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*Research Article*

# Studies on Pyrolysis Kinetic of Newspaper Wastes in a Packed Bed Reactor: Experiments, Modeling, and Product Characterization

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Newspaper waste was pyrolysed in a 50 mm diameter and 640 mm long reactor placed in a packed bed pyrolyser from 573 K to 1173 K in nitrogen atmosphere to obtain char and pyro-oil. The newspaper sample was also pyrolysed in a thermogravimetric analyser (TGA) under the same experimental conditions. The pyrolysis rate of newspaper was observed to decelerate above 673 K. A deactivation model has been attempted to explain this behaviour. The parameters of kinetic model of the reactions have been determined in the temperature range under study. The kinetic rate constants of volatile and char have been determined in the temperature range under study. The activation energies 25.69 KJ/mol, 27.73 KJ/mol, 20.73 KJ/mol and preexponential factors 7.69 $\text{min}^{-1}$, 8.09 $\text{min}^{-1}$, 0.853 $\text{min}^{-1}$ of all products (solid reactant, volatile, and char) have been determined, respectively. A deactivation model for pyrolysis of newspaper has been developed under the present study. The char and pyro-oil obtained at different pyrolysis temperatures have been characterized. The FT-IR analyses of pyro-oil have been done. The higher heating values of both pyro-products have been determined.

## 1. Introduction

Waste management is a big issue nowadays as wastes are being generated in an ever-increasing rate by growing affluent societies. The large amount of nonhomogeneous municipal solid waste has become a tremendous problem for all Indian metropolitan cities. Municipal solid waste (MSW) consists mainly of household and commercial wastes, which are disposed of by, or on behalf of, a local authority. It is composed mainly of paper/cardboard, plastics, glass, metals, textiles, and food/garden waste. Disposal of the massive waste materials poses problems in terms of environmental impact, economic costs and technology implementation. Environment friendly processes must be thoroughly studied for the utilization of the waste materials in view of the increasing demand of energy in this modern era. The recovery of energy from the waste materials may be done through thermochemical processes like combustion, gasification, and pyrolysis. Among all these routes, pyrolysis has been receiving increasing attention in recent years as an acceptable route for waste to energy conversion. The main reason for this is that, in the pyrolysis process production of either char, oil, or gases, the pyrolysis products may be maximized by the adjustment of process condition. Pyrolysis is a thermochemical process in which hydrocarbon rich solid or liquid feed materials are thermally degraded to char, volatile liquid, and noncondensable gaseous component in absence of oxidizing media either air or oxygen. The mechanism of primary pyrolysis of solid and liquid feedstocks is as follows:

$$\text{Pyrolysing solid/liquid} \longrightarrow \text{Char} + \text{Liquid (tar)} + \text{Gas (pyro-gas)} \quad \text{(I)}$$

The usual range of pyrolysis temperature is 573 K to 1273 K. All pyrolysis products have a potential use. For example, char can be burnt as fuel or disposed off (since the heavy metals are fixed inside the carbonaceous matrix), or it can even be upgraded to activated carbon [1]. Gas can be used as fuel [2],

Table 1: Results of proximate and ultimate analyses and higher heating value of newspaper.

| Properties | % (W/W) | Higher heating value (MJ/kg) |
|---|---|---|
| Moisture | 10 | |
| Volatile | 75 | |
| Ash | n.d | |
| Fixed carbon | 15 | |
| Carbon | 59 | 16 |
| Hydrogen | 8.23 | |
| Nitrogen | n.d | |
| Sulfur | 0.31 | |
| Oxygen (by difference) | 32.44 | |

