Effects of Flue Gas Recirculation on NO and N_2O formations in Coal Combustion

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Nagoya University

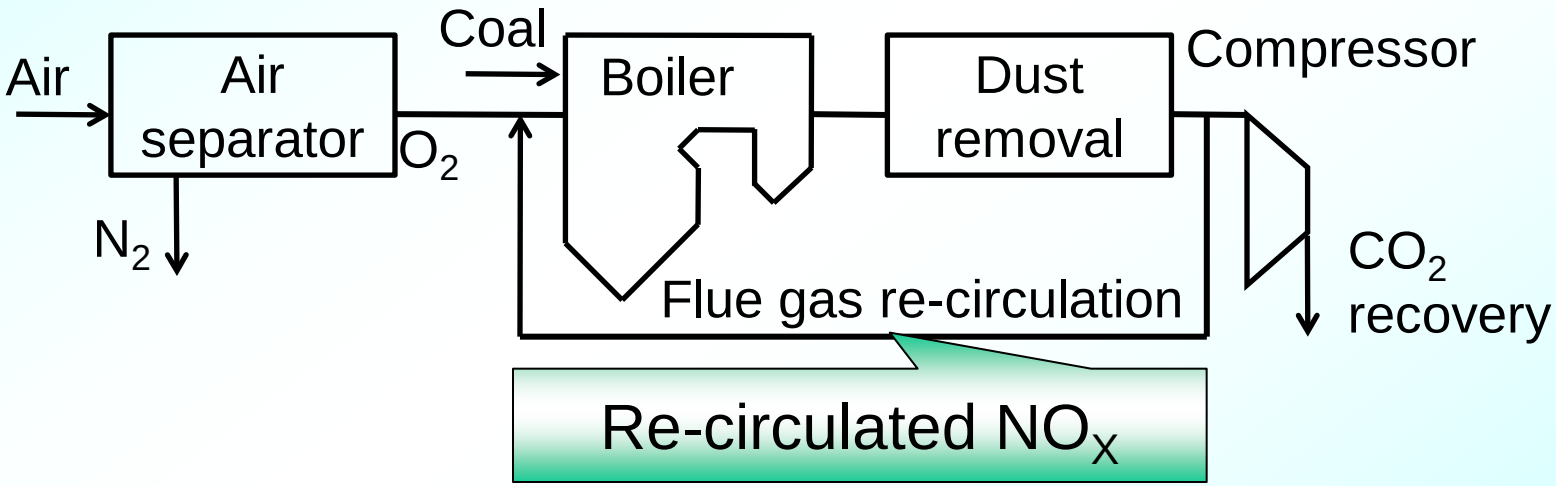
Background

Previous reports:

NO_X emissions

Oxy-fuel coal combustion < Air coal combustion

(the absence of N_2 in gas and lower flame temperature)



There have been few reports describing NO_X formation under oxy-fuel combustion conditions, including the quantitative effect of additional re-circulated NO and N_2O.

Objectives

Oxy-fuel (with flue gas recirculation) ⇔ Air

- **Combustion behaviors (1073K~1223K)**
- **NOx ($NO + N_2O$) emissions**

Contents

CO_2-O_2 atmosphere

Combustion experiment

Combustion behaviors

Flue gas re-circulation

(NOx addition to the inlet CO_2-O_2)

