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INVESTIGATIONS OF THERMAL CONVERSION AND KINETICS OF LOW-GRADE COAL AND BAGASSE USING THERMOGRAVEMETRIC TECHNIQUE.

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Abstract

In the present study thermal characterization of most abundantly available indigenous low grade coal (Chamalang) and residue of agricultural waste (bagasse) have been investigated using thermogravimetric techniques. Proximate and ultimate analyses were also performed and heating values were measured. The operating parameters like heating rate (15, 20, & 40^0C/min), feed composition (85/15, 91/9 & 94/6), equivalence ratio (ER value 0.25, 0.30 & 0 35) that effects the kinetics and thermal conversion process were tested using TGA in non-isothermal mode by maintaining sub-stoichiometric environment. TGA was operated from room temperatures to 950^0C and it was observed that these parameters have very pronounced effects on the kinetics and conversion of these fuels. Coal-bagasse blends 91/9 (% w/w), heating rate of 20^0C/min and ER value 0.30 gives the optimum conversion. These parameters also had shown a substantial effect on the frequency factor (A^{-1}) and little impact on activation energy (Ea) that was determined by the regression analyses of weight loss data of TGA. These results contributed to the understanding of proper thermochemical conversion processes for co-firing at the optimum operating conditions for the gasification reactor.

Key words: Characterization, Thermal conversion, co-gasification, TGA

Nomenclature

CH:	Chamalang mine coal
$_{\text{Sub}}$:	Sub-bituminious caol
Ea:	Activation energy
A^{-1}:	Frequency factor
HHV:	Higher heating value
ER:	Equivalence ratio
CFBG:	Circulating fluidized bed gasifier
LPM:	Litre per minute
TGA	Thermogravimetric Analyser

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1. Introduction

In recent years, due to the continuous increase in oil prices, the utilization of coal and other renewable resources including biomass as an energy source have already shown the potential of economical competitive advantage [1]. Biomass is believed to be economically competitive with the oil. In global terms, biomass ranks fourth as energy resource making up 18 % of the world's primary energy needs [2]. It includes a wide range of organic material such as wood-based energy crops, agricultural residues, algae, municipal wastes, forest product wastes, paper, and cardboard and food waste [3, 4]. Pakistan has the huge resources of both kinds of fuels as it stands 5th in coal reserve as estimated 185 billion tones [5] and the world's 5th largest producer of the sugarcane in terms of acreage. Sugarcane is grown over millions hectares area of the country and provide the raw material for more than 84 sugar mills throughout the country. After textile, sugar industry is the 2nd largest agro base industry in Pakistan. For the year 2011-12, the sugarcane production is estimated at 58 MT over the previous forecast [6]. The total crushing of the sugarcane over the year 2011 was 78.15 MT. The bagasse is 34 % of the cane crushed with average moisture contents of 50%, so total available bagasse from these sugar mills is estimated more than 19.72 million tons.

Both biomass and coal are carbonaceous materials, originating from plants and have the same fundamental elemental constituents [7]. The co-utilization of coal and these renewable resources of biomasses are becoming popular as fuel for all thermal conversion processes like pyrolysis, combustion & gasification due to the dual role of biomasses, both as energy source and carbon dioxide reducing agent in the environment. Moreover, high contents of hydrogen (H) in biomass, making it appropriate as a blend to compensate the often-low hydrogen (H) content of coal. The disadvantages of biomass as gasification feedstock are the low energy density, high moisture and oxygen contents, even though giving a high hydrogen yield. This deficiency is compensated by blending with a higher energy content coal [8]. The significant research interest in co-gasification of various coals and biomass mixtures can be seen such as in Japanese coal and cedar wood [9], coal and saw dust [10].

The high inorganic matter of biomasses contributes to catalyse the gasification of coal. Most of the co-gasification studies have focused on the parametric studies of the gasifier [11, 12], while little attention has been given to the mechanism by which these blends of low-grade coal and biomass thermally interact and degrade in co-conversion environment. The interaction among coal and biomass during thermal co-conversion is still an issue and yet to be solved. Some de-volatilization and pyrolysis results of coal and biomass blends have revealed a very little or no synergy between two fuels [13,14], while some others have revealed significant interaction among them [15, 16, 17 and 18].