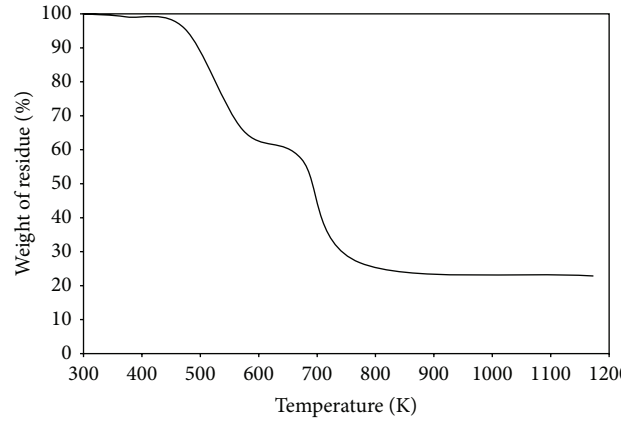


Figure 1: Thermogravimetric analysis graph.

whereas oil can either serve as fuel or as a raw material for chemicals. The yield of either product, namely, char or tar or gas, may be maximized just by adjustment of operating conditions.

In recent years, a few works have been reported in the literature on pyrolysis of waste newspaper or paper mixture [3–9]. Investigation on pyrolysis of mixture of papers with various types of municipal solid wastes has been reported by a few researchers [10–14].

Data solely concerned with pyrolysis of newspaper is however lacking. According to the aforementioned literature, TGA is a common technique to study the thermal decomposition behavior as well as the chemical kinetics of thermal conversion of several biomasses. Newspaper is the principal organic solid waste of Indian metro cities. The composition of newspaper is 62% cellulose, 16% hemicellulose, and 16% lignin. Newspaper has high heating value of about 16 MJ/kg and can be converted through pyrolysis route. As the studies of kinetics of pyrolysis of a feedstock are necessary for proper understanding and application of the process, pyrolysis kinetics of newspaper has been investigated in detail under the present study. Mathematical model incorporating deactivation has also been developed.

## 2. Experimental

*2.1. Materials.* Old newspaper samples were collected from a local residential area for pyrolysis. Table 1 summarizes the results of proximate and ultimate analyses and higher heating value of newspaper.

*2.2. Thermogravimetric Analysis.* Thermogravimetric analysis (Pyris Diamond) of newspaper sample was conducted in the temperature range of 573 K to 1173 K, given in Figure 1. The heating rate was 10°C/min. The flow rate of $N_2$ gas was 150 mL/min.

From this figure, it was clear that the pyrolysis of newspaper started at above 473 K where the weight loss was approximately 20%. Pyrolysis progressed slowly from 473 K to 573 K leading to weight loss up to 35%. Above 573 K, pyrolysis became faster up to 773 K resulting in a weight loss of 74%. This was followed by slow pyrolysis up to 1173 K. The three segments of pyrolysis curve drawn by plotting weight fraction against pyrolysis temperature might signify the pyrolysis of constituent compounds of newspaper, namely, cellulose, hemicelluloses, and lignin. The fractions of cellulose, hemicelluloses, and lignin in newspaper are 40–55%, 25–40%, and 18–20%, respectively [15]. It appears that the hemicelluloses part started pyrolysing at 473 K and the main weight loss occurred for the pyrolysis of hemicellulose portion between 473 K and 573 K. Pyrolysis of cellulose part started at 573 K and proceeded up to 773 K. The last segment of the curve belonging to the temperature zone between 773 K and 1173 K signified the pyrolysis of lignin part of newspaper. Similar observation has been reported by Williams and Besler [16] during their studies on the pyrolysis of wood and its constituent component.

*2.3. Pyrolysis Set-Up.* A 50 mm diameter and 640 mm long cylindrical stainless steel fixed bed pyrolyser was placed horizontally in a tubular furnace (Figure 2).