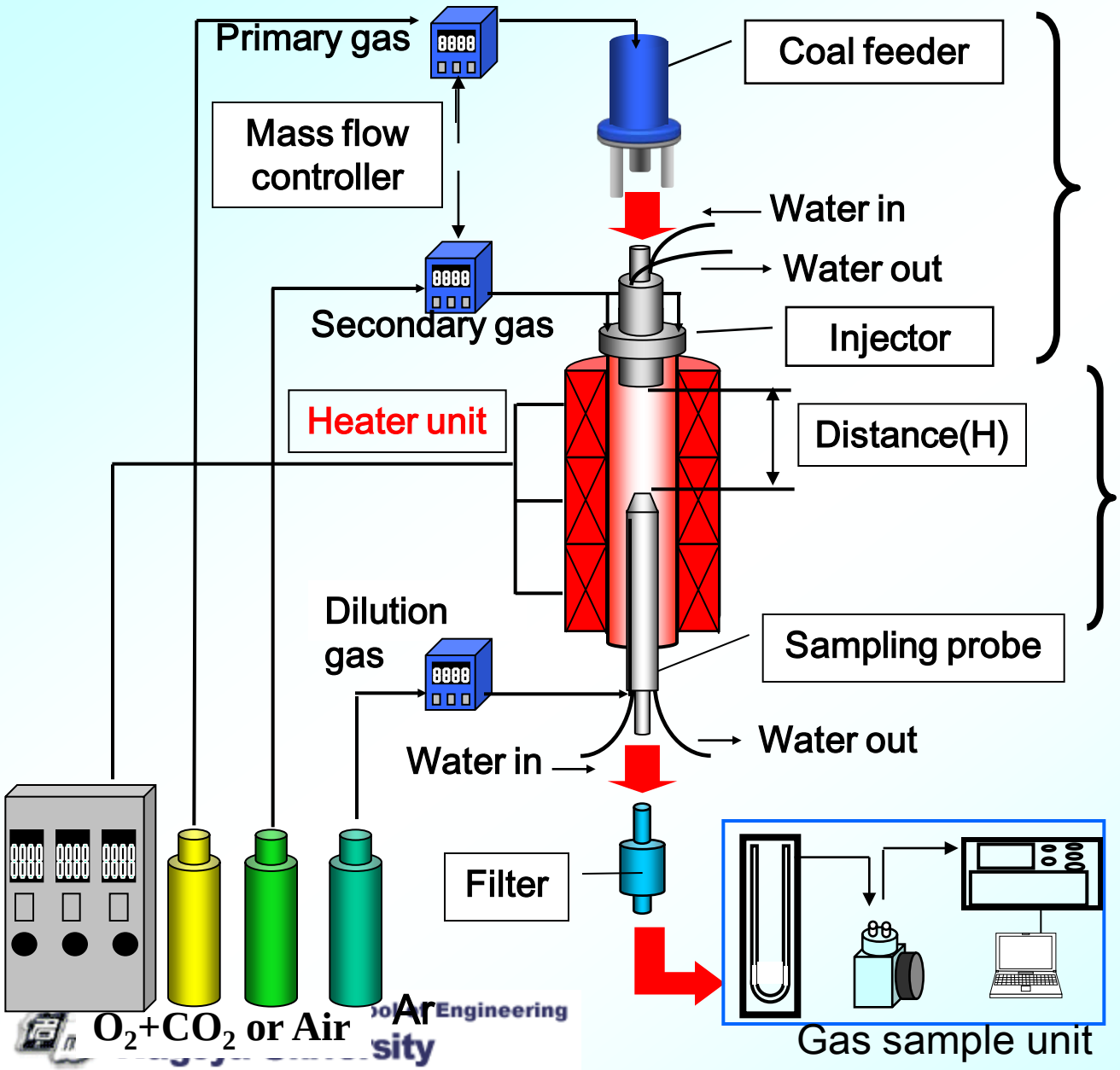
Simulated oxy-fuel

Combustion experiment

Elemental reaction analysis

NO_X emissions

Drop tube furnace for coal combustion experiment



Fuel injection part

Continuous feeding
Entraining gas : CO_2+O_2 or Air

Reaction part

Temperature controlled by electric heater
Residence time : 2s

