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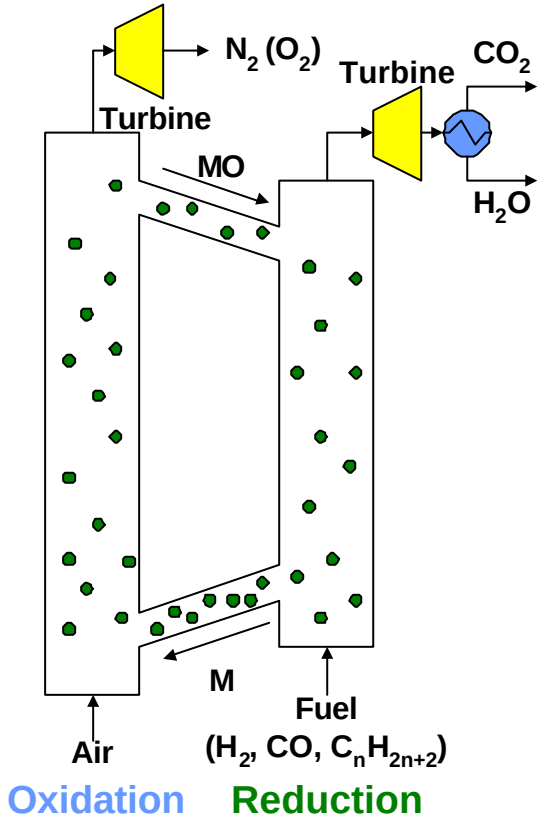
# Reaction Kinetics of Oxygen Carrier Particles in Chemical-Looping Combustor