The pyrolyser was hung by a stainless steel chain attached with a weighing machine for continuous monitoring of the residual mass of solid in the pyrolyser. The furnace temperature varied from 573 K to 1173 K. Once the furnace temperature was raised to a preset value, pyrolyser was inserted into the furnace. Isothermal condition was maintained throughout the entire pyrolysis period. Pyrolysis was carried out for one hour at all temperatures. Experiments were designed to investigate the effects of temperature of pyrolysis on yields of pyro-oil and char and their characteristics. Nitrogen was supplied to the pyrolyser throughout the experiment to sweep the volatiles produced during pyrolysis and to maintain inert atmosphere in the reactor. The volatile product stream along with nitrogen was directed to a water cooled condenser and a series of containers placed in an ice bath. Finally, the gas stream was passed through a silica gel bed and was collected in a gas sampling bottle. The organic part of tar which got dissolved in benzene was extracted in a rotary evaporator and the quantity of pyro-oil was established. The higher heating values of the condensed pyro-oil and char samples,

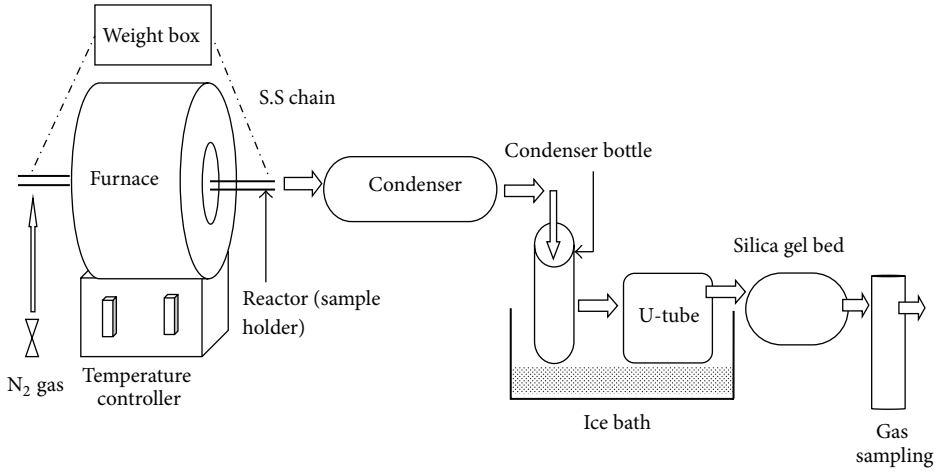


Figure 2: Experimental set up.

obtained as pyrolysis products, were determined using bomb calorimeter.

## 3. Pyrolysis Kinetics

Pyrolysis of newspaper sample proceeds through complex reactions in series, parallel, or combination of both. Under the present study, a parallel reaction model has been attempted to describe pyrolysis kinetics of newspaper. According to this model, pyrolysis of newspaper has been considered to be a homogeneous solid phase reaction and the pyrolysis products have been considered to be char-lumped solid and volatiles made up of tar and gaseous product. The reaction pathway of pyrolysis according to the present model is as follows:

$$\text{Newspaper} \longrightarrow \text{Active complex} \xrightarrow{k_v} \text{Volatile (gas + tar)} \quad \xrightarrow{k_c} \text{Char} \tag{II}$$

The reaction kinetics of volatile and char has been elaborately discussed in the studies of pyrolysis of coconut shell [17], vegetable market waste [18], textile wastes [19], sesame oil cake [20], and mustard press cake [21, 22].

Figure 3 shows the experimental weight fraction profile of residue with respect to time in isothermal conditions at 573 K, 873 K, and 1173 K, respectively. From close observation of the data, it appears that the pyrolysis reactions proceed considerably in the temperature range of 573 K to 1173 K. Below this temperature range, the reactions do not occur at an appreciable rate. From the plots, it is also apparent that at each temperature, a quasiequilibrium of the reaction prevails. The rates of devolatilization reactions decline at temperatures above 673 K. Therefore, the values of frequency factors and activation energies of the reactions of reactant decomposition, volatile formation, and char formation are determined by regression analysis of the rate constant determined in the temperature range of 573 K to 673 K [17–20]. The frequency