The suitability of material for gasification mainly depends on the certain fuel characteristics like heating value, moisture contents, feed size and thermally conversion behaviour [19]. Thermogravimetric is the thermal degradation process which mainly depends on the atmosphere in which it occurs, as it may be inert for pyrolysis, oxidizing for combustion and sub-stoichiometric for gasification. Recently many researchers have paid their attention to the characterization of coal and biomass fuels for co-gasification process that takes place in reducing atmosphere [20, 21]. While some other has investigated the factors effecting the kinetic parameters and thermal decomposition of coal, bagasse and coal-bagasse blends [22].

The kinetic studies can be done both by applying isothermal and non-isothermal techniques; therefore, it is still a controversial issue [22,23]. .Non-isothermal method has been found of much interest for heterogeneous reactions among the researchers [22, 24, 25 and 26]. For example Otero et al [27] have investigated non-isothermal thermo-gravimetric analysis (TGA) of two different carbonaceous materials i.e. coal and sewage sludge, for combustion process and recommended to use non–isothermal TGA with some modification for kinetic measurements of coal reactivity. Investigations have been reported about the reactivity and kinetic behaviour of Thar coal (lignite) using non-isothermal TGA [28].

In the present study the kinetic parameters and conversion behaviour of different coal-biomass blends in sub-stoichiometric environment were investigated by varying the operating parameter such as heating rate, feed composition, and oxidant flow rate using TGA. Non-isothermal method was applied and weight loss studies were carried out for fixed time between atmospheric temperatures to 950^{o}C.

2. Materials and Methods

Low rank coal samples namely CH_{sub} (Chamalang) belongs to Balochistan, province of Pakistan and dry bagasse samples collected from local sugar mill were tested using the TGA (Leco 701) at Coal Research Centre of NFC IET Multan. The grinded coal was segregated into sizes 710, 500 and 355µm. The CH_{sub} coal size 710µm and bagasse of average particle size 490µm were finally selected as co-fuel for characterization in gasification environment.

Table-1: Analysis of low grade coal and bagasse.

Analysis (wt %)	Sugar Cane Bagasse	CH_{sub} Coal
Proximate Analysis		
Fixed carbon	12.28	51
Volatile matter	81.33	39.8
Moisture	1.03	0..9
Ash	5.35	8.3
Ultimate Analysis		
Carbon	43.07	59.08
Hydrogen	6.6	5.60
Sulfur	0.16	2.79
Nitrogen	1.41	1.409
Oxygen*	43.41	22.82
HHV MJ/kg	17.88	22.20

*Oxygen by difference

Table-2: Analysis of coal-bagasse blends

Sample CBB	M (%)	V.M (%)	F. C (%)	Ash (%)	HHV .MJ/ kg
94/6	0.91	42.29	48.67	8.12	21.95
91/9	0.92	43.54	47.51	8.03	21.81
85/15	0.93	46.03	45.21	7.86	21.55

Thermal characterization of coal-biomass blends as fuel for gasification process was investigated using TGA in controlled air environment. Coal-bagasse blend samples of various proportions were prepared and placed in the sealed bags. Tested parameters was feed composition, heating rate and air flow rate for temperatures ranging from atmospheric to 950^{0}C. The analysis were performed for feed compositions (85/15, 91/9 & 94/6), heating rates (Ramp rate 40, 20, 15^{0}C/min) and Air flows of 3.5 and 5 LPM. ER values were determined as 0.25, 0.30 and 0.35 for weight sample 3.5, 3.0 and 2.5 gram respectively. The conversion performance of these samples under said operating conditions was tested. The initial weight & loss in weight with time & temperature were recorded continuously during this thermal conversion process and regression analyses were performed for kinetics data.

3. Results and Discussions

3.1 Effect of Operating Parameters

The extent of conversion for coal biomass blends in series of gasification reactions can be obtained by the following equation:

Conversion = (w_i-w_t)/(w_i-w_∞) (1)

Where w_i represents initial weight, w_t is the weight at any time, t and w_∞ is the residual weight of the sample.