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# Introduction

## CLC : Chemical-Looping Combustor




- **Oxidation Reaction :**

  **exothermic, 800-1300°C**

$$M + \frac{1}{2}O_2 \rightarrow MO$$

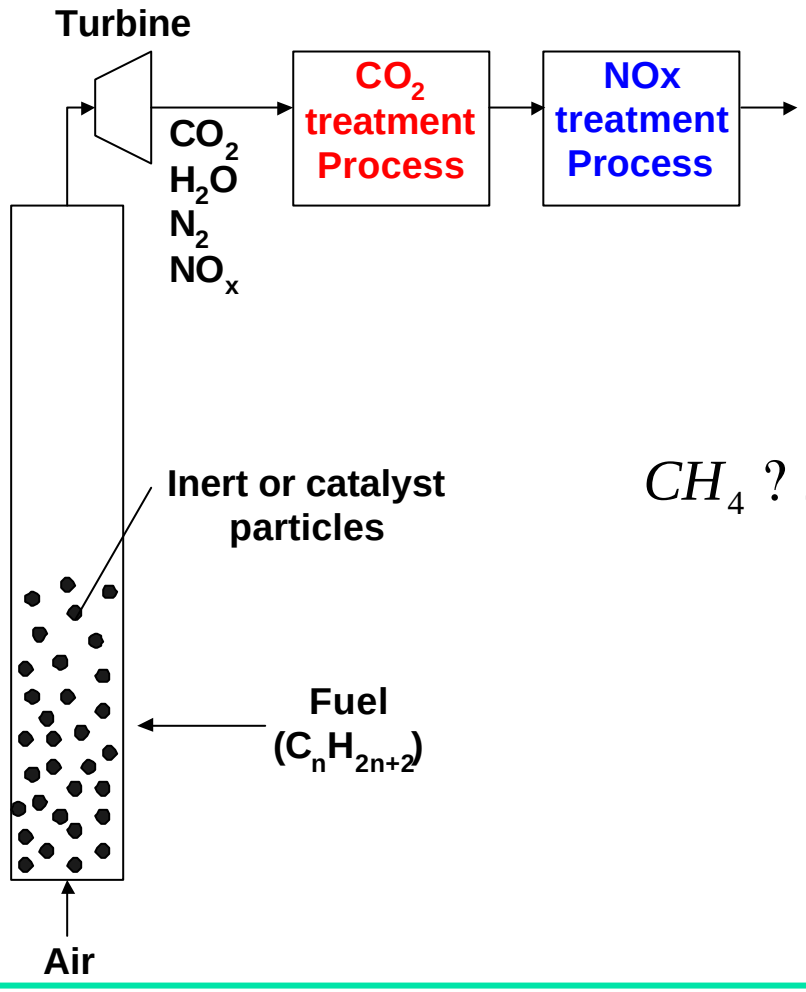
- **Reduction Reaction :**

  **endothermic, 400-800°C**

$$CH_4 + 4MO \rightarrow CO_2 + 2H_2O + 4M$$

**M : metal, MO : metal oxide**

# Traditional LNG combustor




$$CH_4 ? 2O_2 ? \quad CO_2 ? 2H_2O$$

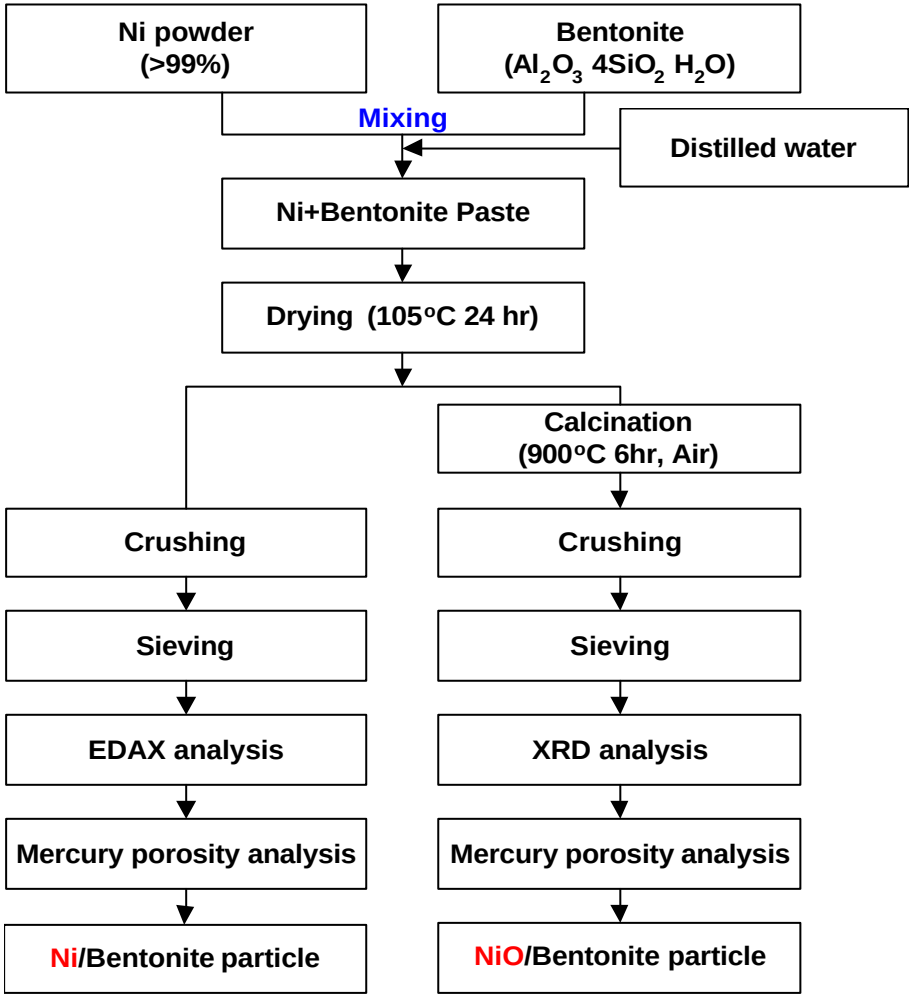
# Advantages of CLC

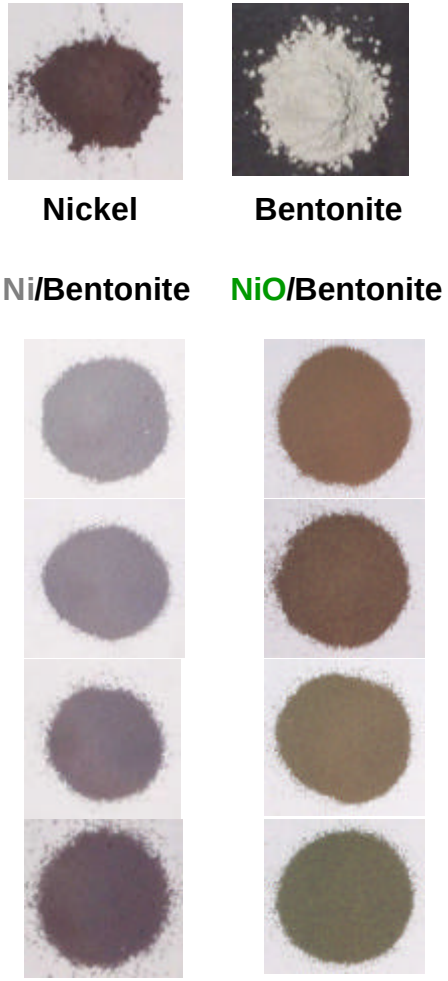
- LNG 를 연료로 사용하는 기존 가스연소기에 비해 열효율이 60%이상 높고(LHV 기준), $CO_2$ 발생량이 적다(0.33kg-$CO_2$/kWh)
- 산화반응기에서 일어나는 산소 carrier 입자의 산화반응은 화염(flame)이 없는 상태에서 일어나므로 thermal NOx 발생이 없다(대기오염 방지).
- 배가스 재순환 연소기($O_2$/$CO_2$ combustor)와는 달리 순수산소가 아닌 공기를 사용할 수 있으므로 산소정제비용이 절감된다.
- 환원반응기에서 배출되는 기체는 $CO_2$ 와 $H_2O$ 뿐이므로 응축에 의해 쉽게 고농도의 $CO_2$를 분리할 수 있다.
  (온실가스 저감, $CO_2$ 농축 비용 절감)

# Purpose of this study

- **Chemical-looping combustor**의 적용 가능성 검토
- **Chemical-looping combustor**에 사용되는 산소 **carrier** 입자의 반응성 비교 및 최적 제법**(recipe)** 결정
- 산소 **carrier** 입자의 산화**-**환원 반응 특성 측정 및 해석
  **(**실제 반응기 설계**,** 운전조건 설정**, scale-up** 기초자료로 활용**)**

# Preparation of oxygen carrier particles



# Particles characteristics

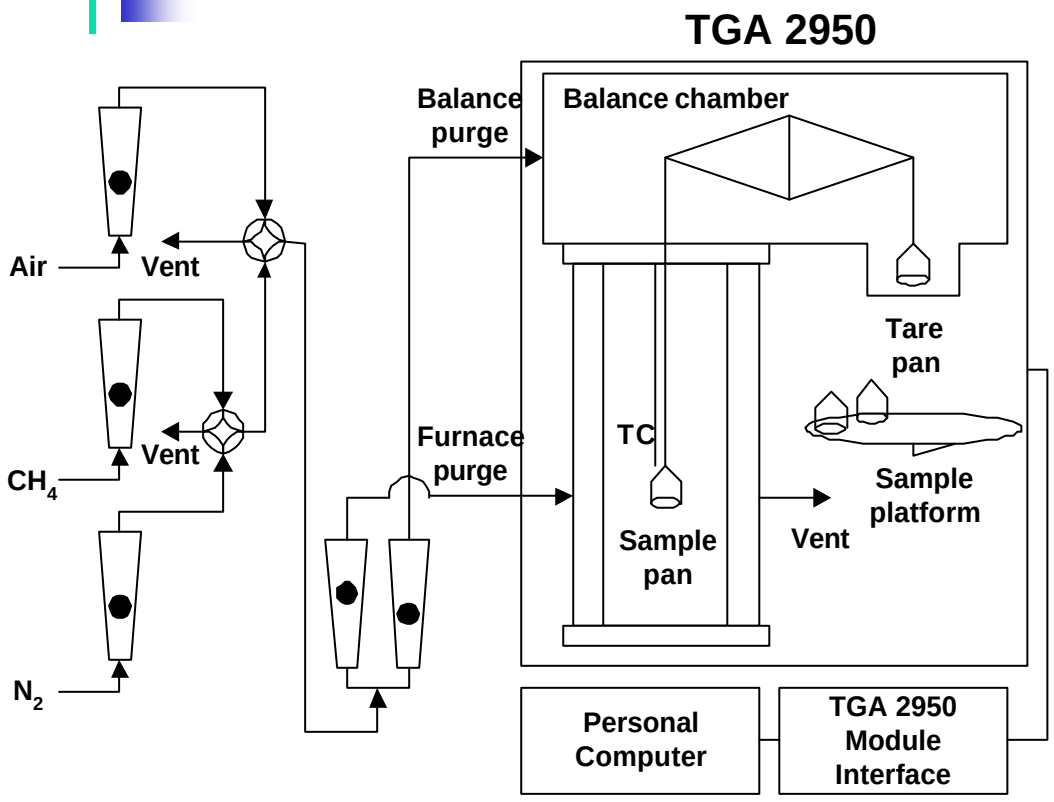
- **Ni / Bentonite particle**

| Name | Nickel wt. % | Porosity [%] | Particle density [g/cm$^3$] | Bulk density [g/cm$^3$] |
|---|---|---|---|---|
| *Particle B* | 21.64 | 68.1 | 2.6545 | 0.8469 |
| *Particle C* | 37.92 | 68.2 | 2.9623 | 0.9406 |
| *Particle D* | 51.86 | 77.5 | 5.1969 | 1.1704 |
| *Particle E* | 74.00 | 79.5 | 7.3739 | 1.5149 |

- **NiO / Bentonite particle**

| Name | NiO wt. % | Porosity [%] | Particle density [g/cm$^3$] | Bulk density [g/cm$^3$] |
|---|---|---|---|---|
| *Particle B* | 26.01 | 71.37 | 3.5619 | 1.0199 |
| *Particle C* | 43.74 | 71.25 | 3.6288 | 1.0433 |
| *Particle D* | 57.82 | 64.54 | 3.5892 | 1.2729 |
| *Particle E* | 78.36 | 58.99 | 3.7309 | 1.5302 |