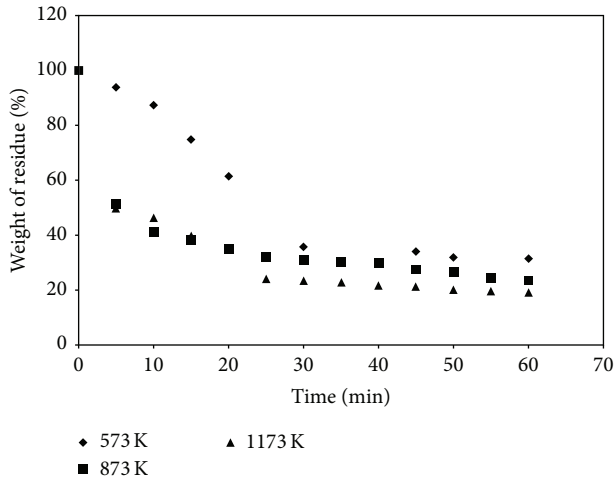


Figure 3: Variation of percentage of weight of residue of newspaper sample with respect to time at different pyrolysis temperature.

factors and activation energies of different reactions are given in Table 2.

In Figure 4, the rate constants $k$, $k_v$, and $k_c$, calculated using activation energies and frequency factors reported in Table 2, have been plotted in the logarithmic scale against reciprocal temperature. All the rate constants calculated from the experimental results in the temperature range of 573 to 1173 K have also been superimposed on the same figure. From the figure, it is apparent that for temperatures higher than 673 K, the actual values of rate constants are far below the predictions of Arrhenius law. The deviation of the pyrolysis rate constants from the Arrhenius law may be due to the deactivation of the solid reactants with temperature. Thus, a deactivation model has been introduced. In the present study, a deactivation model has been developed on the following assumption.

(i) Deactivation occurs with the increase of pyrolysis temperature beyond 673 K.

Table 2: Calculated activation energies and frequency factors as per Arrhenius law.

| Reaction rate constant | Frequency factor ($min^{-1}$) | Activation energy (kJ/mol) |
|---|---|---|
| $k$ | 7.69 | 25.69 |
| $k_v$ | 8.09 | 27.73 |
| $k_c$ | 0.853 | 20.73 |

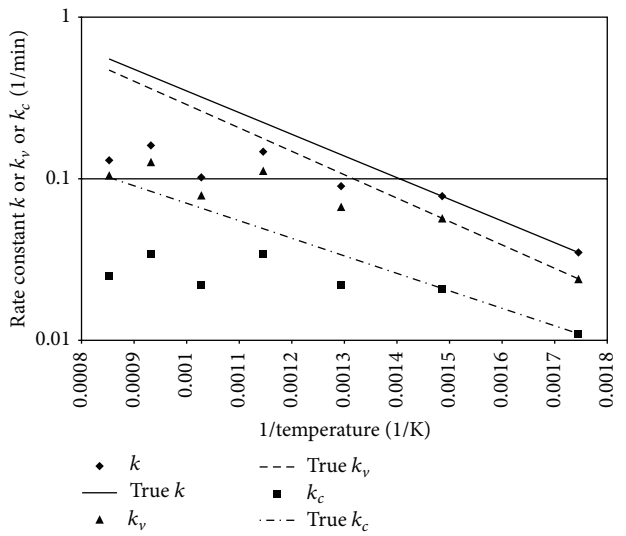


Figure 4: Simulated true $k$, true $k_v$, true $k_c$, and experimental $k$, $k_v$, and $k_c$ values of rate constant for weight loss of newspaper, volatile formation, and char formation in logarithmic scale against reciprocal temperature.

(ii) The apparent reaction rate constants may be written in the following form:

$$k_{ap} = ak_0 \exp\left(\frac{-E}{RT}\right),$$
$$k_{vap} = a_v k_{v0} \exp\left(\frac{-E_v}{RT}\right), \quad (1)$$
$$k_{cap} = a_c k_{c0} \exp\left(\frac{-E_c}{RT}\right),$$

where $a$, $a_v$, and $a_c$ may be defined as the activities of the solid towards corresponding reactions.

(iii) The values of activities $a$, $a_v$, and $a_c$ are unity at 673 K and they decrease to the minimum of zero as the temperature increases to 1273 K.

