ATTRITION OF LIGNITE CHAR DURING FLUIDIZED BED GASIFICATION: EXPERIMENTAL AND MODELING STUDIES

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Introduction

- The urgent need to capture and sequester CO_2 in particular emitted from the use of coal has triggered the development of new processes like chemical looping combustion and sorption enhanced gasification.
- Both these processes require that fuel gasification is carried out at a relatively low temperature, limiting the types of fuel that could be used to those with highly reactive chars, such as low-rank coals and biomass.
- Lignite is an abundant fuel and makes up approximately 40% of coal reserves in the world. Its commercial use is forecast to grow substantially.
- However, lignite usually has high water and oxygen content, low energy density, and it is expensive to transport. All these characteristics have limited its utilization.

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Introduction

- ✓ Fluidized bed (FB) gasification is acknowledged to have great flexibility in conversion of several solid fuels, including low rank coals, into synthesis gas.
- ✓ Fuel attrition and fragmentation phenomena are well known to affect the reliability and efficiency of FB combustion and gasification processes.
- ✓ Attrition/fragmentation cause the elutriation of fine material from the bed that results in the loss of unconverted carbon.
- ✓ Attrition/fragmentation may also significantly change the particle size distribution of the materials in the bed which influences the bed fluid-dynamics, heat and mass transfer coefficients and reaction rates.
- ✓ Several attrition studies are reported under FB combustion conditions, but only limited activity is reported under gasification conditions, and no study could be found in the literature on attrition of lignite during gasification.

Introduction

✓The authors started a study on attrition of lignite char, pointing out that fragmentation and attrition of the char particles during gasification were significant, and suggested a *gasification-assisted attrition* enhancement effect.

✓This mechanism, associated to the low reactivity of the generated fines, made the loss of carbon by fines elutriation during char gasification more significant than that typically found under combustion conditions.

t_0=0min	t_1=30min	t_2=50min	t_3=70min	t_4=90min	t_5=110min
X_{C0}=0%	X_{C1}=35.9%	X_{C2}=56.3%	X_{C3}=71.2%	X_{C4}=81.0%	X_{C5}=86.2%

t = gasification time, X_C = carbon conversion degree

T= 850 °C, U= 0.4 m/s, Gas = 100% CO_2

Scope of the work

- ✓ In this work the attrition behaviour of char from an Italian lignite (Sulcis) was studied under CO_2 gasification conditions in a lab-scale FB apparatus.
- ✓ In particular, attrition was characterized as a function of the char conversion degree.
- ✓ The influence of bed temperature and CO_2 concentration on attrition and carbon conversion was investigated.
- ✓ On the basis of the experimental results, a kinetic model of lignite char under CO_2 gasification conditions was developed in order to predict the time resolved profiles of carbon conversion.
- ✓ The mechanical resistance of the char particles was also characterized at different stages of char conversion by specific attrition experiments.
- ✓ A semi-empirical model was finally developed to simulate the experimental profiles of carbon elutriation rate as a function of time.

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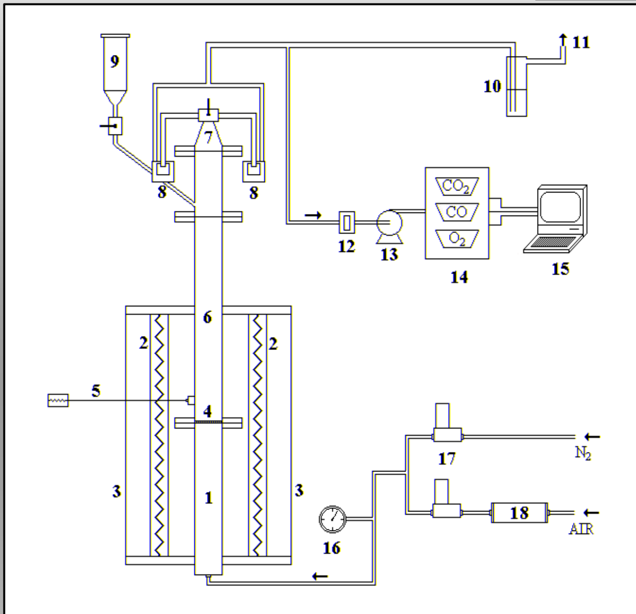
Experimental

Two-exit head FB



1) gas preheating section; 2) electrical furnaces; 3) ceramic insulator; 4) gas distributor; 5) thermocouple; 6) fluidization column; 7) head with three-way valve; 8) sintered brass filters; 9) hopper; 10) SO2 scrubber; 11) stack; 12) cellulose filter; 13) membrane pump; 14) gas analyzers; 15) personal computer; 16) manometer; 17) digital mass flowmeters ; 18) air dehumidifier (silica gel).