3.1.1 Heating Rate

As depicted in the Figures 1 and 2 there was a little shift of thermo-grams for heating rates of 40, 20 & 15°C/min. Due to non uniform distribution of heat at higher heating rate, there was slow conversion but with the passage of time, conversion rates of as high as 49.82% for ramp rate 20 was observed which corresponded to 37.25 mins. Similarly, conversion rates of 37.49% & 37.16% were observed for ramp rate 40 & 15^{o}C/min, corresponding to 18.72 and 48.88 mins respectively. De-volatilization was completed at 750^{0}C. This was perhaps due to release of highest amount of volatile matters and moisture during this temperature range (0-750^{0}C), which causes series of chemical reactions. In de-volatilization step the fuel releases CO, CO_2, H_2O & hydrocarbons (i.e. CH_4, C_2H_4, and C_2H_6) in the product gas [29]. Some of these reactions are endothermic, which caused to slow down conversion at higher heating rate earlier. However, as it exceeded the said temperature range and attained temperature 750-950^{0}C, the heating rate of 40^{0}C/min regained the highest conversion rate i.e. 47 % as against the conversion of 36.5% & 27.3% for heating rate 20 & 15^{0}C /min respectively. Under these operating conditions, 20 ramp rate was found to be optimum with the highest overall conversion value i.e. 86% as against 80% and 64% for ramp rates of 40 and 15, respectively

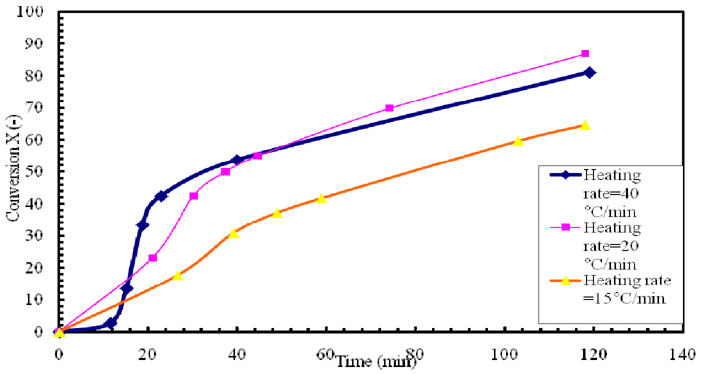

Fig. 1 Effect of heating rate on Conversion of coal-bagasse blend (91/9) at ER value =0.30

.As suggested by some researchers that between the temperatures 700-950^{0}C, a series of endothermic reactions between residual char of de-volatilization step and oxidant took place which resulted in higher yields of CO, CO_2 and H_2 in the product gas[30]. Thus higher heating rate provided the necessary heat for this conversion. Hence the rate of conversion of blended fuel at various heating rates forms the basis of designing a gasification system for the production of producer gas using circulating fluidized bed gasifier (CFBG).

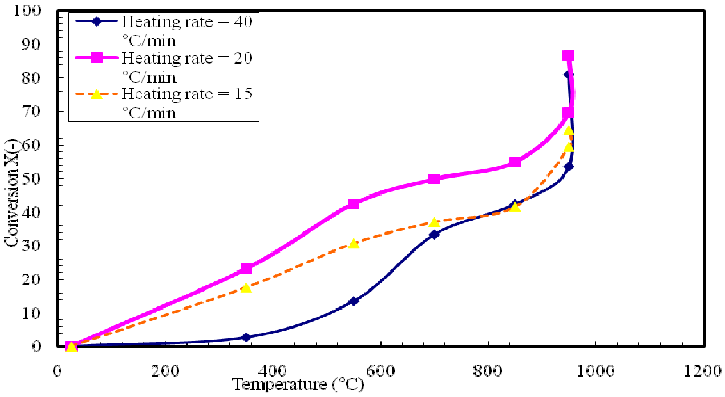

Fig. 2 Time dependent conversion of coal-biomass blend (91/9) at different heating rate (ER=0.30)

3.1.2 Feed Composition:

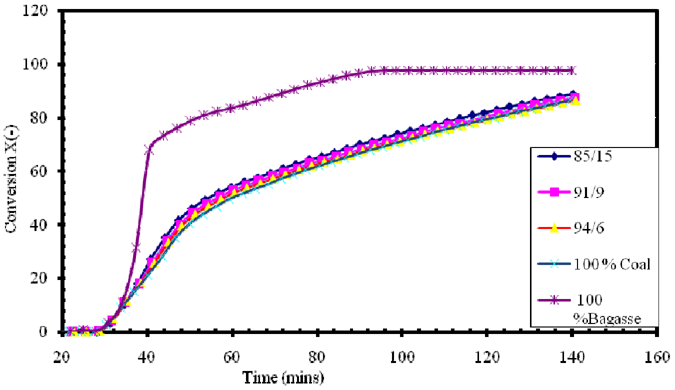

Fig.3 Effect of feed composition on thermal conversion (ramp rate 20^{0}C/min and ER value =0.30)

Bagasse contains high fraction of oxygen as compared to coal as shown in Table 3. The higher proportion of oxygen increases the ER value and promotes the partial oxidation or combustion reaction of gasification [31]. Ash is another important factor of feedstock as it promotes the clinker formation and can cause the problems for the gasifier operation due to slugging and consequently ash agglomeration due to fusion of ash at low temperature. Biomass having ash-content above 5%, can initiate the clinkering formation. Successful gasification with ash-content up to 25% has been reported.