The normalized temperature parameter has been defined as

$$\theta = \frac{T - T(a=1)}{T(a=0) - T(a=1)}. \quad (2)$$

Here, $T(a = 1) = 673$ K and $T(a = 0) = 1273$ K.

Table 3: Values of constant for deactivation model.

| Model parameters | Values | Correlation coefficient |
|---|---|---|
| $n$ | −0.067 | 0.897 |
| $n_v$ | −0.089 | 0.901 |
| $n_c$ | −0.046 | 0.874 |

*3.1. Deactivation Model.* In this model, the rates of deactivation are considered as function of activities themselves with exponents other than unity:

$$\frac{-da}{d\theta} = \beta a^n \quad (\text{where } n \neq 1),$$
$$\frac{-da_v}{d\theta} = \beta_v a_v^{n_v} \quad (\text{where } n_v \neq 1), \quad (3)$$
$$\frac{-da_c}{d\theta} = \beta_c a_c^{n_c} \quad (\text{where } n_c \neq 1).$$

The boundary conditions are as follows:

$$a = a_v = a_c = 0 \quad \text{at } \theta = 1,$$
$$a = a_v = a_c = 1 \quad \text{at } \theta = 0. \quad (4)$$

Applying the boundary conditions represented by (4), the solutions of the differential equations may be expressed as

$$a = \exp\left[\frac{\ln(1-\theta)}{1-n}\right],$$
$$a_v = \exp\left[\frac{\ln(1-\theta)}{1-n_v}\right], \quad (5)$$
$$a_c = \exp\left[\frac{\ln(1-\theta)}{1-n_c}\right].$$

Therefore, the rate expression may be represented as follows:

$$k_{ap} = k_0 \exp\left(\frac{-E}{RT} + \frac{\ln(1-\theta)}{1-n}\right),$$
$$k_{vap} = k_{v0} \exp\left(\frac{-E_v}{RT} + \frac{\ln(1-\theta)}{1-n_v}\right), \quad (6)$$
$$k_{cap} = k_{c0} \exp\left(\frac{-E_c}{RT} + \frac{\ln(1-\theta)}{1-n_c}\right).$$

Values of different parameter of deactivation model have been determined using nonlinear regression analysis. The values of parameters are given in Table 3.

In Figure 5, logarithms of simulated values of $k$, $k_v$, and $k_c$, predicted by Arrhenius law using data of 573 K and 673 K as well as those predicted by deactivation model in the temperature zone beyond 673 K, have been plotted against inverse of temperature.

Comparison with experimental data suggests that for $k$, $k_v$, and $k_c$, the deactivation model can explain the reality, except at 973 K. The experimental findings at 973 K indicate comparatively low reaction rate. Actually, other than deactivation of single pyrolysis component due to tar clogging

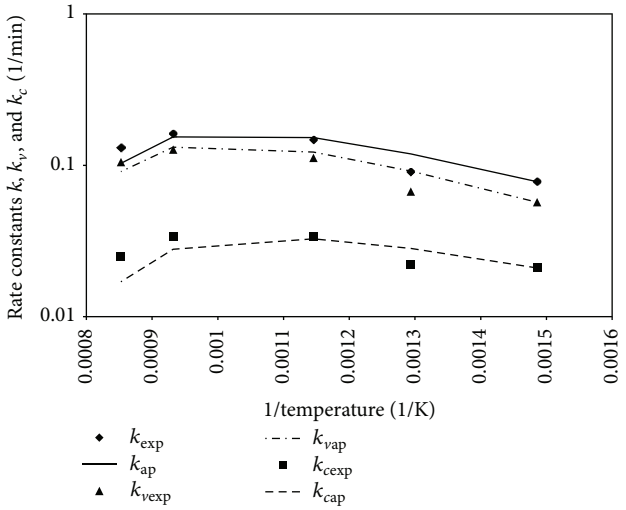


FIGURE 5: Comparison of simulated rate constants $k_{app}$, $k_{vapp}$, and $k_{capp}$ as per deactivation model with the experimental results $k_{exp}$, $k_{v\,exp}$. $k_{c\,exp}$ for weight loss of newspaper, volatile, and char formation, respectively.

of active sites, conformational changes, and so forth, as observed by Bandyopadhyay et al. [17], Ray et al. [18], Sarkar and Chowdhury [19], use of different types of woods, both soft and hard, as the source of Indian newspaper may lead to abnormal pyrolysis behaviour.