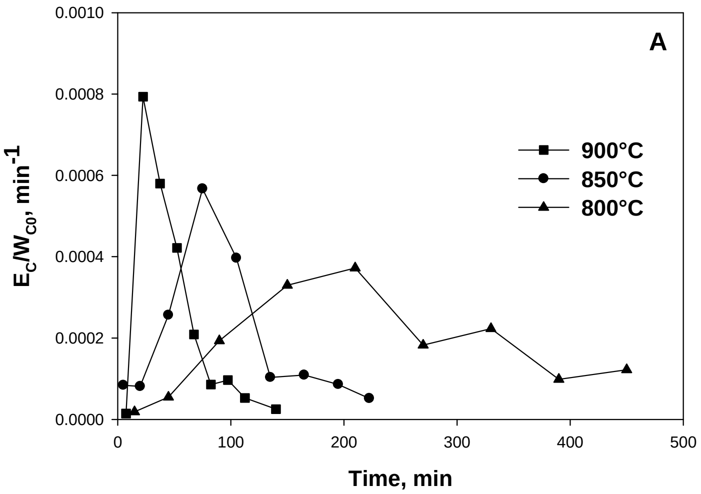
ID = 40 mm
T= 800-900°C
U= 0.4 m/s
CO_2 = 20-100%
d_{fuel} = 6.3-8.0 mm
W_{sand} = 180 g
d_{sand} = 0.3-0.4 mm

Sulcis lignite properties

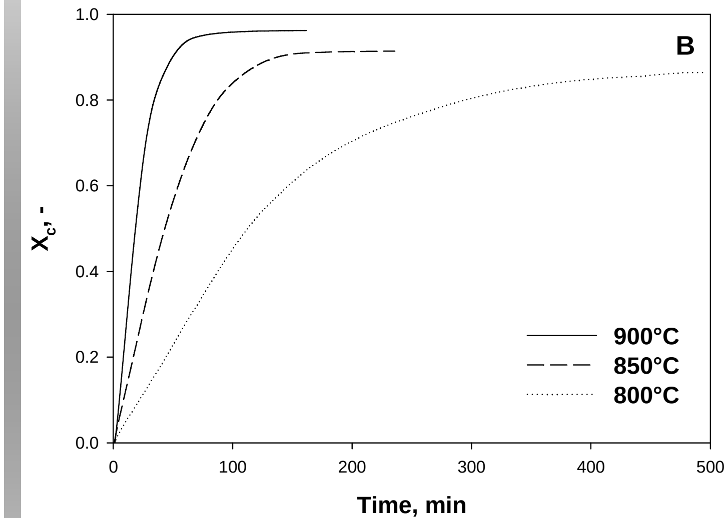
Proximate analysis, % (as received)	
Moisture	6.3
Ash	16.9
Volatile Matter	49.7
Fixed Carbon	27.1
Ultimate analysis, % (dry and ash free basis)	
Carbon	71.7
Hydrogen	5.7
Nitrogen	1.8
Oxygen	13.1
Sulphur	7.7
LHV, kcal/kg	5137