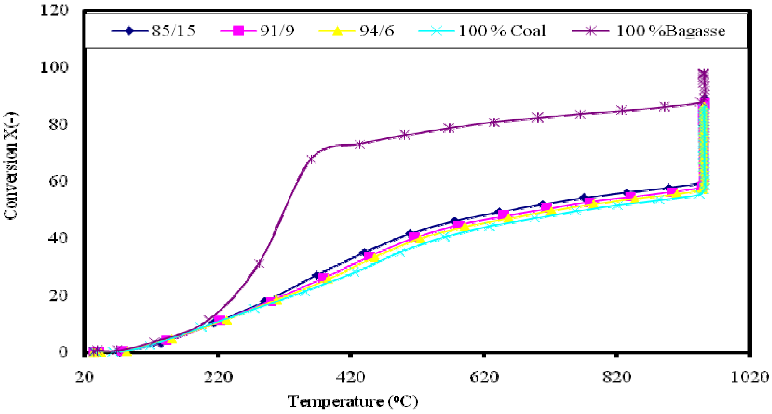

Fig. 4 Time dependent conversion of coal, bagasse and blends (ramp rate 20 ^{0}C/min and ER value =0.30)

Figures 3 & 4 depicted that with increase in the fraction of biomass in coal-biomass blends (100% coal, 100% bagasse, 94/6, 91/9 & 85/15), the conversion of the feedstock increased with temperature or time due to increasing fraction of volatile matters, available oxidant and decreasing ash content. However, heating value decreased with increasing fraction of biomass in feedstock as given in Table2. Similar values have been reported by Sonobe and Worasuwannarak [32]**.**

3.1.3 Equivalence Ratio (ER)

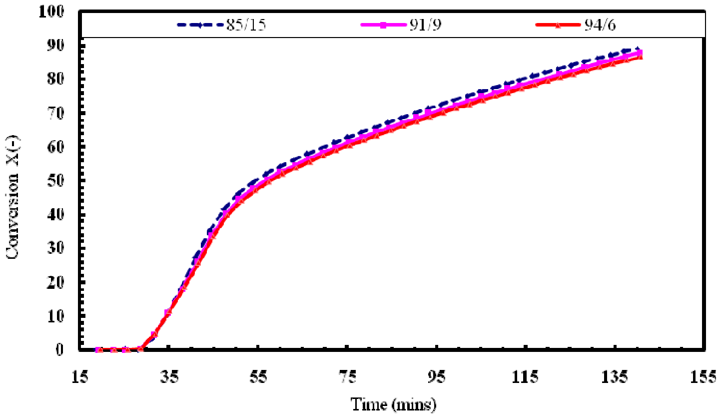

Fig. 5 Conversion of coal-bagasse blends at ER=0.25 and ramp rate 20 ^{0}C/min

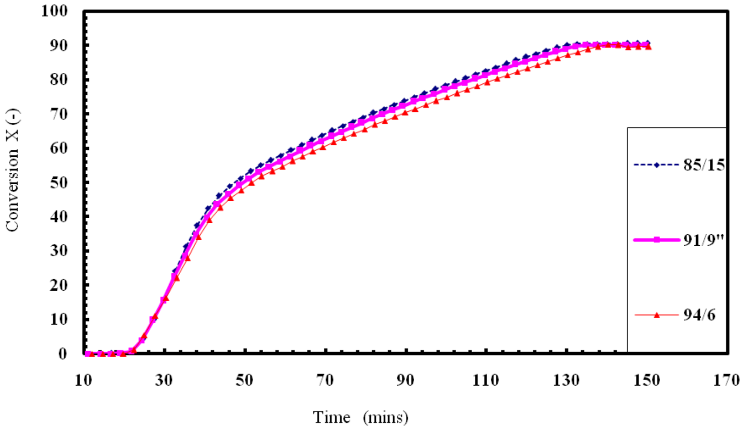

Fig.6 Conversion of coal-bagasse blend at ER=0.30 at constant ramp rate 20 ^{0}C/min

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In gasification process, the amount of air supplied determines its degree of combustion that causes to increase the combustion zone temperature and provide heat for directly heated gasifier [29]. The composition of producer gas is strongly influenced by the air flow in many ways. It supplies the oxygen for combustion and fluidizing media in case of fluidized bed and also affects the residence time and superficial velocity.