Sampling part

Iso-kinetic sampling
Gas analysis
Particle analysis

Coal sample and experimental conditions

Coal properties

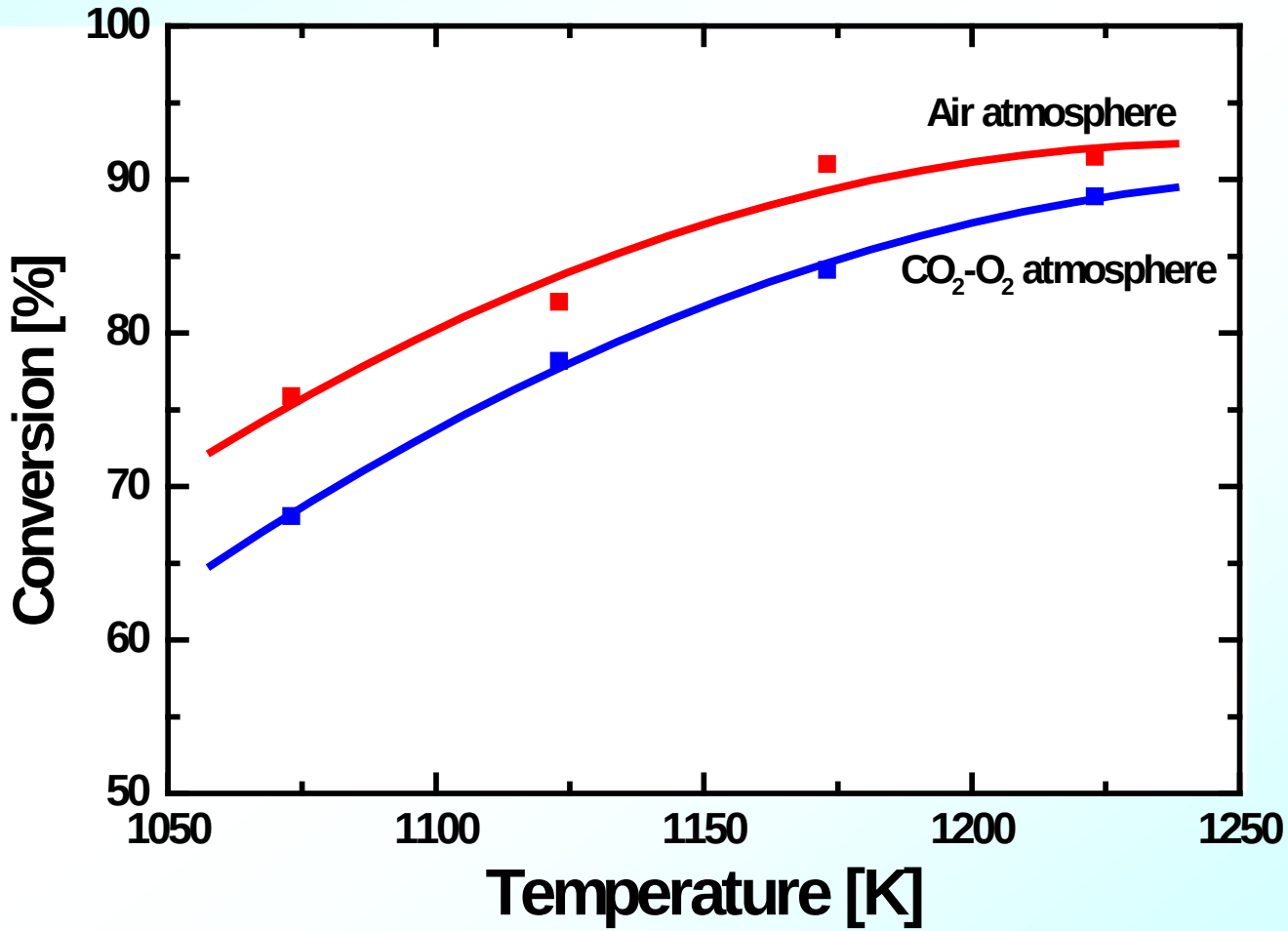
	Proximate analysis [wt.%dry]				Ultimate analysis [wt.%d.a.f]				
	Volatile matter	Fixed carbon	Ash	Moisture	C	H	N	O	S
Coal A	30.3	56	13.7	2	80.97	9.32	1.47	7.79	0.45