## 4. Results and Discussions

*4.1. Effects of Pyrolysis Temperature on Product Yields.* After the completion of pyrolysis of newspaper, the solid residue part was collected from the reactor. The unreacted newspaper and the left char yield were determined. While the condensable part of volatile was considered as a tar yield, the organic part of tar, soluble in benzene, was considered as pyro-oil. The gas yield was calculated by subtracting the amount of tar from volatile yield. These yields of char, tar, and gases in relation to reactor temperature are shown in Figure 6.

The char yield decreased with a rise in pyrolysis temperature from 32 wt% at 573 K to 20 wt% at 1173 K. On the other hand, the yield of tar increased to 43 wt% at 773 K and then it decreased to 12 wt% at 1173 K. Figure 6 also shows that while gas yield increased gradually from 573 K to 873 K, it increased appreciably as the pyrolysis temperature increased from 973 K to 1173 K.

Appearance of a maximum in the trend of yield of tar against temperature may be due to the commencement of further cracking of tar molecules to lower gaseous molecules at higher temperatures. Under the present experimental conditions, the results showed a clear influence of temperature on the fractional yields of char and volatiles. The decrease in the char yield with pyrolysis temperature is to be attributed to an increasing devolatilization of the solid hydrocarbons in the char. Partial gasification of the carbonaceous residue is also possible [23].

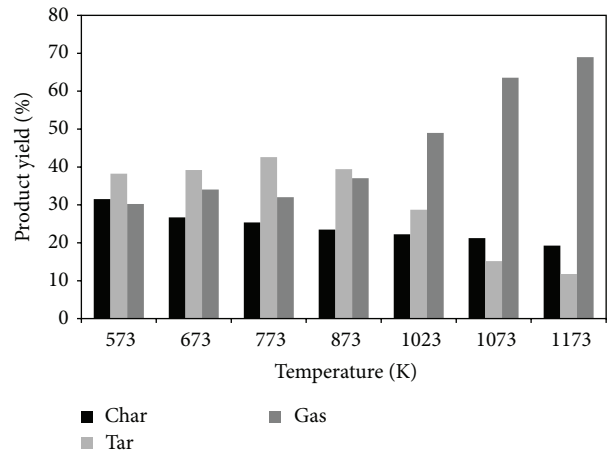


FIGURE 6: Percentage of product yields as char, tar, gas, and unreacted reactant in different pyrolysis temperature (K).

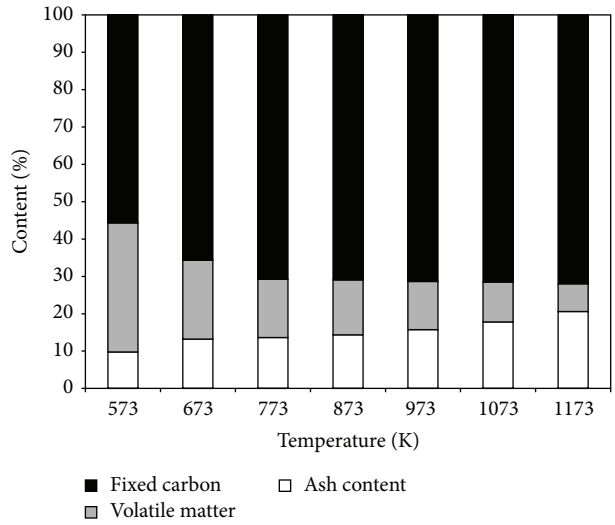


FIGURE 7: Percentage of volatile, ash, and fixed carbon present in char obtained at different pyrolysis temperatures.