Moreover, air flow rate controls the degree of combustion which in turn, affects the conversion temperature and composition of product gas. Figures 5 to 7 presents the effect of ER values 0.25, 0.30 & 0.35 on conversion of coal-biomass blends 85/15, 91/9 and 94/6 respectively for change in temperature from atmospheric to 950°C while keeping other conditions constant. Increasing the air supply increased the ER, which in turn improved the conversion process. Steeper conversion was observed for coal-bagasse blend (85/15) containing larger fraction of biomass.

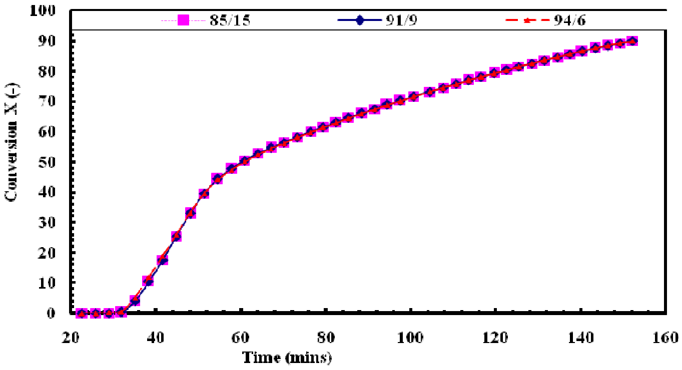

Fig.7 Conversion of coal-bagasse blends at ER=0.35 at ramp rate 20 ^{0}C/min

Increasing fraction of biomass in blends increased the conversion, due to more availability of elemental oxygen from biomass. Many of the researchers have reported the increase in gas yield with increase in ER. It implies that an increase in the availability of oxidant faster the conversion. This is in agreement with the previous research work of the researchers [33, 34].

4.2 Estimation of Kinetics Parameters

To characterize the selected coals, biomasses and coal-bagasse blends thermogravimetric data was used to calculate the kinetics of the reaction that occurred during thermal conversion in specific conditions. As TGA measured the overall weight loss due to the reaction; hence the overall kinetics can be determined. For kinetic calculations following assumptions were made;

1. Reaction is purely kinetics controlled
2 The conversion process follows the first-order reaction
3. Eliminates the effects of heat transfer due to smaller particle size.

Several methods are available in literature that can be used to calculate the activation energy [35]. The fundamental rate equation for heterogeneous solid-state reactions can generally be explained by:

$$\frac{d\alpha}{dt} = K(T)\,f(\alpha) \qquad (2)$$

Where t is the time K (T) is the temperature dependent rate constant and f (α) described the reaction model, a function depending on the actual reaction mechanism, which presents the dependence of the reaction rate on the extent of reaction α. Generally well known Arrhenius equation is used to explain the temperature dependence of the rate Constant.

$$K(T) = Ae^{-Ea/RT} \qquad (3)$$

$$\frac{d\alpha}{dt} = Ae^{-Ea/RT}(1-\alpha)^n \qquad (4)$$

Where A is the frequency factor in (min^{-1}) Ea is the activation energy in KJ/mole for the conversion process . R is the universal gas constant (8.314 J K^{-1} mole $^{-1}$) T (K) is the absolute temperature; n is the order of reaction.

Data obtained at constant heating rate $\beta = \frac{dT}{dt}$, following equation can be expressed:

$$\beta = \frac{d\alpha}{dT} = \frac{d\alpha}{dt} \times \frac{dt}{dT} \tag{5}$$

The term $\frac{d\alpha}{dT}$ is the non-isothermal reaction rate and substituting eq. (5) into eq. (4) will result as follow:

$$\frac{d\alpha}{dT} = \frac{A}{\beta} e^{-Ea/RT} (1-\alpha)^{n} \tag{6}$$

Rearranging and integrating eq. (6), the following expression can be obtained:

$$\frac{1-(1-\alpha)^{1-n}}{1-n} = \frac{A}{\beta} \int_{0}^{T} e^{-Ea/RT} dT \tag{7}$$