Char attrition tests results – temperature effect

Elutriation rate



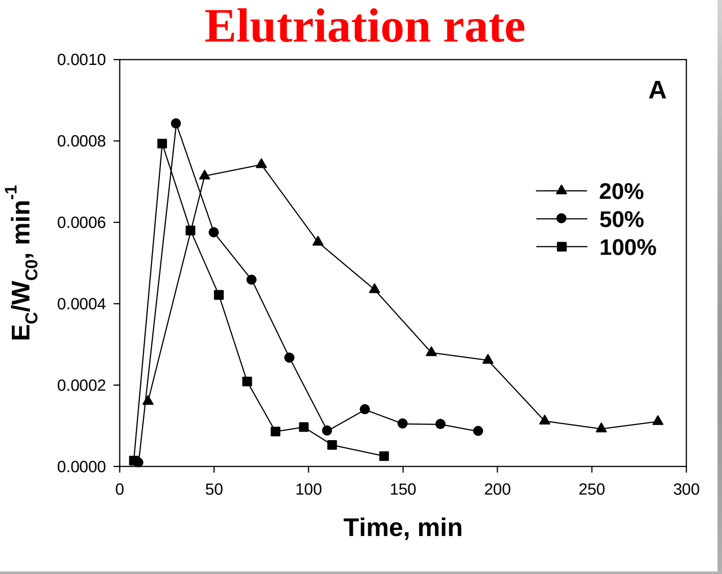
Carbon conversion

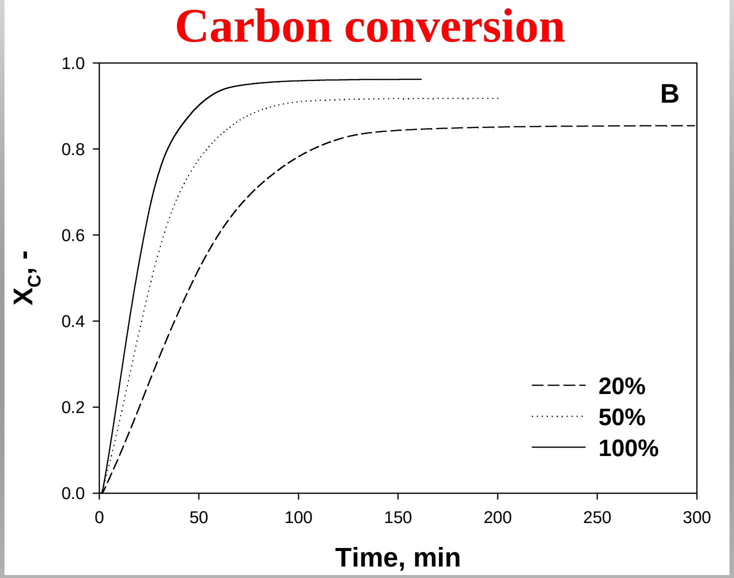


U= 0.4 m/s
W_{batch} = 1 g
Gas = 100% CO_2

Operating Temperature	Gasification time (min)	Gasified carbon (%)	Elutriated carbon (%)	Unconverted carbon (%)
T = 800°C	480	86.4	8.9	4.7
T = 850°C	240	91.4	4.8	3.8
T = 900°C	160	96.2	3.4	0.4

Char attrition tests results – CO_2 effect





T= 900°C
U= 0.4 m/s
W_{batch} = 1 g
Gas = 20-100% CO_2 in N_2

CO_2 inlet concentration (in N_2)	Gasification time (min)	Gasified carbon (%)	Elutriated carbon (%)	Unconverted carbon (%)
20%	300	85.4	10.0	4.6
50%	200	91.8	5.2	3.0
100%	160	96.2	3.4	0.4