Experimental conditions

Atmosphere	CO_2+O_2	Air
CO_2 : O_2	79 : 21	
Primary gas [L/ min]	0.22 (O_2) 0.83 (CO_2)	1.05 (Air)
Secondary gas [L/ min]	0.81 (O_2) 3.04 (CO_2)	3.85 (Air)
Dilution gas (Ar) [L/ min]	1.23	
Dilution rate (Sample gas : Ar = 1 : 4)	5	
Stoichiometric air ratio	1.2	
Pressure [atm]	1	
Coal feed rate (g/ min)	0.5 ± 0.05	
Sampling distance from injecter [mm]	1300	
Furnance temperature [K]	1073, 1123, 1173, 1223	

Carbon conversions under air and CO_2-O_2 combustion

ash tracer analysis of particulates



Carbon conversions: CO_2-O_2 < air @all temperature

High concentrations of CO_2→Low reaction rate, low heat transfer

Combustion experiment under flue gas re-circulation condition

In oxy-fuel system,
minor components in flue gas was re-circulated to the combustion zone.

Focusing on NO and N2O

Procedures to make up re-circulated gas with additional NO and N_2O

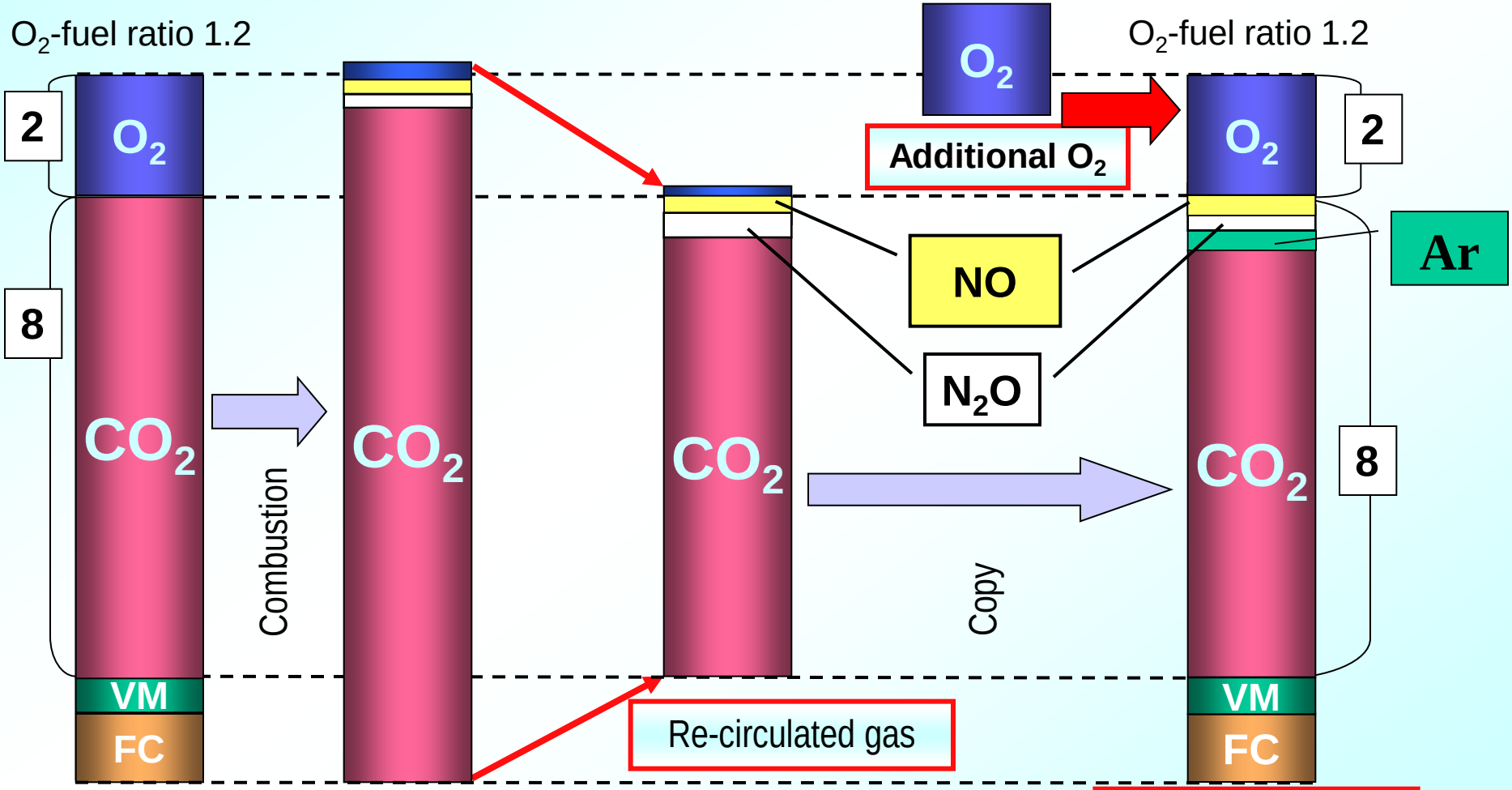
Gas	O_2	NO	N_2O	CO_2
Vol%	21	αx_{i-1}	αy_{i-1}	Balance