# Experimental apparatus




**TGA 2950, TA Instrument**

**Temperature range : 25-1000 ºC**

**Heating rate range : 0.1-100ºC/min**

**Gas flow rate : 100 ml/min <**

**Weight range :**

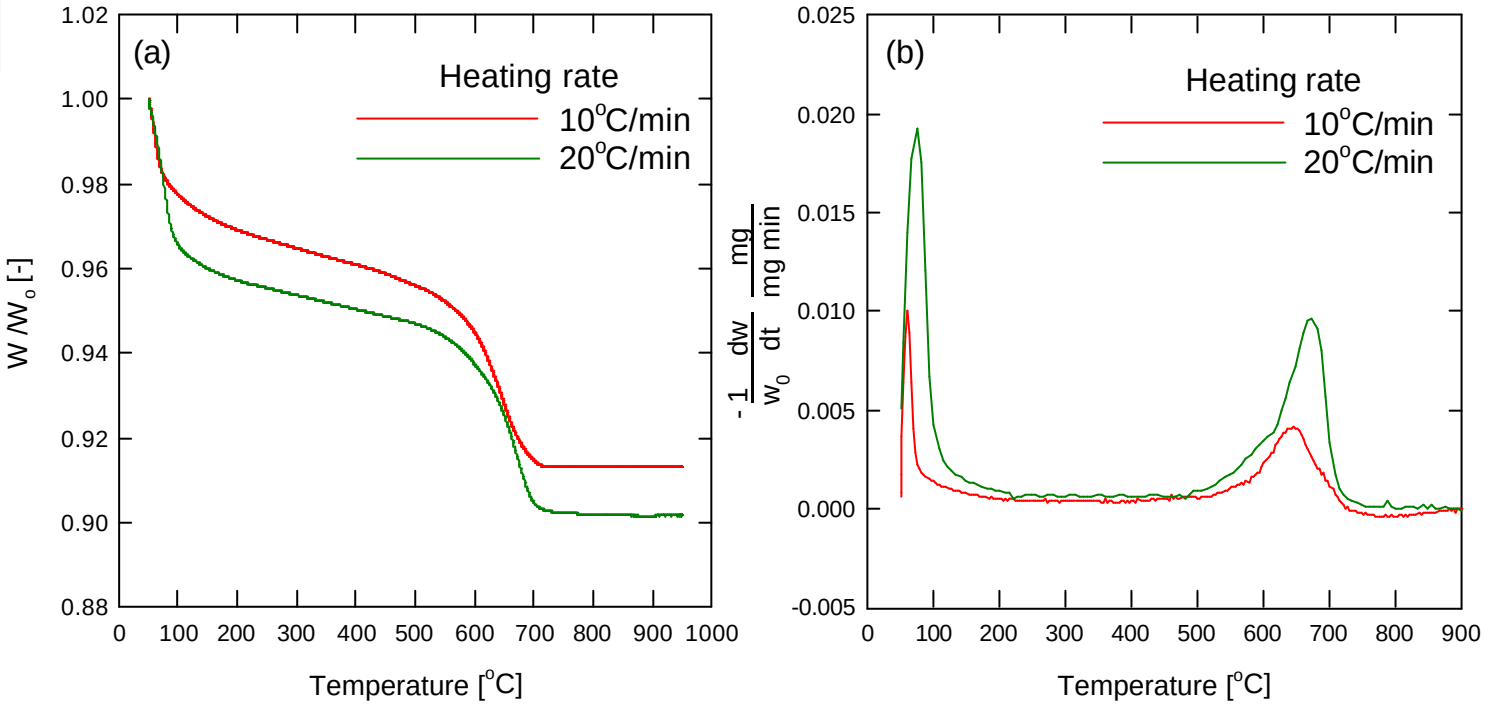
**0.1?g-100mg or 1?g-1000mg**



# Experimental method and variables

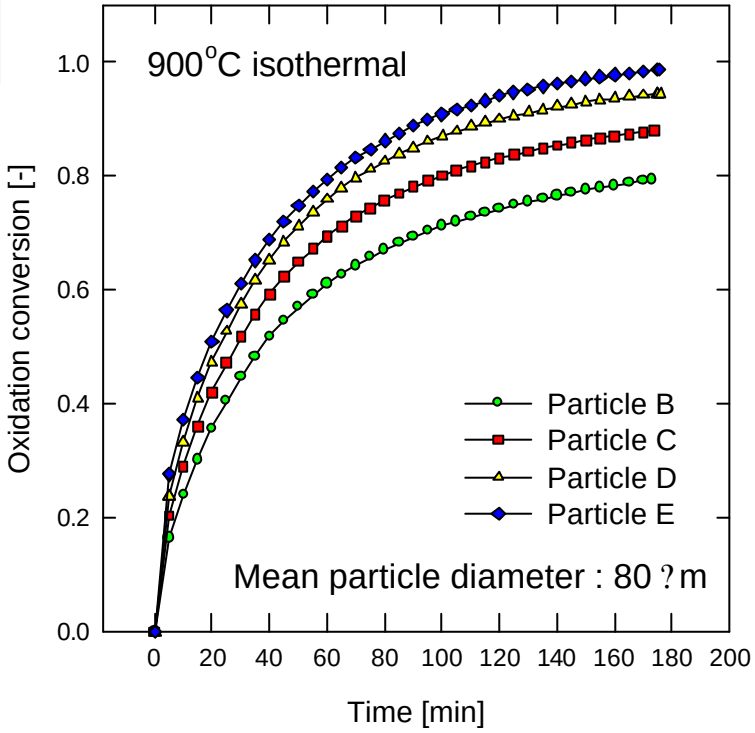
| *Reaction* | *Dehydration of bentonite* | *Oxidation* | *Reduction* |
|---|---|---|---|
| Particles | Pure Bentonite | Ni / Bentonite | NiO / Bentonite |
| Method | Non-isothermal | Isothermal | Isothermal |
| Inert gas | | $N_2$ | $N_2$ |
| Reacting gas | Air (100 m*l*/min) | Air (100 m*l*/min) | $CH_4$ (5.04%, 100m*l*/min) |
| Heating rate [°C/min] | 10, 20 (Non-isothermal) | | |
| Variables | | | |
| Ni or NiO content | | Particle B, C, D, E | Particle B, C, D, E |
| Temperature [°C] | | 850, 900, 950, 1000 (Isothermal) | 650, 700, 750, 800, 850, 900 (Isothermal) |
| Particle size [? m] | | 80, 128, 181, 284 | 80, 128, 181, 284 |