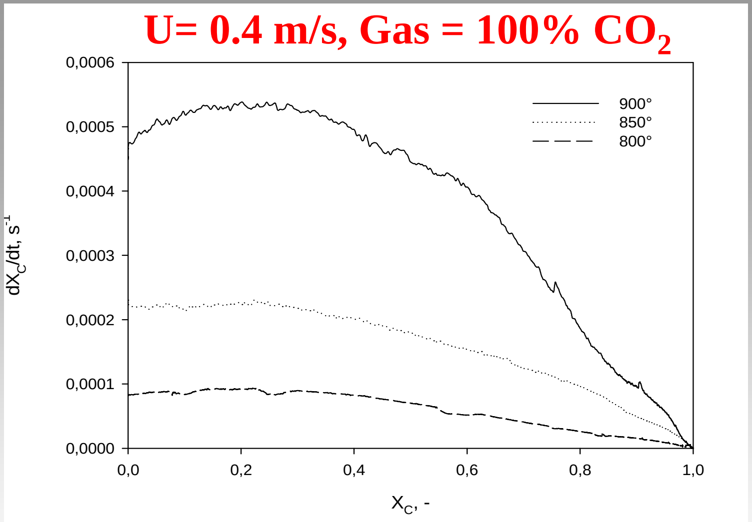
Kinetic model

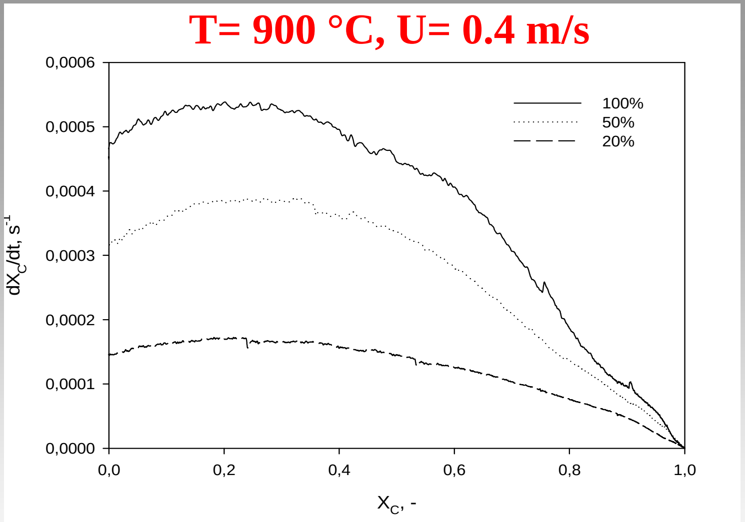
- ✓ Char gasification in the present operating conditions is controlled by intrinsic kinetics.
- ✓ The concentration of produced CO is negligible (< 1% of CO_2 concentration).

$$\frac{dX_C}{dt} = A(X_C) \cdot k_0 \cdot \exp(-E_a/RT) \cdot \left[C_{CO_2} / \left(C_{CO_2} + \beta \right) \right]$$

$$A(X_C) = a + b \cdot X_C + c \cdot X_C^2$$

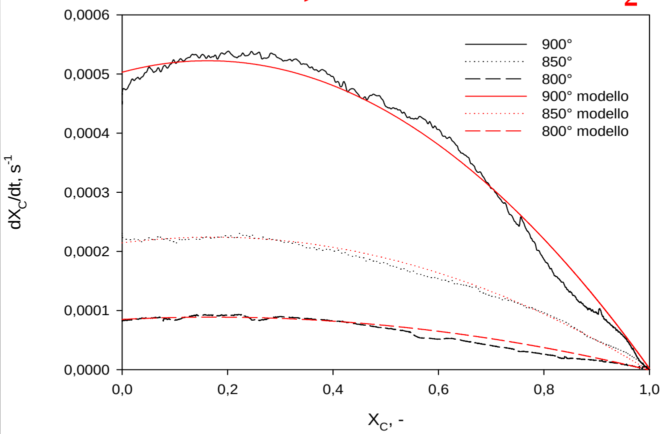
a (-)	b (-)	c (-)	k_0 (s^{-1})	E_a/R (K)	β (-)
0.964	0.460	1.42	$1.53 \cdot 10^5$	22300	0.625