Since $\int e^{-Ea/RT} dT$ has no exact integral,
$e^{-Ea/RT}$ can be expressed as an asymptotic series and its integration with ignoring the higher–order terms gives:

$$\left[\frac{1-(1-\alpha)^{1-n}}{(1-n)}\right] = \frac{ART^{2}}{\beta Ea}\left[1 - \frac{2RT}{E} e^{-Ea/RT}\right] \tag{8}$$

Expressing eq. (8) in logarithmic form result in following equation

$$\ln\left[\frac{1-(1-\alpha)^{1-n}}{T^{2}(1-n)}\right] = \ln\left[\frac{AR}{\beta Ea}\left(1-\frac{2RT}{Ea}\right)\right] - \frac{Ea}{RT} \text{ for } n \neq 1 \tag{9}$$

If assuming that 2RT/Ea<< 1 than eq. (9) becomes:

$$\ln\left[\frac{1-(1-\alpha)^{1-n}}{T^{2}(1-n)}\right] = \ln\left[\frac{AR}{\beta Ea}\right] - \frac{Ea}{RT} \qquad \text{for } n \neq 1 \tag{10}$$

In order to simplify the calculations, the order of the reaction, n is assumed to be unity, and hence eq. (10) can be presented as follow

$$\ln\left[-\frac{\ln(1-\alpha)}{T^{2}}\right] = \ln\left[\frac{AR}{\beta Ea}\right] - \frac{Ea}{RT} \tag{11}$$

Above eq. (11) will result in a straight line with slope E/R and an intercept of in [AR/βEa]. This was done by plotting graph between following

$$\ln\left[-\frac{1-(1-\alpha)}{T^{2}(1-n)}\right] = versus \frac{1}{T} \text{ (For n≠1)} \tag{12}$$

$$\ln\left[-\frac{\ln(1-\alpha)}{T^{2}}\right] versus \frac{1}{T} \text{ (For n=1)} \tag{13}$$

The values of α and T obtained from the TG analysis would have been used. Using data from thermograms of a variety of coals, biomasses and coal-biomass blends discussed earlier, the kinetic parameter, activation energy (Ea) was estimated. The linear correlation coefficients criterion was used for the best acceptable value of Ea. The orders of the reactions were first order for all the operating conditions.

Using the data from the thermo grams shown in Figure.1, the kinetic parameters i.e. activation energy (Ea) and frequency factors (A^{-1}) were estimated. For all the operating parameters, 1st order reaction for the thermal conversion process was assumed. The results of the kinetic parameters at various conditions were tabulated in Table 3.

Table 3 Kinetic parameters and respective R^2 constant values at various operating conditions

Parameter	**Values**	**Kinetic parameters**		**R^2-Constant**
		Ea KJ/mole	**A^{-1}** min	
Heating rate °C/min	40	20.363	7.25e+2	0.88
	20	24.05	1.235e+3	0.92
	15	24.5	1.338e+3	0.94
Feed composition by weight %	100% coal	23.83	1.604e+3	0.95
	100 % bagasse	33.6181	8.422e+3	0.96
	85/15	26.539	1.985e+3	0.90
	91/9	26.54	1.606e+3	0.89
	94/6	24.286	1.418e+3	0.88
ER values	0.35	25.64	1.604e+3	0.94
	0.30	24.98	1.607e+3	0.90
	0.25	26.755	1.68e+3	0.93

The results indicated that there were slight variations in activation energies (Ea), however a larger variation in frequency factors was observed against the various heating rates. Thus, it indicated that heating rate has more pronounced effect on frequency factor (A^{-1}) rather than on activation energy (Ea). Therefore, it suggested that higher heating rate could ease and promote the conversion of fuel blend under this sub-stoichiometric environment. A similar value of activation energies (Ea) close to this study for coal-biomass blends in oxidizing atmosphere has been reported in the literature [36]. These investigations were found quite in fair agreement with the findings of researchers [30], whom has reported the Ea values 54.1, 54.8, 55.2 and 55.3kJ/mole for heating rate 5,10,20 & 30^0C/min respectively for the pyrolysis of palm solid waste. Hence, the results of this study also show the similar variation trend against the various heating rates for coal bagasse blend for this sub-stoichiometric environment.

