



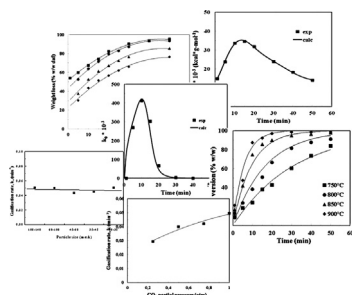
Low rank coal – CO₂ gasification: Experimental study, analysis of the kinetic parameters by Weibull distribution and compensation effect



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GRAPHICAL ABSTRACT



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ABSTRACT

Gasification is generally considered as the most effective for low rank coal exploitation and CO₂ gasification offers the advantage of upgrading an environmentally detrimental gas. Isothermal CO₂ gasification tests of Greek low rank coal were performed in a tubular fixed bed reactor, to study the effect of the various reaction conditions. Greek lignite was quite reactive with CO₂, resulting in high conversions even at short reaction times, while the obtained chars remained reactive after devolatilisation, as the continuous increase of conversion indicated. Conversion was very sensitive to temperature due to both pyrolysis and C–CO₂ reactions, and for the granulometries studied (up to 1000 μm) the gasification rate was independent of particle size. CO₂ partial pressure affected the gasification rate at low CO₂ partial pressures, since the reactivity of lignite remained high (conversion ~85% w/w daf) even for 0.25 atm CO₂ partial pressure. The presence of CO retarded the reaction and the gasification rate decreased rapidly as the concentration of CO increased. The first order Volume Reaction Model (VMR) adopted was proven quite effective in describing the experimental results. Experimental results showed that the activation energy (*E*) varied with time, passing through a wide maximum between 10 and 15 min. The pre-exponential factor (*k*₀) follows the behaviour of the activation energy, however, it exhibits a more sharp maximum at the same time region. The variations of the kinetic parameters with time could be attributed to the different nature of carbon atoms gasified and the changes of the number of active collisions, as the gasification reaction proceeds. As the gasification reaction advanced the Arrhenius parameters (*E* and *k*₀) increased or decreased simultaneously, exhibiting a compensation effect. The probability density function (pdf) of the Weibull distribution was employed successfully to depict the variations of the kinetic parameters, *E* and *k*₀.

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1. Introduction

Coal won the energy race in the first decade of the 21st century, as it accounted for nearly half of the increase in global energy use

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over the past decade, with the bulk of the growth coming from the power sector in emerging economies [1]. Poor quality coal (lignite) poses a special place in the Greek energy system, since it is the most important domestic source and thus, the main fuel for power generation. The main deposits in Greece are that of Ptolemais in Northern Greece (Western Macedonia Region) and of Megalopolis in the South (Peloponnese) and lignite-fired power plants contribute about 50% of the total electricity production in Greece. Therefore, coal is the main energy source for electricity generation today and in the near future, and the CO₂ emitted contributes to the global warming.

A considerable reduction in CO₂ emissions from coal could be obtained by improving the energy efficiency of equipment, using alternative technologies (i.e. IGCC) and/or development and deployment of carbon capture and storage (CCS) technologies. Towards that, carbon capture and storage (CCS) technologies have been developed and deployed recently. Despite the great body of research that has been pursued to control global warming through the sequestration of CO₂ from power plants and oil refineries (the largest single source emitters of CO₂), major shortcomings exist, namely environmental and safety concerns, inefficient capture rates and high costs.

Current efforts to sequester CO₂ usually involve CO₂ capture followed by CO₂ underground storage which requires storage sites adequate to receive CO₂ that should prevent any significant leakage. Geologic sequestration is most popular as a long-term solution however any geologic shift could trigger the release of CO₂, which asphyxiates any life-form that relies on oxygen. In addition, most sites usually are very far from the locations where CO₂ is produced in large quantities, such as power stations. Over the years to come it is believed that the sites capable of storing CO₂ may be depleted and this will then create problems of storage. Most other alternatives for CO₂ sequestration, such as deep ocean sequestration or conversion to mineral carbonates, apart from technical problems, are prohibitively expensive.