Initial gas composition at the inlet in i^{th} recycle experiment

i : recycle times (i=0 to 5)
x_i : NO concentration in flue gas in i^{th} recycle experiment (x_0=0)
y_i : N_2O concentration in flue gas in i^{th} recycle experiment (y_0=0)
α : dilution ratio defined by additional O_2 to the flue gas

1st step to simulate flue gas re-circulation condition

Inlet gas approaches to oxy-fuel with increase in gas recycling times



CO_2-O_2 combustion experiment (i=0)

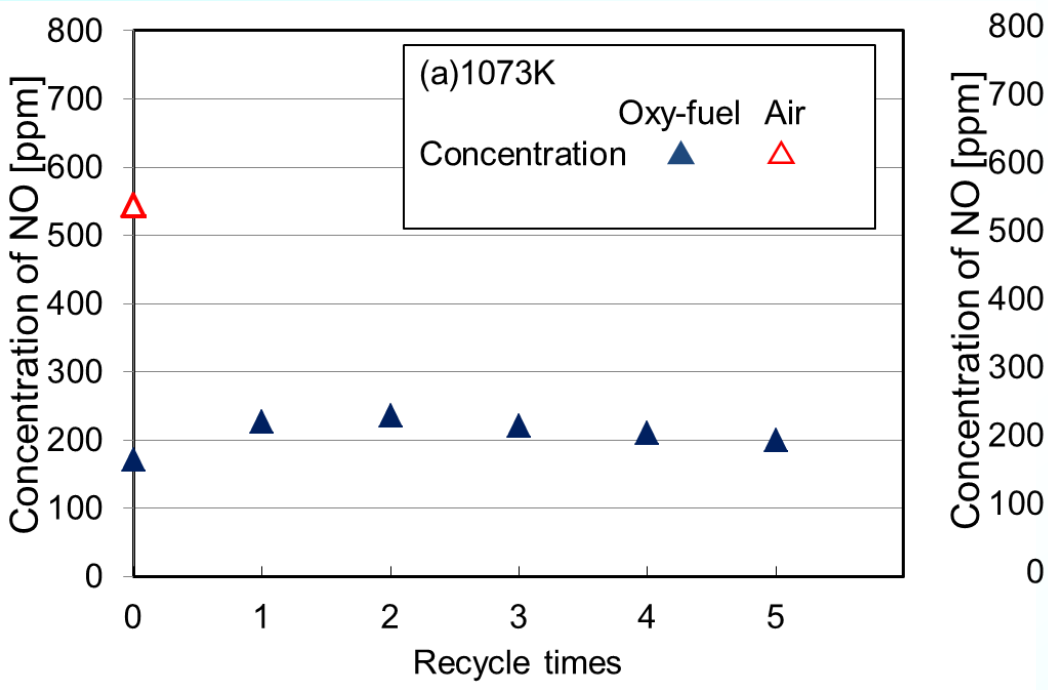
Flue gas

VM = Volatile matter
FC = Fixed Carbon

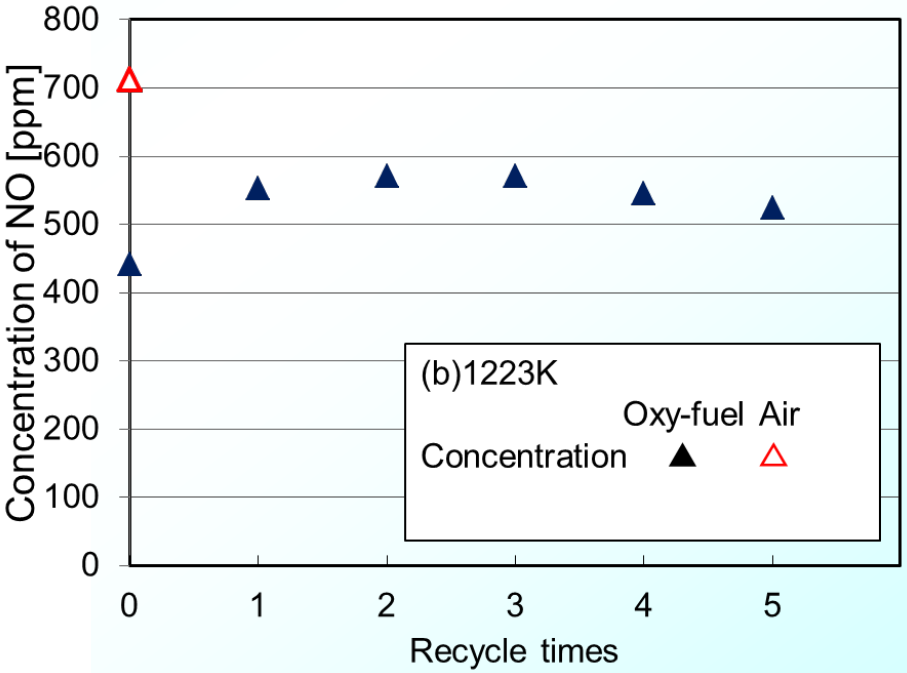
1st re-circulation experiment (i=1)