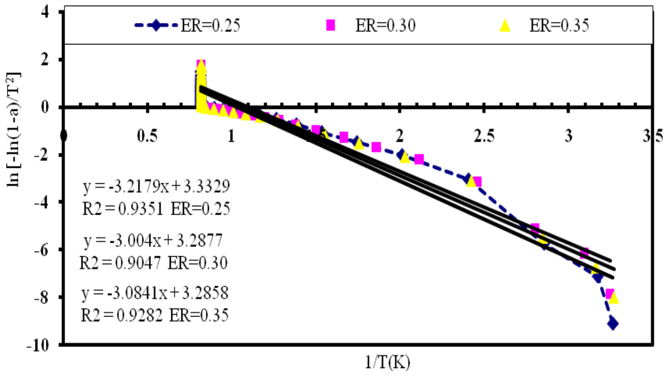

Fig-8 Kinetic data of coal-bagasse blend 91/9

Data of thermo grams from Figure-3 was used to estimate, the kinetic parameters of CH_{sub} coal, bagasse and various coal-bagasse blends. It was observed that the bagasse was highest in Ea and A^{-1} values as compared to coal and coal-bagasse blends. Increased in kinetics parameters (Ea & A^{-1}) with increasing proportion of bagasse was due to higher fraction of V.M and other constituents like cellulose and hemi-cellulose present in biomasses. Hence, increased fraction of bagasse in coal increased the frequency factor (A^{-1}), which in turn ease and faster the conversion as shown in Figure 3. These observations were found quite in line with the findings of some other investigators [37], who characterized the varieties of coal, biomasses and coal-biomass blends for pyrolysis process.

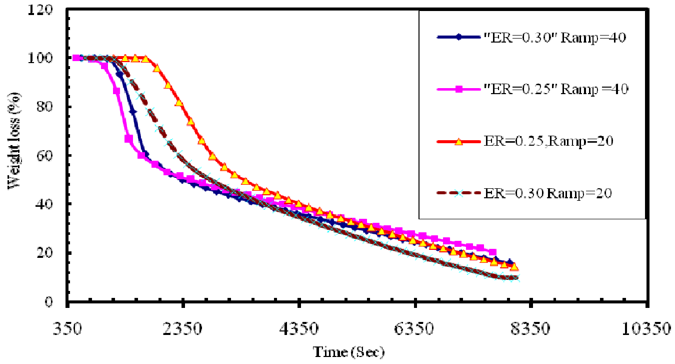

Fig-9 Optimum thermal conversion of coal-bagasse blend 91/9

Similarly, the Kinetic data derived from thermal conversion data of thermo-grams shown in Figures 5 to 7,there was a little variation in kinetics parameters (Ea and A^{-1}) with increasing Equivalence ratio (ER). So with increasing ERs values from 0.25 to 0.35 the values of Ea decreased which in turn promoted the conversion. as shown in Table-3 for coal-bagasse blend (91/9).

4. Conclusion:

This study has investigated the impact of operating parameters (heating rate, feed composition and equivalence ratio) on the conversion and kinetics of co-gasification using TGA in controlled air environment. It was concluded that increased fraction of bagasse and ER value increased the conversion. While in case of heating rate the highest conversion was observed for medium heating rate 20^0C/min due to uniform distribution of heat. Overall highest conversion rate (90.2%) was observed for coal-bagasse blend 91/9 at ER=0.30 and ramp rate 20^0C /min. whereas, the conversion (84.8%) was lower at the ramp rate

(40^0C/min). Perhaps, these are the optimum operating conditions for the TGA in the stated operating environment.
Estimated values of Ea and A^{-1} obtained showed that there was little change in activation energy and a reasonable variation in frequency factor(A) with increasing heat rate and ER values, however it increased with increasing the weight fraction of bagasse in coal. The values determined for the activation energy (Ea), for coal-biomass blend (91/9) were 26.75 ,24.98 and 25.64 kJ/mole for ER values 0.25, 0.30 and 0.35 at constant heating rate of 20^0C/min respectively. At ER=0.30 the value of activation energy (22.98kJ/mole) was found almost acceptable which gave optimum conversion rate.

Finding from the present work shows the optimum values for the thermal conversion of low grade coal-bagasse blends. Additionally, determination of exact impact of these parameters on activation energy is important to understand the thermal decomposition of this low grade coal, bagasse and their blends. This knowledge can provide the guide line to the investigators to design and operate the laboratory scale co-gasifier.

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