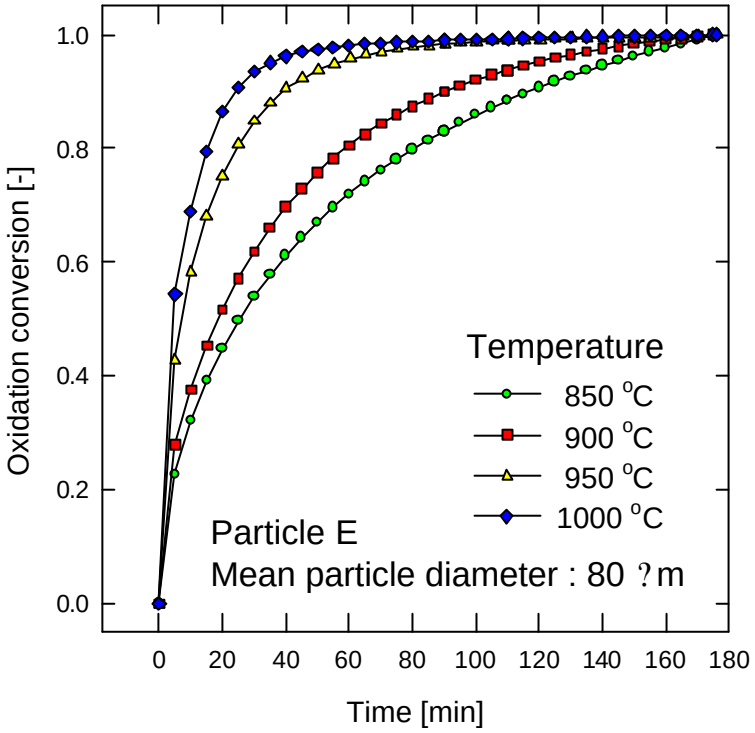
# Results : Dehydration of bentonite




- 60-75°C : 입자 제조시 첨가된 수분 제거됨
- 650-675°C : bentonite($Al_2O_34SiO_2H_2O$) 중에 포함된 결정수가 제거됨
- Ni/Bentonite 입자의 TGA 산화실험시 결정수 제거후(700°C 이상) 실험