NO concentrations at the reactor outlet with gas recycle times

1073K NO



1223K NO



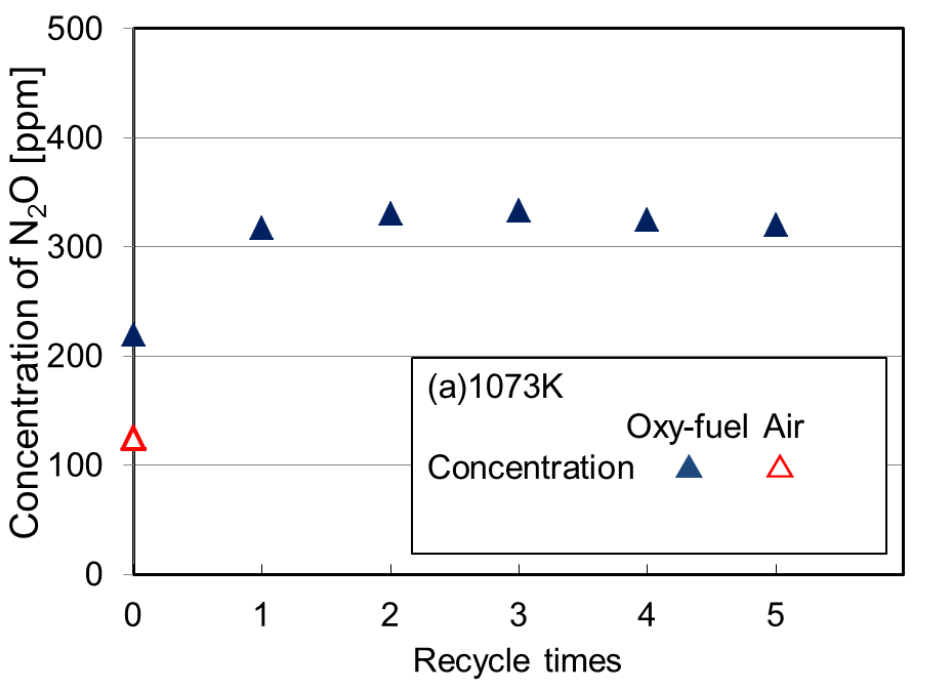
NO in flue gas: not accumulate

NO under oxy-fuel combustion < NO under air combustion

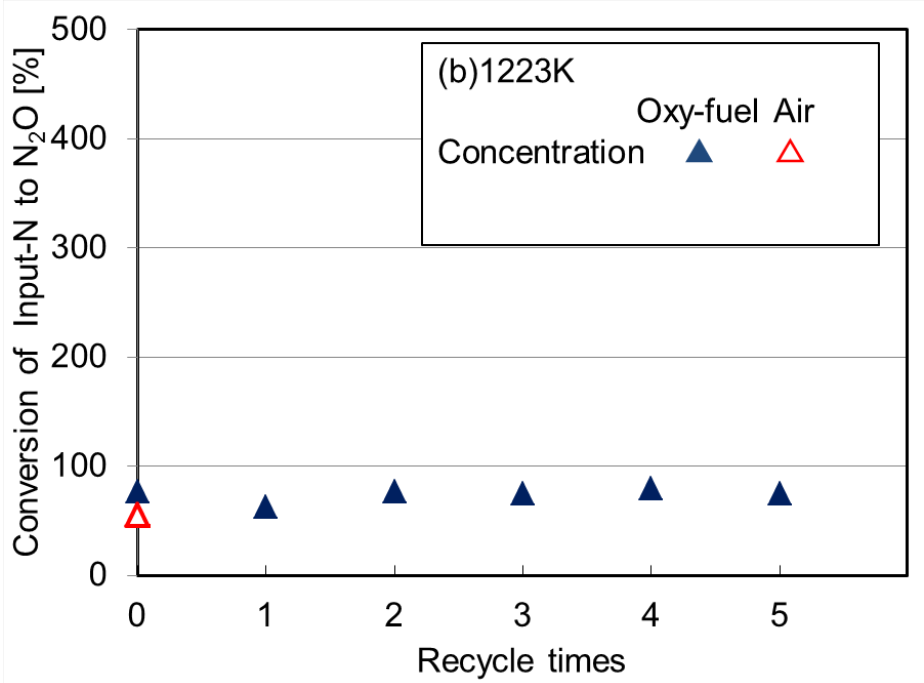
Higher temperature ➔ Smaller difference

N_2O concentration at the reactor outlet with gas recycle times

1073K N_2O



1223K N_2O



N_2O in flue gas: accumulate due to additional N_2O

N_2O under oxy-fuel combustion > N_2O under air combustion

Higher temperature ➔ Smaller difference

Elemental reaction analysis

Based on
- 52 chemical species, 323 elemental reactions (GRI-mech 3.0)
- 4 surface reactions (BRAIN-C, NEDO, 1989)

Pressure and temperature: constant through reaction time (3s)