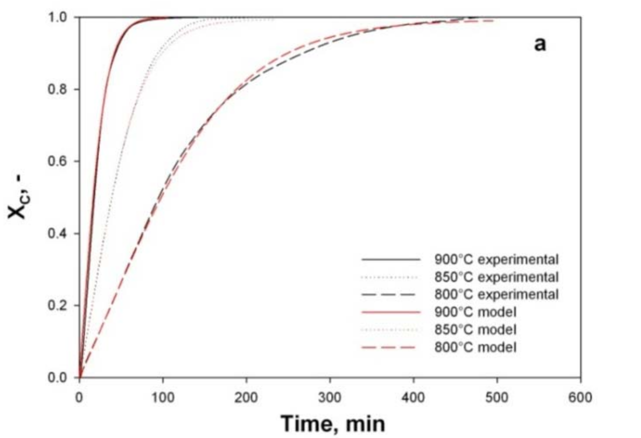




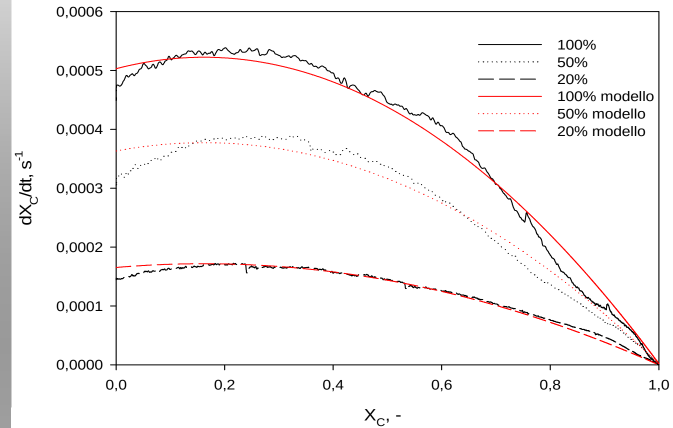
Kinetic model - results

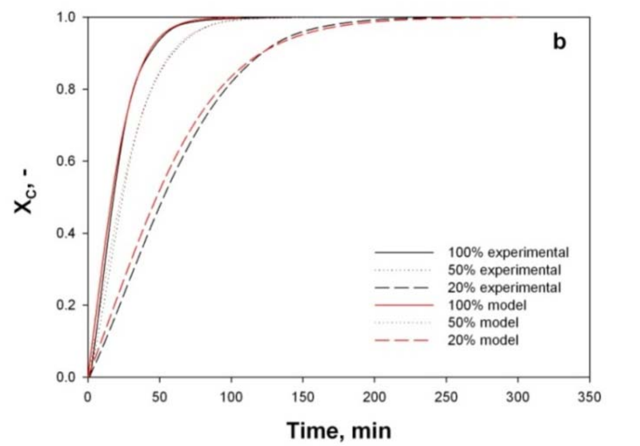
U= 0.4 m/s, Gas = 100% CO_2





T= 900 °C, U= 0.4 m/s



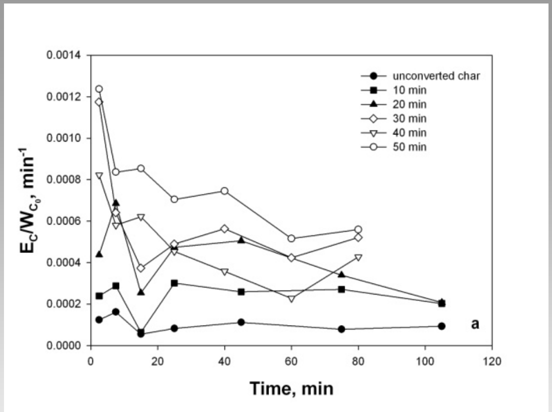


Elutriation model - 1

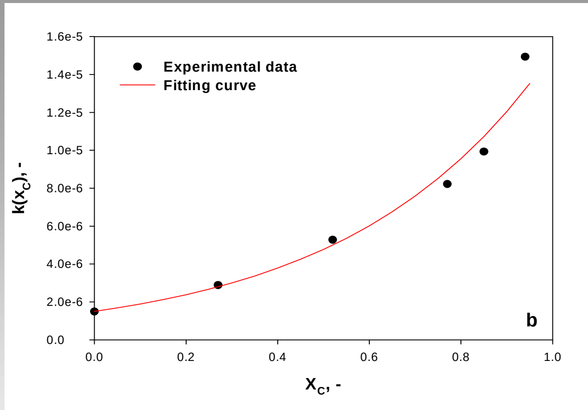
$$E_C = k \cdot (U - U_{mf}) \cdot (W_C / d)$$ **Attrition rate**

$$W_C = W_{C0} \cdot (1 - X_C)$$ **Carbon mass**

In order to understand the relationship between the char attrition constant (**k**) and carbon conversion (**X_C**), attrition experiments were carried out under inert conditions (100% N_2) using char samples pre-gasified for different times (ranging from 10 to 50 min).



Initial values



$$k = 1.5 \cdot 10^{-6} \cdot \exp(2.31 \cdot X_C)$$

Pre-conversion of the char significantly enhances the mechanical attrition of the particles.