Coal gasification is a process for utilizing the energy contained in coal resources, without the traditional process of combustion. The coal is gasified and the product gas is used as feedstock to produce numerous end-use products. Potential end-uses include power generation, liquid fuels and chemical feedstock. Furthermore, in situ coal gasification (i.e. underground coal gasification-UCG) has the potential to greatly increase the availability of otherwise unusable coal resources and, when utilized for power generation or liquid fuels, to reduce the environmental impacts of coal utilization. The application of modern gasification technology (i.e. IGCC) is generally considered as the most appropriate for low rank coal exploitation for energy production. In the case that carbon dioxide (CO₂) is used as gasification medium, it can be more cost effective than conventional carbon capture technologies. Thus, the Boudouard reaction may act as an alternative route for CO₂ sequestration, thereby upgrading an environmentally detrimental gas and adding on climate change mitigation. Therefore, the study of coal – carbon dioxide gasification is an essential feature towards understanding the reaction mechanisms, kinetics and for sizing gasifiers.

Several researchers have studied the influence of the temperature and time, particle size and CO₂/CO partial pressures (i.e. feed gas composition). The CO₂ gasification rate was found to depend strongly on reaction temperature [2–9] while prolongation of reaction time increases conversion [2,7,8]. Contradictory results are given in literature [9–15] for the particle size effect, since some investigators considered to decrease gasification rate [10,11,13,14] while others found insignificant dependence [12,15]. It has also been found that increasing CO₂ partial pressure enhances CO₂ gasification rate, while the presence of CO retards the reaction

[4,5,9,11,13,16]. Low rank coals are characterised by high elemental oxygen content highly dispersed inherent inorganic matter and micro-porous structure. Previous investigators [16–18] concluded that the concentration of active sites is directly related to the oxygen-containing groups of the coal. It has been found also that mineral matter in the coal influences extensively gasification rate by acting catalytically on this reaction [10,18–20]. The volume reaction model is generally employed [12,21–26] for the kinetic analysis of experimental data for coal gasification. However, limited information is given with respect to possible variations of the kinetic parameters, E and k_0 , with reaction time [23–26]. As the gasification reaction proceeds, carbon atoms of different nature and different energy status are gasified, while, changes of the number of active collisions are also taking place. The activation energy distribution is generally assumed to follow the Gaussian distribution, while, the modified Gaussian, the Weibull, the log-normal and the Maxwell–Boltzmann distributions have also been used [27–31]. However, there is no information for possible variations of the pre-exponential factor with the reaction time. The Weibull distribution [32], originally introduced by Rosin and Rammler [33] is a continuous probability distribution with two parameters, convenient to use for a mathematical distribution model and has recently been extensively used for investigating thermal decomposition of carbonaceous materials [33–39].

The effect of the gasification temperature and time, the CO₂ partial pressure, the presence of CO and the particle size were investigated, at ambient total pressure. The volume reaction model was used to study the kinetics of gasification of lignite with CO₂ and the Weibull distribution was elaborated to describe possible variations with reaction time of the activation energy and the pre-exponential factor.

2. Methodology

Greek lignite from Ptolemais reserve was used in this work, and coal samples were kept sealed with their moisture and in the absence of air to avoid any weathering. Proximate, ultimate and ash analyses, of both the raw and catalyst impregnated samples, were performed according to the ASTM Standard Methods [ASTM D 5142–90, ASTM D 5373–93 and ASTM D 6349–98] and the obtained results are given in Table 1. This lignite is characterised by its high moisture, high ash and high volatile, high oxygen and low sulphur

Table 1
Proximate and ultimate analyses of Ptolemais lignite.

(a) Proximate and ultimate analyses			
Proximate	Ultimate (% w/w dry)		
Moisture (% as received)	60.00	C	47.16
Volatiles, (% w/w dry)	47.17	H	4.68
Fixed carbon, (% w/w dry)	36.23	N	1.27
Ash, (% w/w dry)	16.6	S	0.99
CO ₂ , (% w/w dry)	0.87	O ^a	29.3
HHV, (kcal kg ⁻¹)	4393	Ash	16.6
(b) Ash analysis			
Compound	(% w/w)		
SiO ₂	27.80		
Al ₂ O ₃	14.01		
CaO	33.10		
MgO	1.26		
Fe ₂ O ₃	7.99		
SO ₃	17.07		
K ₂ O	0.6		
Na ₂ O	0.6		
TiO ₂	0.1		

^a By subtraction.

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