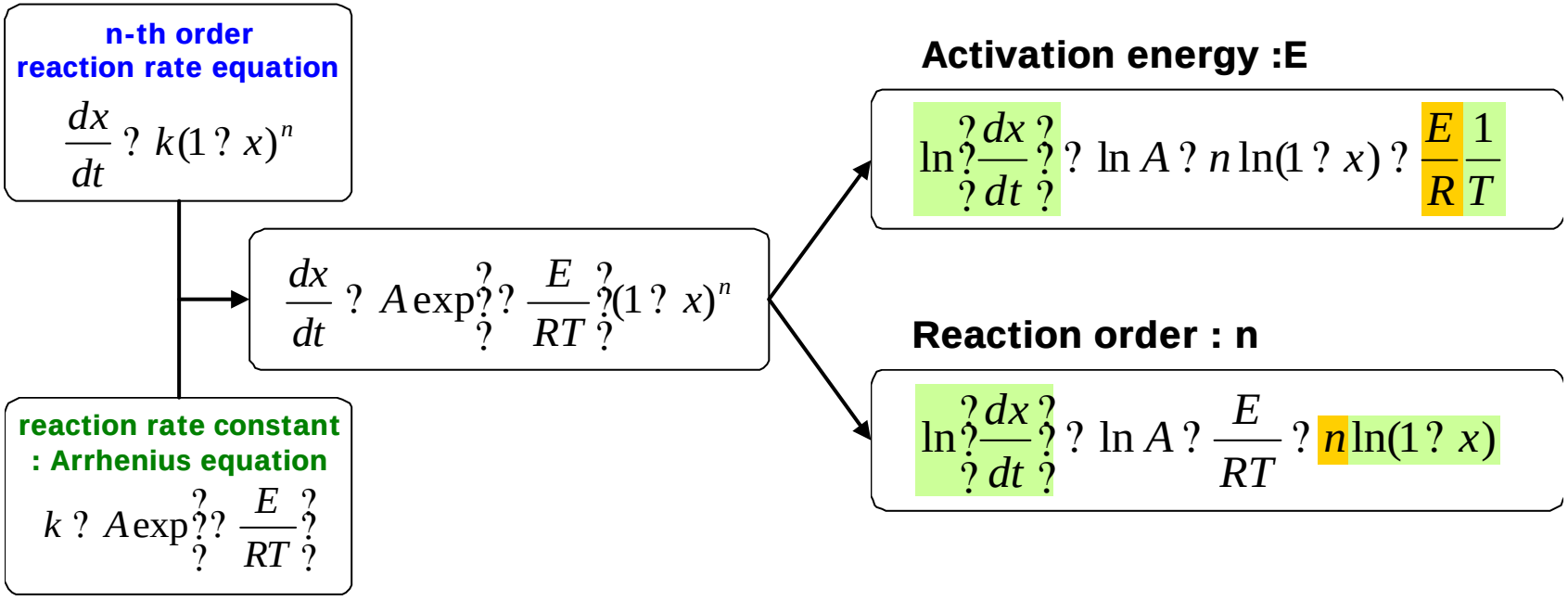
# Results : Oxidation reaction



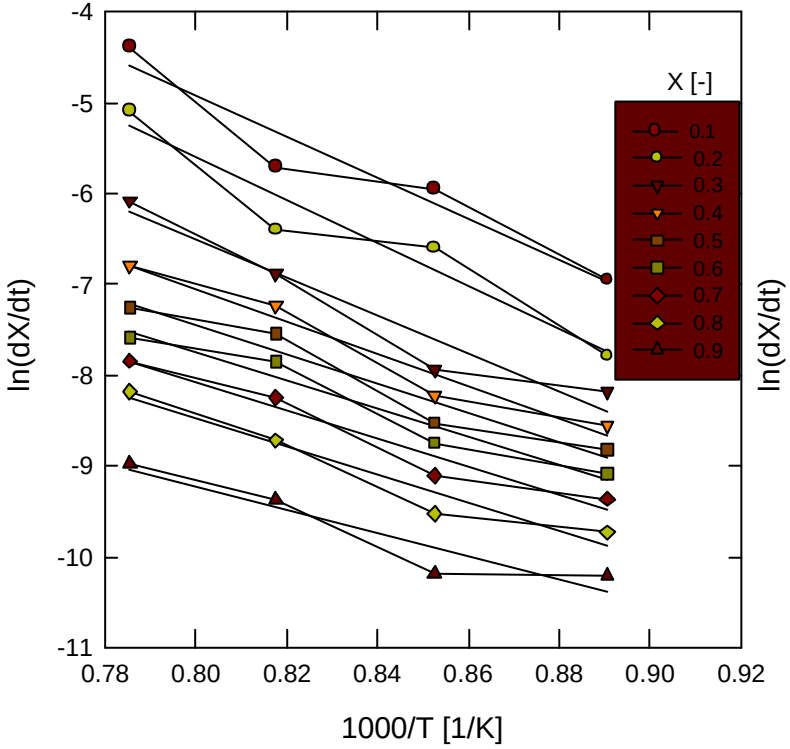



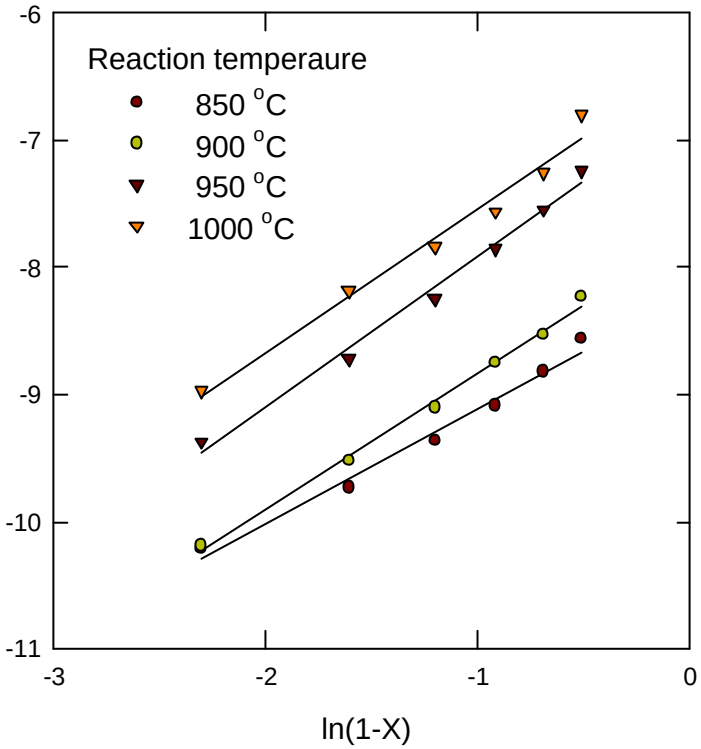

- **Reaction rate :**
  - **Ni 함량이 증가함에 따라 반응속도 증가 : Particle E > D > C > B**
  - **반응온도가 증가함에 따라 반응속도 증가 : 1000 > 950 > 900 > 850 ºC**
  - **순수 Ni powder의 경우 소결되어 괴상으로 변화, 유동화 물질로 부적합**

# Results : Reaction kinetics



# Results : Oxidation reaction kinetics

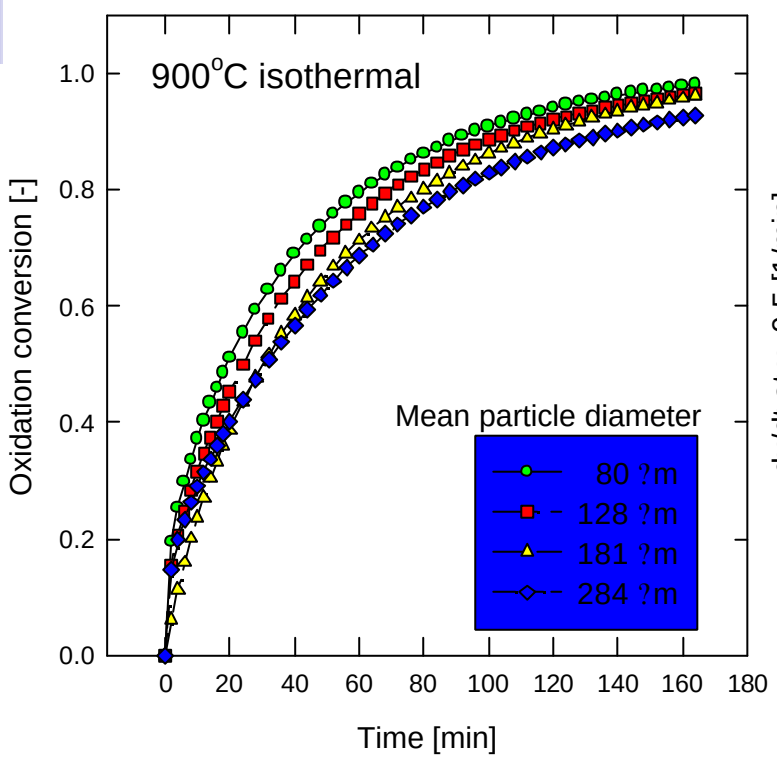


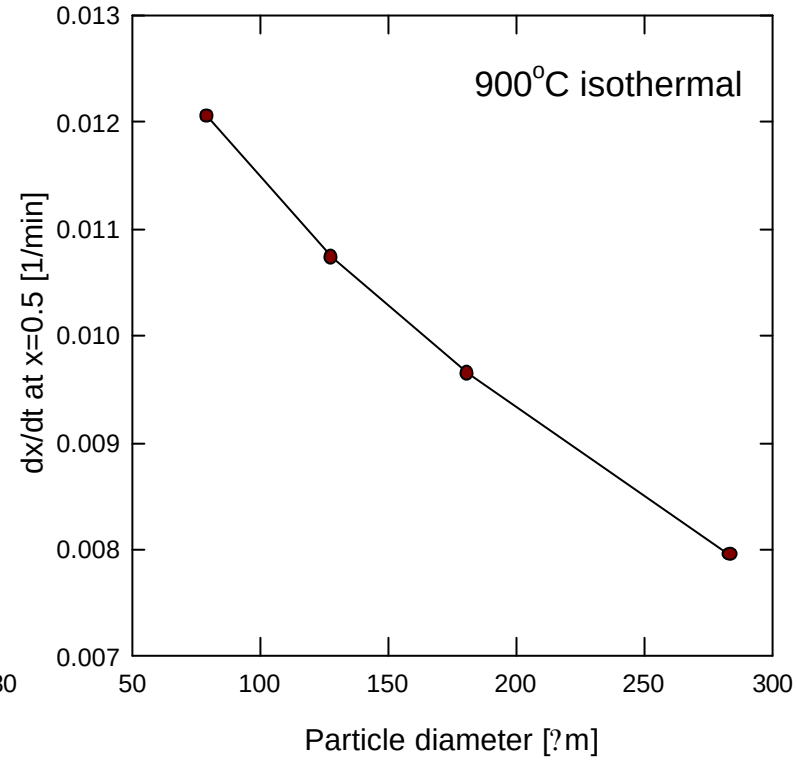




- **ln(dx/dt) vs. 1/T**
  - **Slope = -E/R**
  - **E = 35.3 Kcal/mol**

- **ln(dx/dt) vs. ln(1-x)**
  - **Slope = n**
  - **n=1.0748 (first-order reaction)**