*4.2. Products Characterization*

*4.2.1. Proximate Analyses of Char.* Proximate analyses of char obtained at different pyrotemperatures from 573 K to 1173 K are shown in Figure 7.

Proximate analysis of char sample has been done to measure the fixed carbon that is present in it. From this figure, it may be inferred that volatile content of char gradually decreases with temperature, while contents of fixed carbon and ash show increasing pattern with temperature. The fixed carbon and ash content of char product increased with temperature from 70.72 and 13.61 wt% at 773 K to 71.97 and 20.58 wt% at 1173 K, respectively. While volatile matter of char product decreased from 15.66 to 7.44 wt% at 773 to 1173 K.

*4.2.2. Chemical Characterization of Product Yield.* The empirical formulas of pyroproducts obtained at different pyrolysis

Table 4: Empirical formula of char and pyro-oil.

| Temperature (K) | Char yield | Tar yield |
|---|---|---|
| 573 | $C_3H_2O$ | $CH_2O$ |
| 673 | $C_3HO$ | CHO |
| 773 | $C_6H_3O$ | $CHO_2$ |
| 873 | $C_4HO$ | $C_3HO_3$ |
| 973 | $C_5HO$ | $C_3HO_3$ |
| 1073 | $C_6HO$ | $C_3HO_3$ |
| 1173 | $C_7HO$ | $C_2HO_4$ |

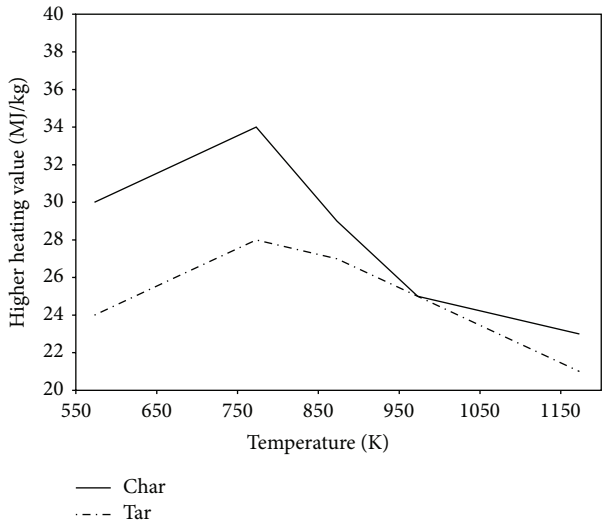


Figure 8: Pattern of higher heating values of pyro-oil and char at different pyrolysis temperature.

temperatures are listed in Table 4. It was clear from the table that the char products became carbon rich with the rise of pyrolysis temperature. The ratio of H/C decreased with the higher temperature. On the other hand, the pyro-oil became highly oxygenated with the rise of temperature. The ratio of H/C decreased with the higher temperature. At initial temperatures, namely, 573, 673, and 773 K, the H/C ratios of pyro-oil were 14, 9, and 5, respectively. Beyond this temperature, the ratio of H/C of pyro-oil was similar to alkenes/cycloalkanes (~2).

*4.2.3. Effects of Pyrolysis Temperature on Higher Heating Values of Product Yield.* The higher heating values of char and tar yield of different pyrolysis temperature are determined using the bomb calorimeter given in Figure 8. The higher heating value of char increases gradually from 30 to 34 MJ/Kg as the temperature increases from 573 K to 773 K. Beyond 773 K, the higher heating value decreases from 34 to 23 MJ/Kg as the temperature is changed from 873 K to 1173 K. Pattern of temperature dependence of higher heating value of tar is also similar to that of char. This may be justified by the fact that fraction of carbon in char increases as the temperature increases up to 773 K, beyond which char further participates in heterogeneous reactions with gaseous product. In case of tar, fraction of volatile components increases with the temperature, resulting in the increase of higher heating value up to 773 K. At temperatures above 773 K, secondary cracking of tar takes place causing decrease in higher heating value of tar.

