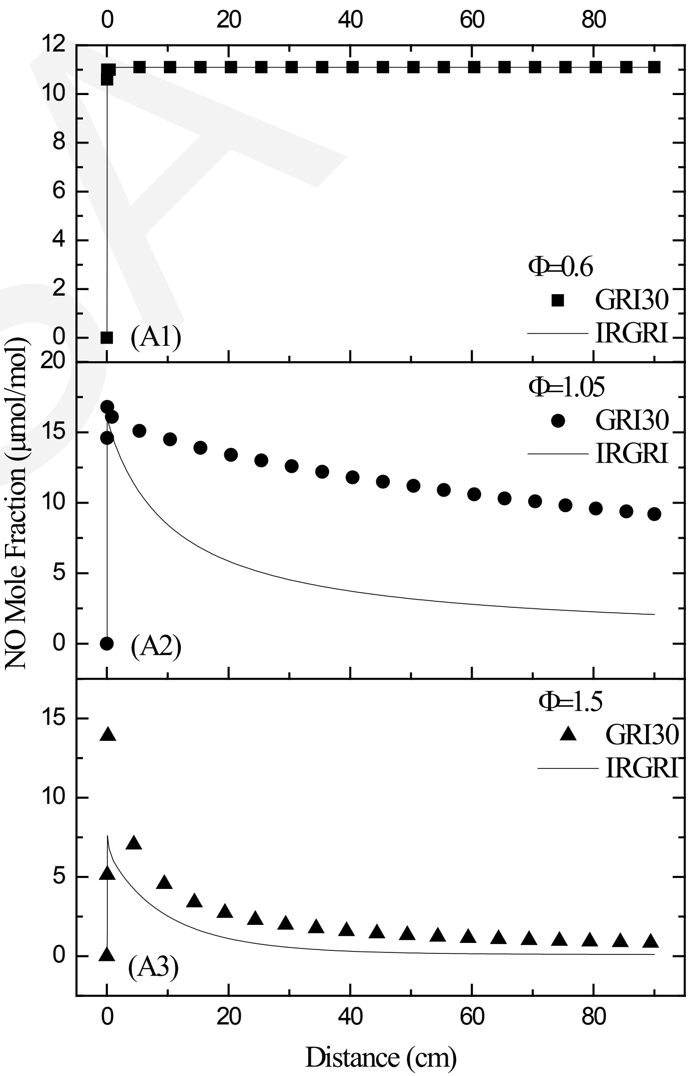

Fig. 2 Profile of NO along the reactor under conditions A1, A2 and A3 at T=1523 K. Closed symbols denote the GRI30 mechanism, while lines denote the IRGRI mechanism. A1, A2 and A3 correspond to conditions described in Table 1

Main result

· The promotion effect of Reaction 1 on NO during the formation period is weakened gradually with CO_2 dilution. NO formation is inhibited under high CO_2 dilution and fuel-lean conditions by Reaction 1 indirectly .

$$CO_2 + H \rightarrow CO + OH \qquad (1)$$

Table 2 The effect of Reaction 1 on formation and reduction of NO at T=1523 K

Equivalence ratio		A	B	C
0.6	Formation[a]	-0.3	-2.5	-21.9
	Reduction[b]	0.0	0.0	-0.1
1.05	Formation	0.2	-2.8	-23.1
	Reduction	-36.4	-3.6	-1.4
1.5	Formation	28.8	-1.1	-26.9
	Reduction	25.8	-43.0	-8.9

a. Comparison of the peak values of NO along the reactor between the full mechanisms (GRI30 or M09) and the artificial mechanisms (IRGRI or IRM09). A positive value means formation of NO is facilitated by Reaction 1. A negative value means formation of NO is inhibited by Reaction 1.

b. Comparison of Peak (NO)-End (NO) along the reactor between the full mechanisms (GRI30 or M09) and the artificial mechanisms (IRGRI or IRM09). A positive value means reduction of NO after the peak is facilitated by Reaction 1. A negative value means reduction of NO after the peak is inhibited by Reaction 1.

Main result

3. The competition for H as CO_2 concentration increases influences the conversion of NH_3, changing from facilitation to inhibition of NH_3 conversion under fuel-rich conditions, and from no influence to inhibiting conversion under near stoichiometric and fuel-lean conditions.

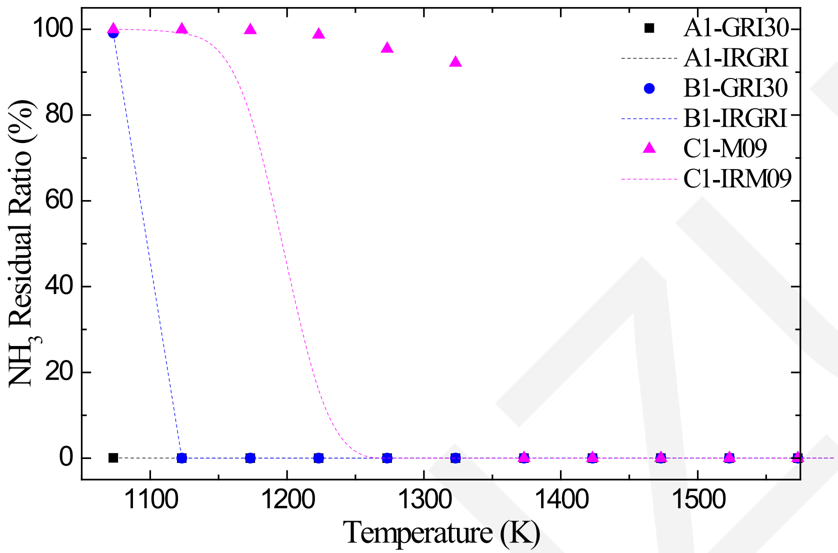

Fig. 3 The residual ratio of NH_3 at the end of the reactor under fuel-lean conditions, Φ=0.6. Symbols denote results simulated in GRI30 or M09, while dashed lines denote results from IRGRI or IRM09

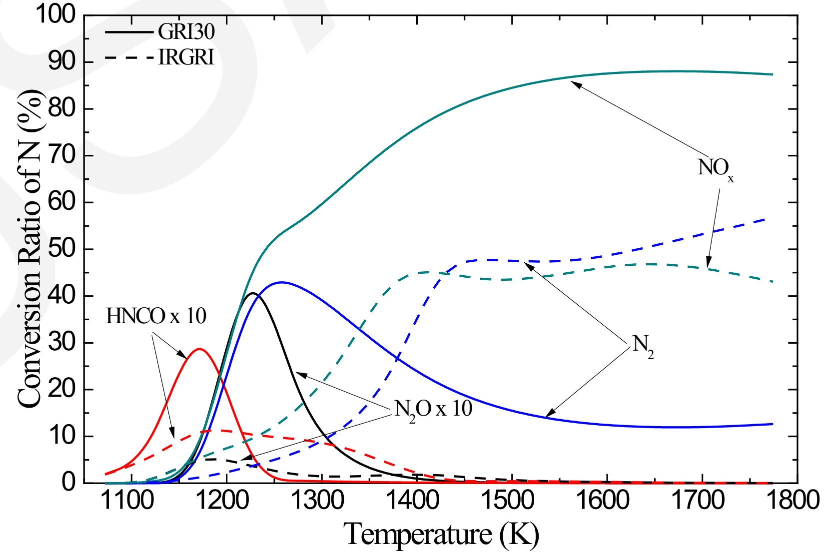

Fig. 4 Conversion ratio of nitrogen to main products, Φ=1.5, Condition B3. Solid lines denote the GRI30 mechanism, while dashed lines denote the IRGRI mechanism