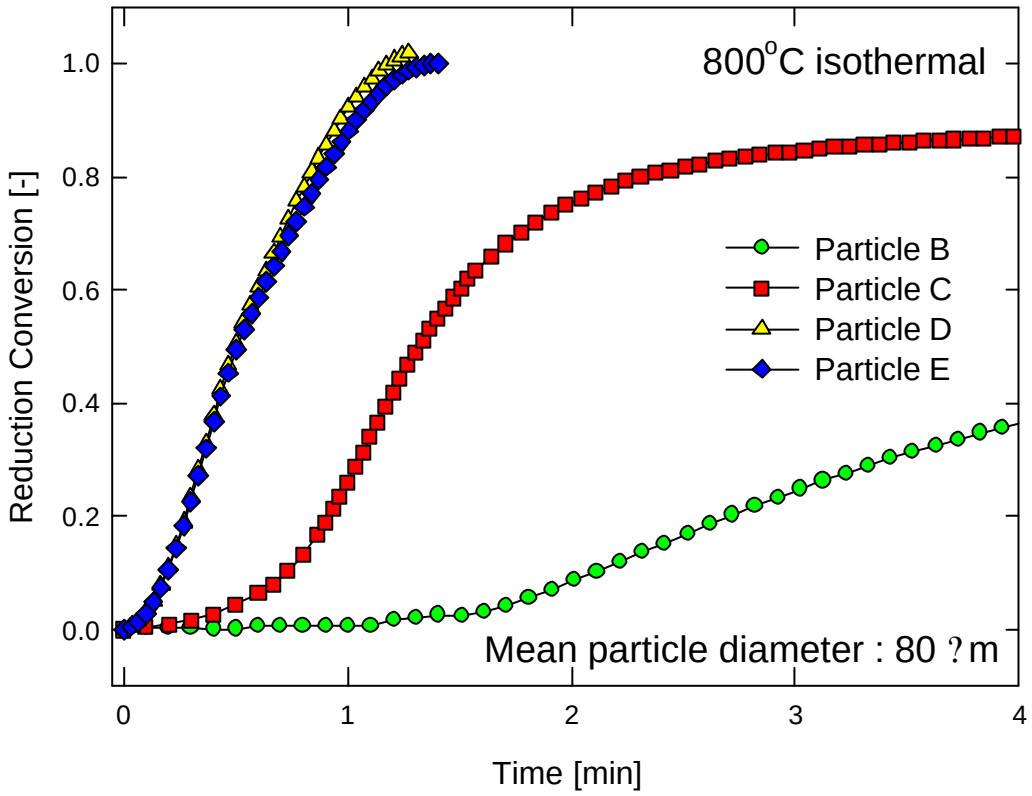
# Results : Oxidation reaction kinetics



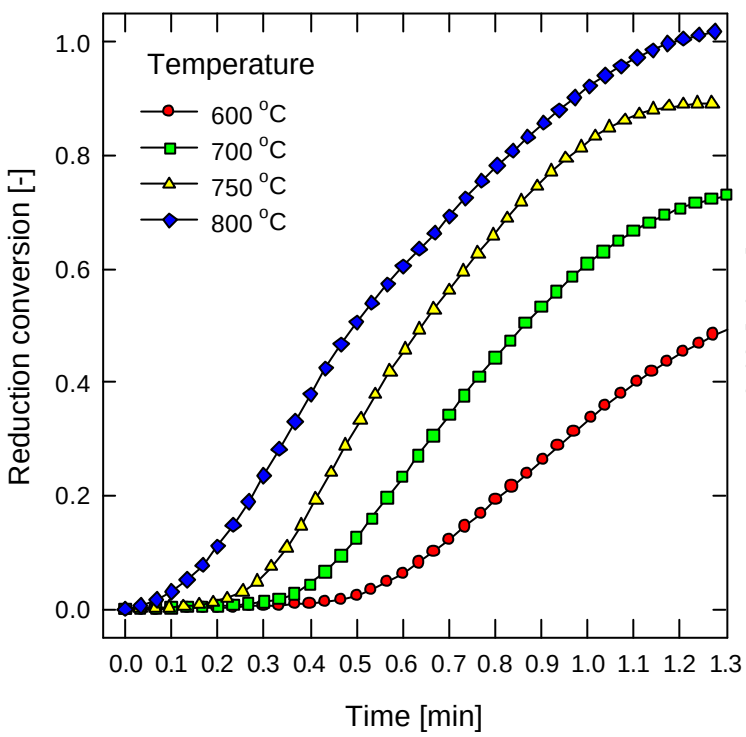

- **Effect of particle size on reaction rate : 80 > 128 > 181 > 284 ?m**
- **dx/dt proportional to $d_p^{-0.313}$**