Assumption for compositions of volatiles from coal

NH_3 , HCN , H_2 , C_2H_4 , CO

N → NH_3 : HCN = 1 : 1
Rest of H → H_2 : C_2H_4 = 1 : 1
Balance C → CO

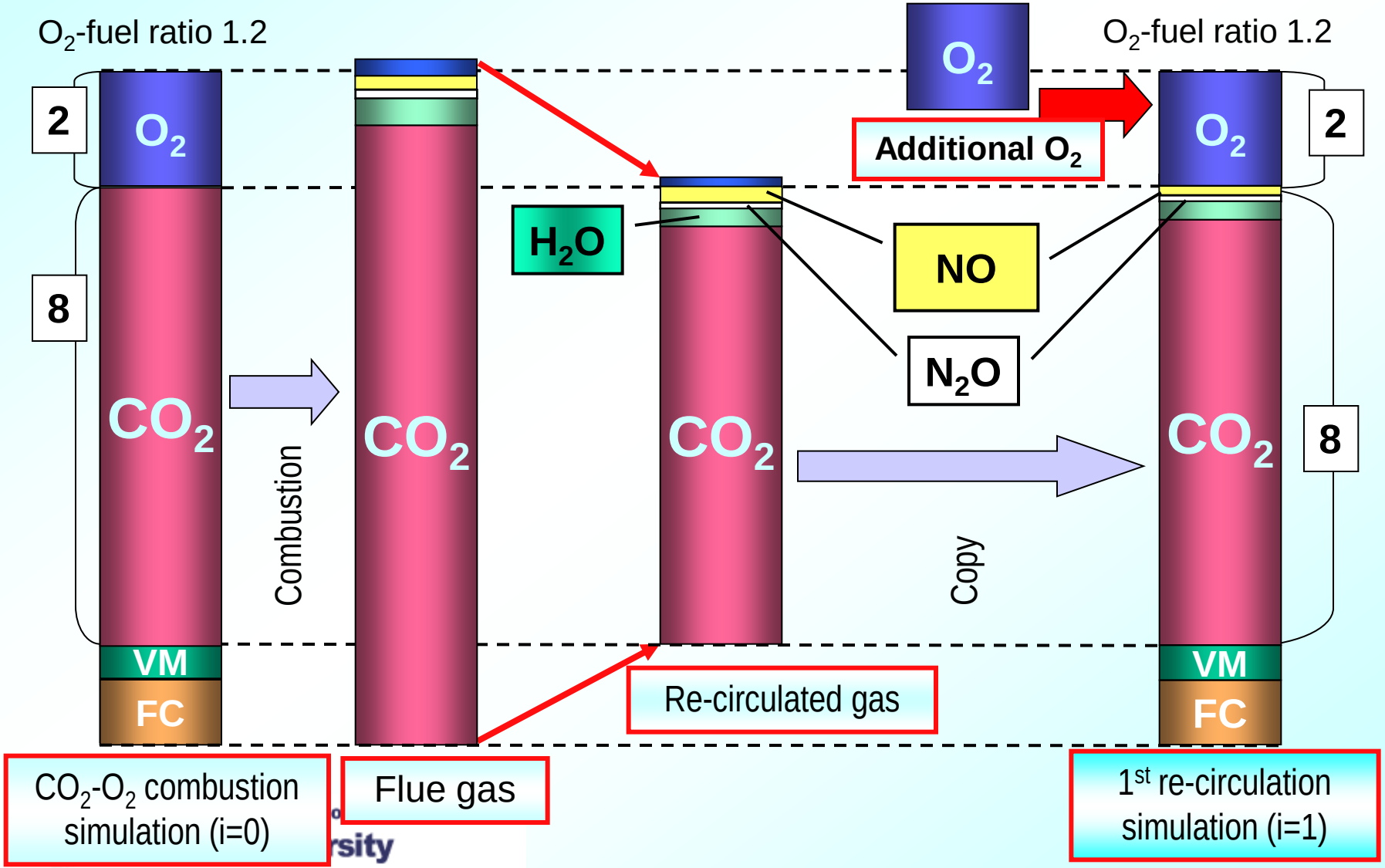
O_2 fuel ratio → 1.2
CO_2 : O_2 = 8 : 2

VM

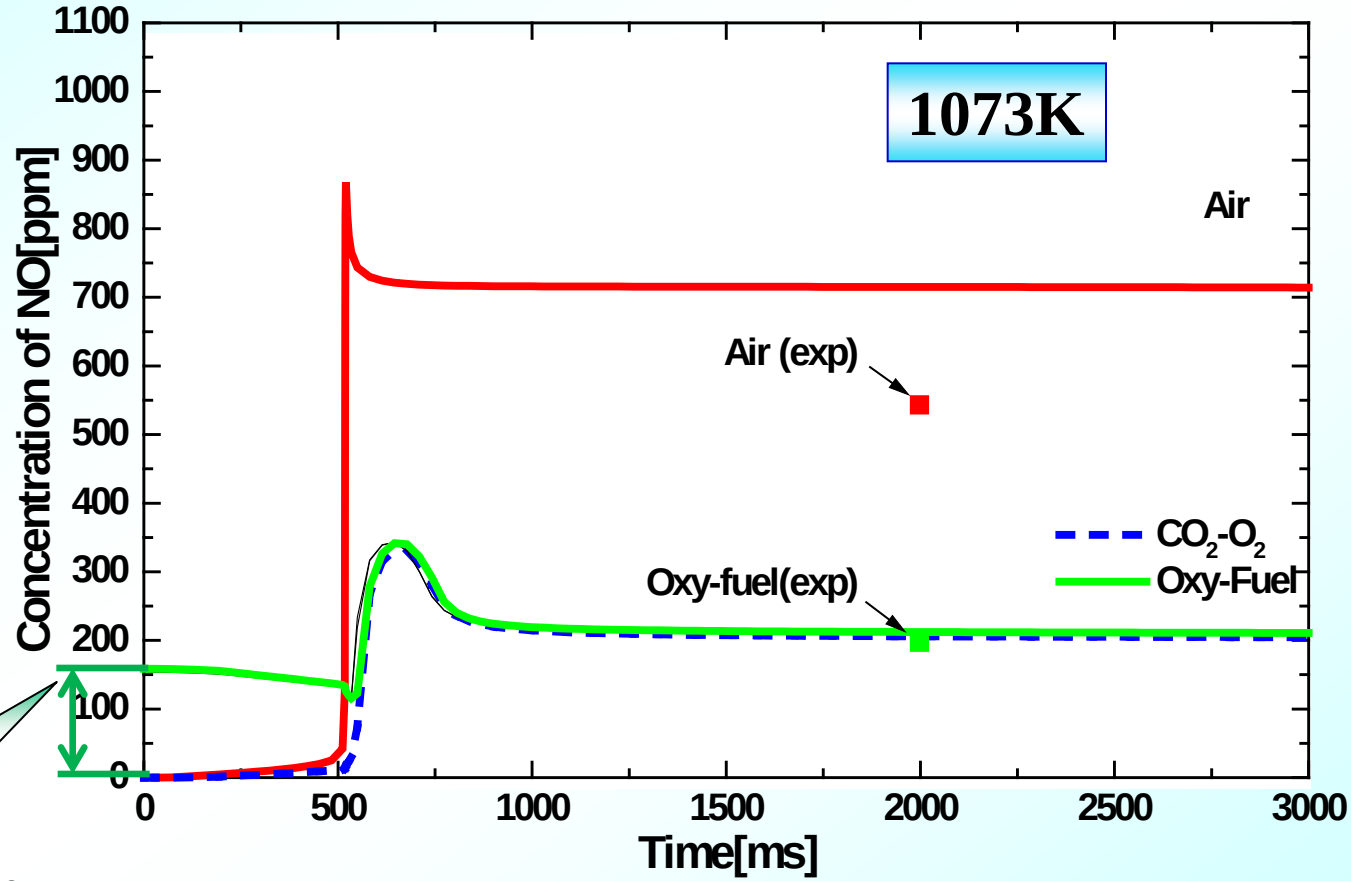
	[mol.%]
NH_3	0.00040
HCN	0.00040
H_2	0.01147
C_2H_4	0.01147
CO	0.02763
O_2	0.17089
CO_2	0.68356
FC	0.09419

Procedures to make up re-circulated gas (Sim.)