Elutriation model - 2

✓ Only limited secondary fragmentation occurs at low carbon conversions, while significant shattering is evident at conversions larger that 50%.

Char particle size

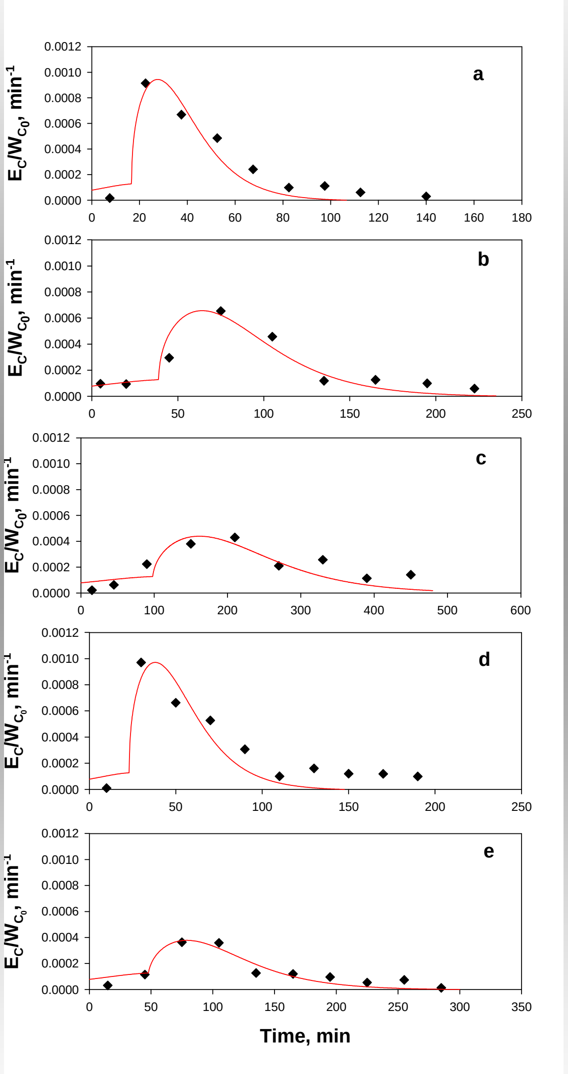
$$d(X_C) = d_0 \cdot \sqrt[3]{1/n(X_C)}$$

Number of char particles

$$n(X_C) = 1 + (n_{max} - 1)\cdot\left[(X_C - X_C^*)/(1 - X_C^*)\right]$$

$X_C^* = 0.5$
n_{max} = fitting parameter

Comparison between experimental and model time resolved carbon elutriation rates obtained during CO_2 gasification experiments at different operating conditions



Conclusions - 1

- A *gasification-assisted attrition* mechanism is proposed. Internal reaction progressively weakens the structure inside the particle, enhancing particle attrition.
- The low reactivity of the generated fines under gasification conditions makes the loss of carbon by fines elutriation significant.
- The total elutriated carbon increased when the bed temperature is decreased. Two reasons are likely to determine this behaviour: i) at lower temperatures the residence time for complete gasification increases; ii) the extent of in-bed conversion of the attrited fines increases with the bed temperature.
- The total elutriated carbon increased when the CO_2 concentration was decreased. At lower CO_2 concentrations the residence time of particles in the bed increases and the extent of in-bed conversion of the attrited fines decreases.
- On the whole, the carbon loss by elutriation is certainly one of the critical factors during the gasification process, especially at low gasification rates (i.e. at low CO_2 concentration and bed temperature).

Conclusions - 2

- A char gasification kinetic model was proposed which was able to correctly predict the carbon conversion as a function of time found in the experiments, including the effect of temperature and CO_2 concentration on carbon conversion.
- A semi-empirical carbon elutriation model was also presented, where the char attrition constant was expressed as a function of the carbon conversion degree.
- The change of the average char particle size due to secondary fragmentation was taken in account.
- Model results were in good agreement with the curves obtained from the experimental tests.
- Particle fragmentation appears to be a key phenomenon in determining the attrition rate of the char in the bed.
- Nevertheless, the presence of a fitting parameter, i.e., the maximum number of generated fragments, implies that the model is not yet predictive at this stage.