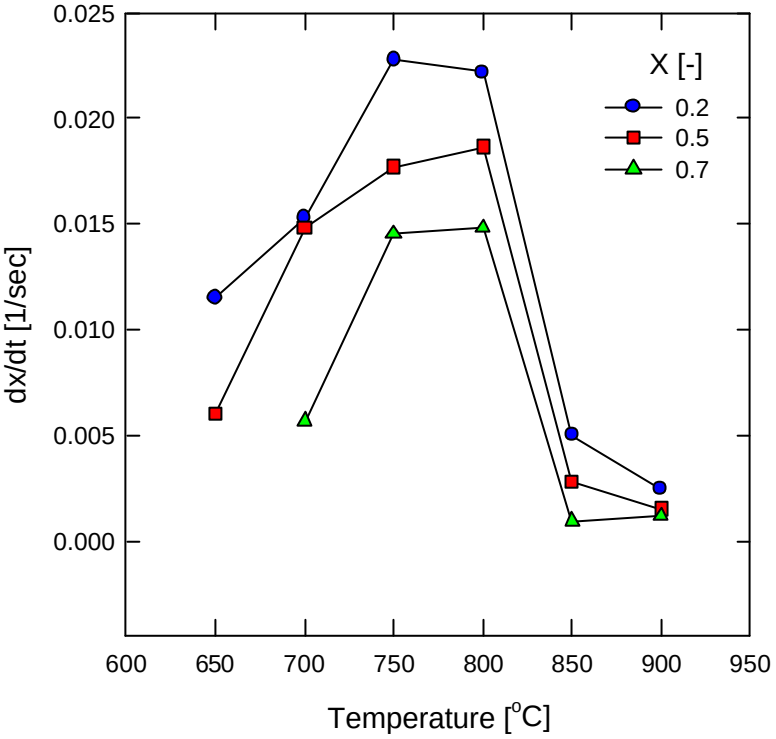
# Results : Reduction reaction




• **Reaction rate : Particle D > E > C > B**

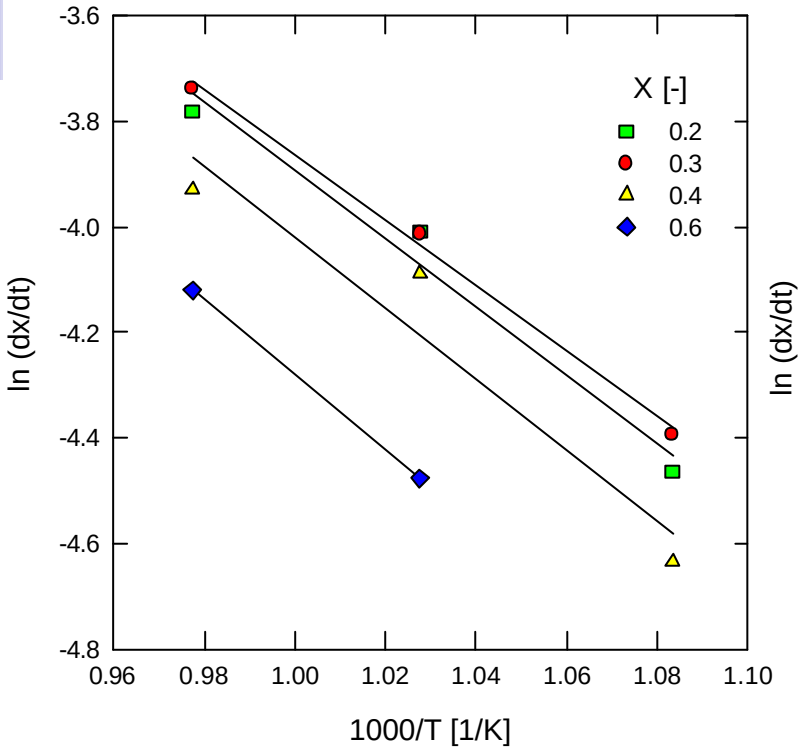
# Results : Reduction reaction

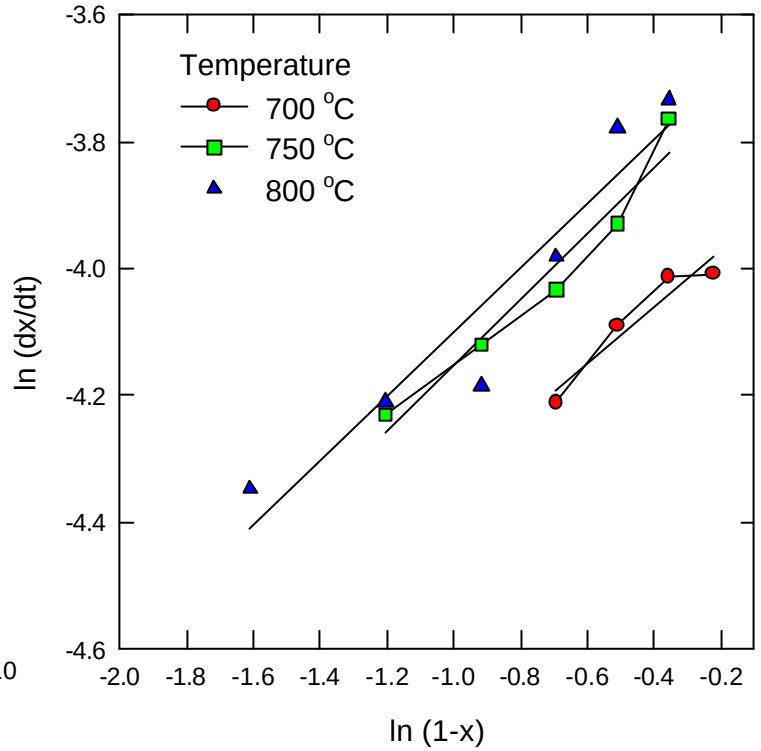



- **Reaction rate : maximum at 750 or 800 ºC**

# Results : Reduction reaction kinetics

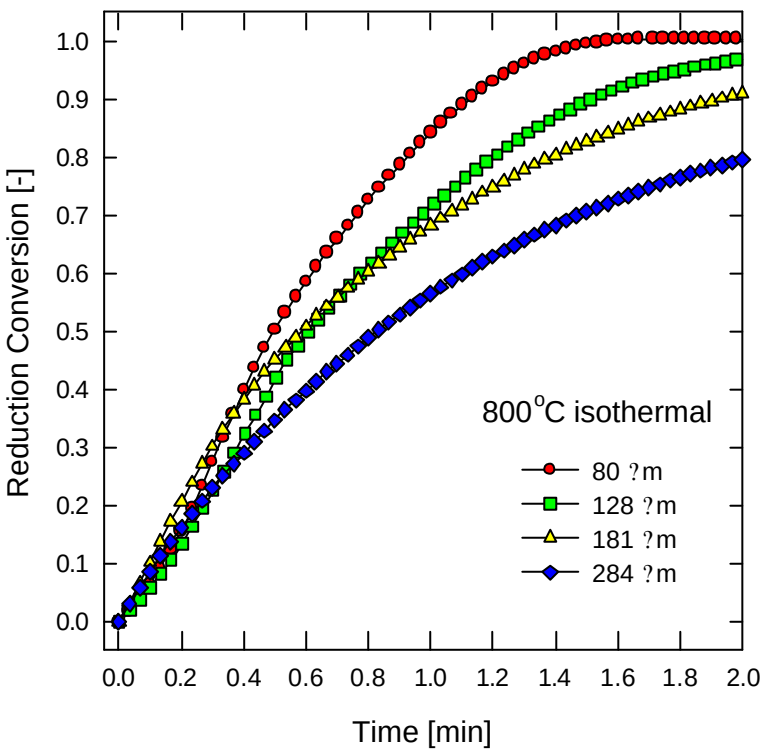


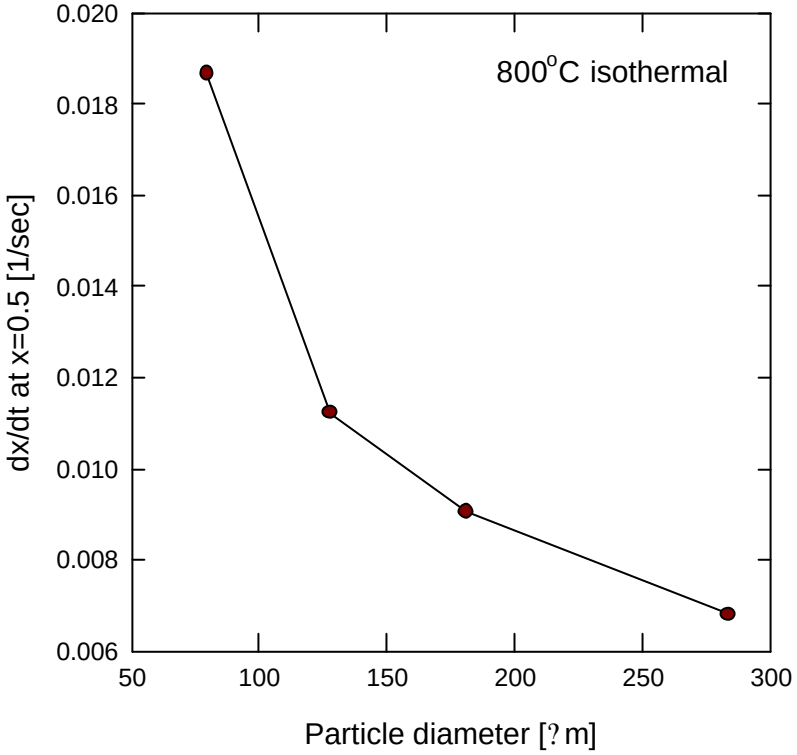




- **ln(dx/dt) vs. 1/T**
  - **Slope = -E/R**
  - **E = 13.1 Kcal/mol**

- **ln(dx/dt) vs. ln(1-x)**
  - **Slope = n**
  - **n=0.491 (0.5th-order reaction)**