Table 5: Main atomic groups and structure of pyro-oil.

| Wavenumber ($cm^{-1}$) | Infrared spectrum | Atomic groups and structures |
|---|---|---|
| 3200–3700 | O–H stretching | Hydroxyl |
| 2800–3000 | C–H stretching | Aliphatic structures |
| 1650–1770 | C=O stretching | Carbonyl |
| 1610–1680 | C=C stretching | Oliefinic structures |
| 1450–1600 | C=C stretching | Aromatic structures |
| 1420–1480 | C–H bending | Aliphatic structures |
| 1360–1430 | O–H and C–H bending | Hydroxyl, acid, phenol, olefins, and methyl |
| 1200–1300 | C–O stretching | Unsaturated ethers |
| 1000–1200 | C–H out-of-plane blending | Aromatic structures |

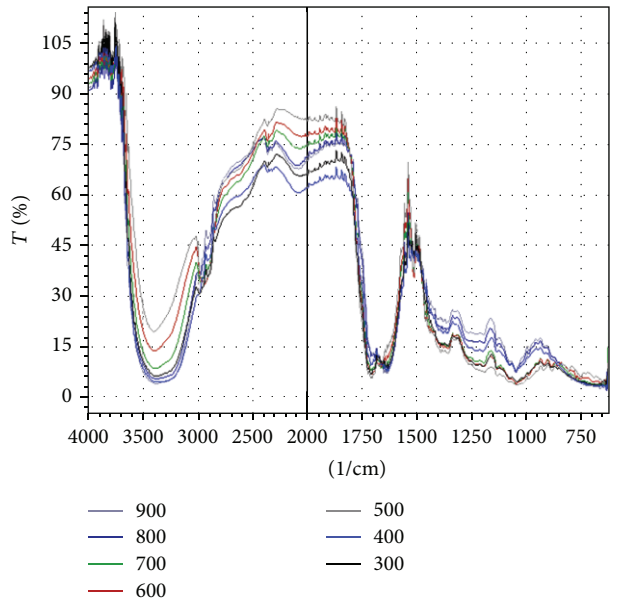


Figure 9: FT-IR spectrum of pyro-oil obtained at different pyrolysis temperatures.

*4.2.4. FT-IR Analyses of Pyro-Oil.* The FT-IR spectra of pyro-oils obtained at different temperatures from pyrolysis of newspaper waste feedstock are provided in Figure 9. Band assignments of IR spectrum of pyro-oil, which are summarized in Table 5, indicate that the pyro-oil contains a number of atomic groupings and structures.

It appeared from Figure 9 that the intensity of spectrum of pyro-oil has changed with the increase of temperature.

At higher temperature, the hydrogen bonded OH stretching decreased due to the loss of phenolic or alcoholic groups of the pyro-oil [21, 24].

## 5. Conclusion

In the present investigation, pyrolysis of newspaper has been studied in the temperature range of 573 K to 1173 K. FT-IR analyses of pyro-oil obtained at different reaction temperatures have been done. The effects of pyrolysis temperature on higher heating value and ratio of H/C of pyro-oil and char have been discussed. These properties of the pyrolysis products can be used as fundamental data for the design of a pyrolysis process for biomass wastes. The system has been mathematically modeled in a deterministic way incorporating the deactivation phenomenon. A deactivation model representing the deactivation rate as a nonlinear function of activities has been found to be successful to explain the reaction engineering behaviour of the system.

## Nomenclature

$a$: Activity of solid
$E$: Activation energy (kJ/mol)
$k$: Rate constant ($min^{-1}$)
$k_{ap}$: Apparent reaction rate constant (k/min)
$R$: Gas constant (kJ/mol/K)
$T$: Temperature (K)
$t$: Time (min)
$\beta$: Deactivation rate constant
$\theta$: Dimensionless temperature parameter
$v$: Volatiles
$c$: Char
0: Initial condition.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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