Output under oxy-fuel combustion can be derived as a convergence



NO concentration with reaction time under air, CO_2-O_2 and oxy-fuel combustion conditions



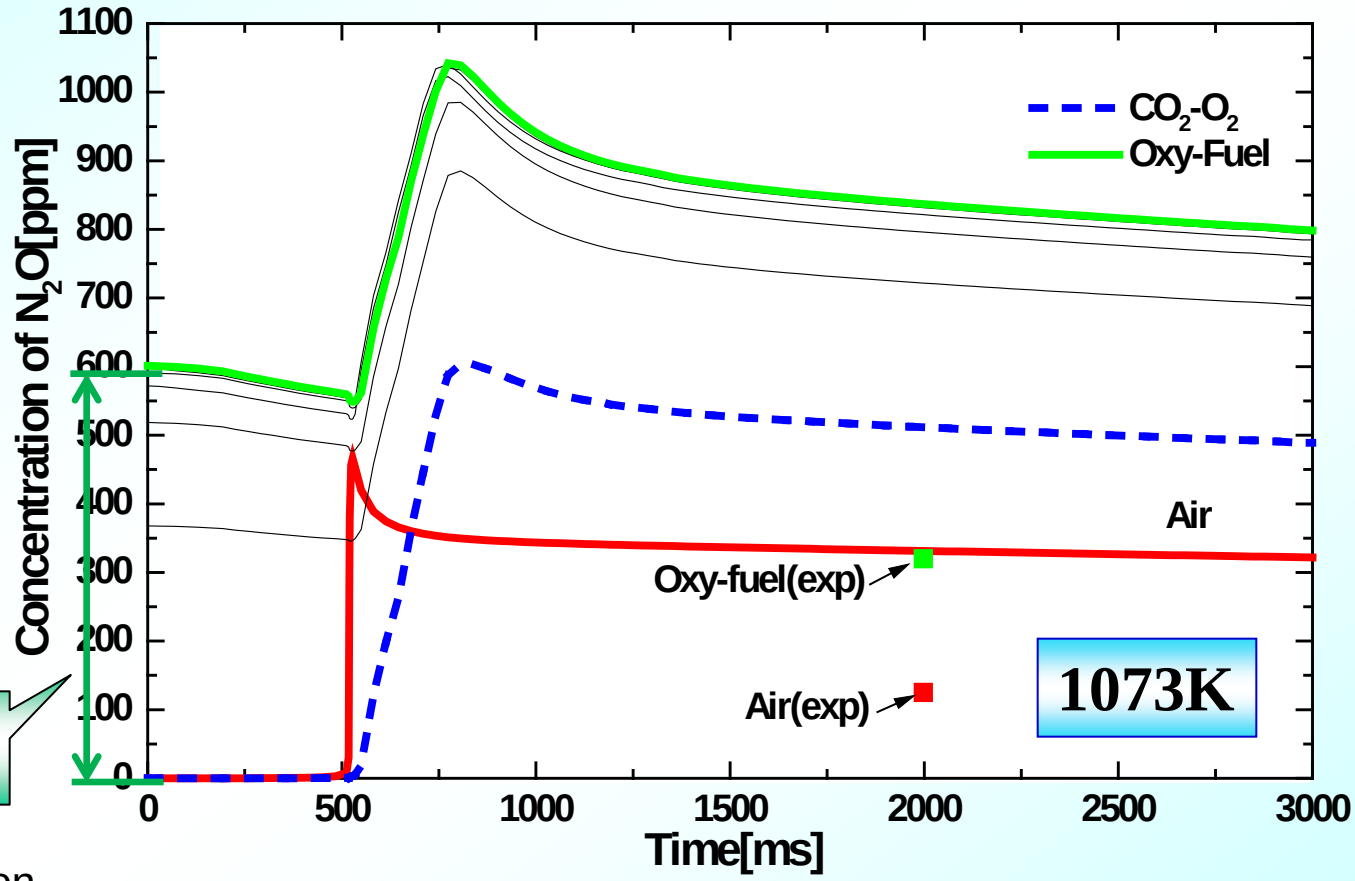
Additional NO

Oxy-fuel combustion

NO accumulation doesn't happen in spite of additional NO

Consistent with exp.

N_2O concentration with reaction time under air, CO_2-O_2 and oxy-fuel combustion conditions



Additional N_2O

Oxy-fuel combustion

N_2O accumulation happens due to additional NO

Consistent with exp.

Summary

1. Carbon conversion under CO_2-O_2 combustion was lower than that under air combustion.

2. NO concentration from coal combustion in CO_2-O_2 atmosphere was lower than that in air, and it did not increase in spite of additional NO input under oxy-fuel condition at 1073K. However, NO emissions accumulated under oxy-fuel conditions at 1223K.

3. N_2O concentration from coal combustion in CO_2-O_2 atmosphere was higher than that in air, and it accumulated under oxy-fuel condition at 1073K. The accumulation of recycled N_2O and the differences between the outcomes of oxy-fuel and air combustion had practically disappeared at 1223K.