# Results : Reduction reaction kinetics



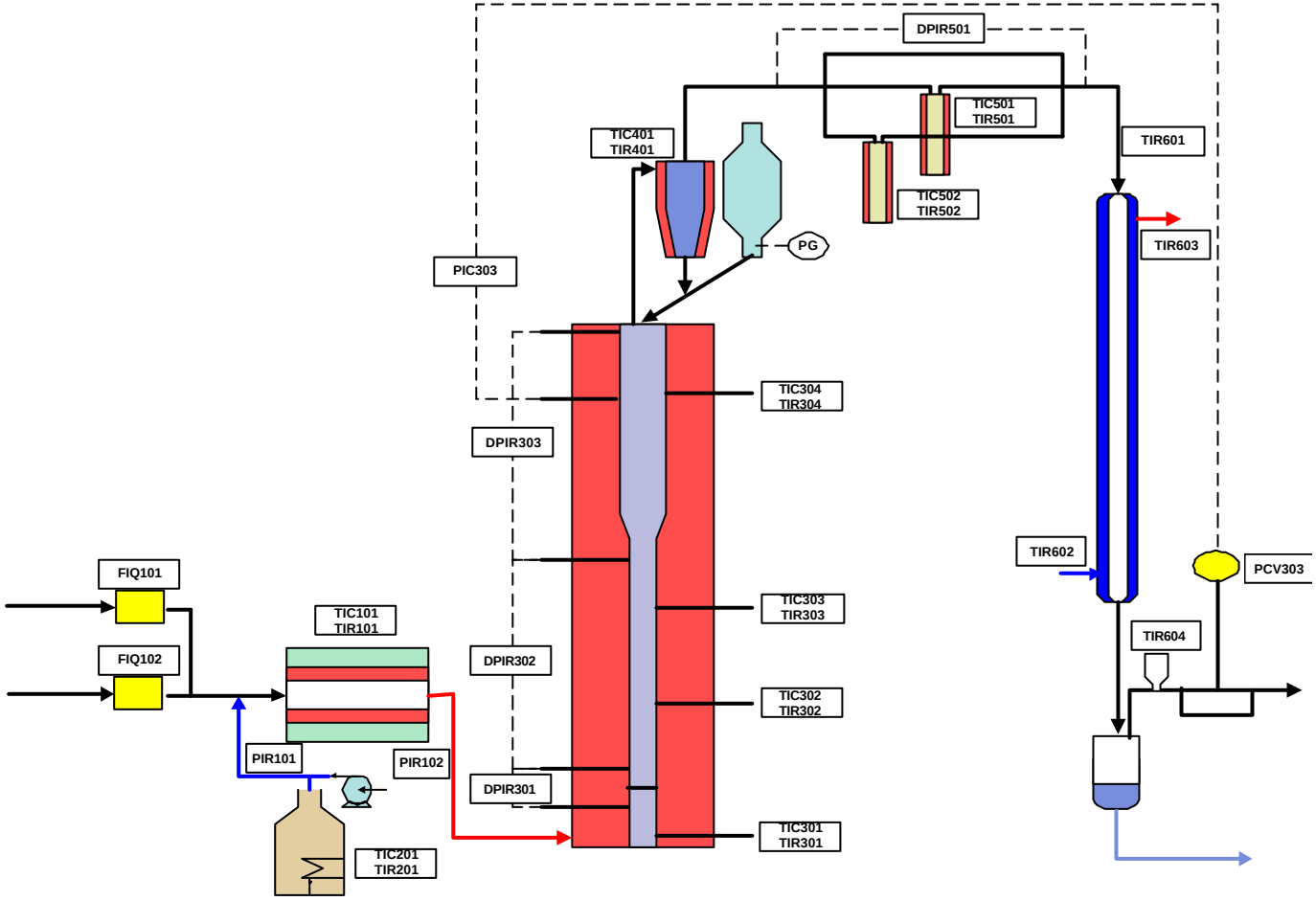




- **Effect of particle size on reaction rate : 80 > 128 > 181 > 284 ?m**
- **dx/dt proportional to $d_p^{-0.857}$**

# Conclusions

| | *Oxidation* | *Reduction* |
|---|---|---|
| *Ni or NiO content* | Particle E > D > C >B | Particle D > E > C >B |
| *Temperature* | 1000 > 950 > 900 > 850 °C | Show maximum at 750 or 800°C |
| *Particle diameter* | 80 > 128 > 181 > 284 ?m | 80 > 128 > 181 > 284 ?m |
| *Reaction order* | 1 | 0.5 |
| *Activation energy* | 35.3 Kcal/mol | 13.1 Kcal/mol |

# Batch Fluidized bed gas combustor

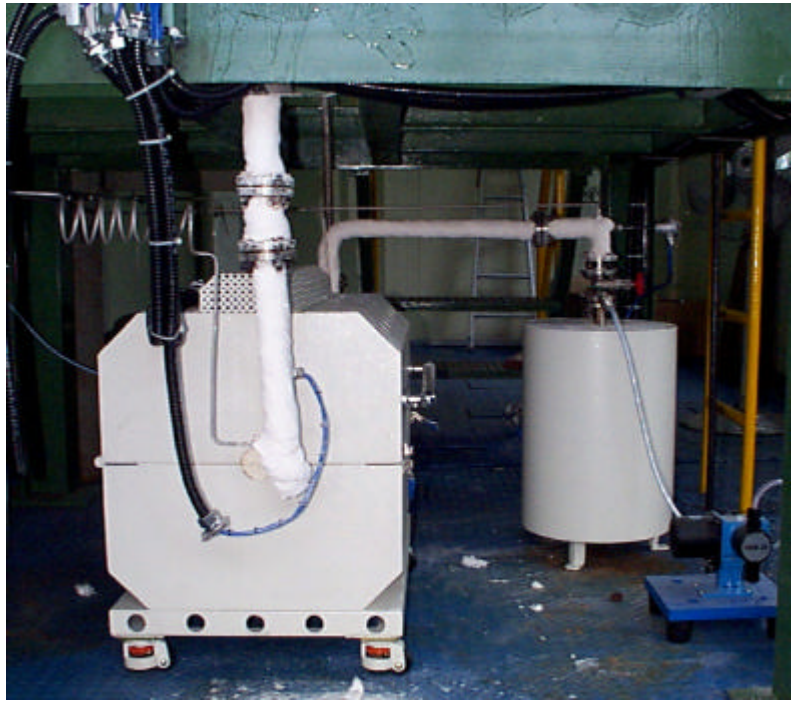


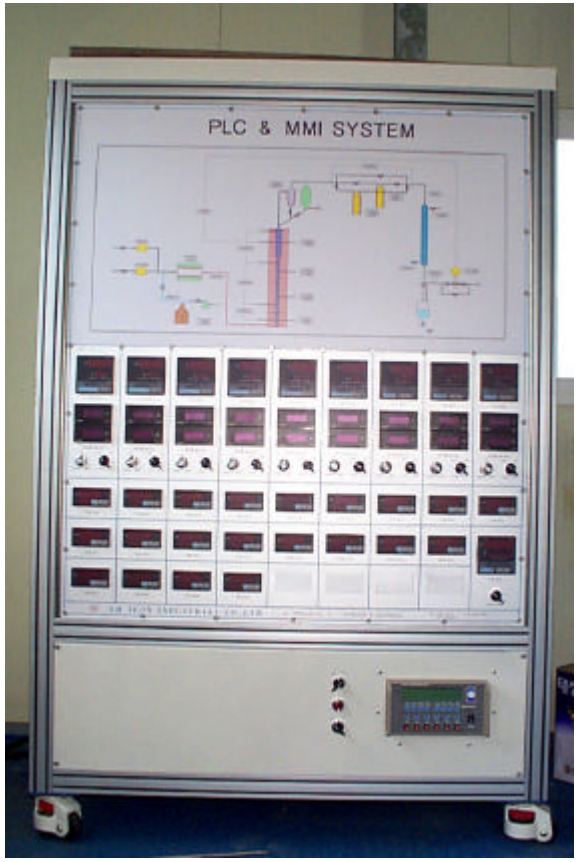
# Batch Fluidized bed gas combustor





# Batch Fluidized bed gas combustor





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