



## Energy and Useful Products from Waste Using Pyrolysis: A State-of-the-Art Review

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### ABSTRACT

*Waste to energy is an answer to the growing demand for fossil fuels. Pyrolysis plays an important role in waste to energy technologies. The product obtained from pyrolysis depends on various physical parameters that influence the reaction mechanism. Pyrolysis reactions can be classified onto flash, fast and slow pyrolysis depending on temperature and heating rate. This paper discusses the effect of parameters like temperature, heating rate, residence time and particle size for each of the above mentioned pyrolysis types. The product distribution for each type of pyrolysis is discussed. Moreover many feed materials are being researched to find out the best possible feed in terms of product yield. The paper brings out a list of various feed tried out in recent times and also the yield obtained from each of the feed material.*

### INTRODUCTION

Waste is an inevitable consequence of numerous human activities. The average amount of waste generated, in India, by an individual ranges from 100g in rural areas to 500g in urban areas (Shaleen & Pandey, 2001). Waste can be categorized into solid, liquid and gas. Solid waste management has gained a lot of importance these days and ways of handling and disposing them safely are being researched. In the past two decades many researchers have worked on solid waste management. Many efforts were towards reusing and recycling the waste. When it comes to reusing organic waste, the main method is to convert the waste into energy. Waste to energy is an upcoming field that aims in converting organic waste into fuel feedstocks by methods like incineration, gasification or pyrolysis. Each of the methods is widely researched and finds application as the situation demands. The main objective of this paper is to review the recent trends in pyrolysis of solid waste disposal. Solid waste has a major portion of it as organic matter. Organic matter has absorbed carbon dioxide as a consequence of photosynthesis. Upon thermal destruction in presence of excess air, the absorbed carbon dioxide is liberated back. Under controlled thermal destruction, we can obtain valuable organic products like CO, H<sub>2</sub>, CH<sub>4</sub> etc. The suitability of a biomass fuel for the production of energy in a combustion process is based on the inherent energy content of the elemental constituents, the density, and the moisture content of the material.

## **PYROLYSIS**

Pyrolysis is the thermal decomposition of complex organic matter to simpler molecules, which can be used as a feedstock. The main products obtained by pyrolysis are char, oil and gas. Char can be used for combustion or used as activated carbon. Bio-oil can be used directly by upgrading or added to petroleum products and used. The gas obtained can be directly used for combustion. The main constituent of biomass is cellulose, hemi cellulose and lignin. Each of this constituent is different in structure, elemental composition and thermal behavior. The pyrolysis behavior of biomass is the sum of pyrolysis behavior of individual constituent's viz. cellulose, hemi cellulose and lignin. The thermal behavior of each of these constituents is affected by a set of parameters which govern the conversion, product yield and distribution. The main parameters affecting the pyrolysis behavior are temperature, heating rate, solid residence time, volatile residence time, particle size and density of the particles (Babu & Chaurasia, 2004a, 2004b). Pyrolysis is classified into three types: (1) Flash (2) Fast Pyrolysis (3) Slow Pyrolysis, according to the temperature, heating rate and residence time. The product obtained depends mainly of the type of pyrolysis.

The main component of any organic matter is cellulose. Cellulose is again the important constituent that contributes to maximum tar and volatile compounds. The pyrolysis behavior of any biomass is the sum of decomposition characteristics of cellulose, hemi cellulose, lignin and their interactions. Thermo gravimetric studies have been conducted to find the rate of decomposition of each of the above constituents by Besler & Williams (1996). Cellulose decomposes between temperature 325<sup>0</sup>C and 400<sup>0</sup>C, hemi cellulose decomposes between 250<sup>0</sup>C and 350<sup>0</sup>C and lignin starts at 200<sup>0</sup>C and continues till 700<sup>0</sup>C. It is also important to observe that there is a lateral shift in the temperatures of decomposition as the rate of heating is increased. The results show that the primary decomposition of wood takes place between 250<sup>0</sup>C and 450<sup>0</sup>C. It would be interesting to find out the product composition obtained by pyrolyzing different biomass feed under different conditions. The following text is divided into flash & fast pyrolysis and slow pyrolysis. Under each section, the pyrolysis behavior, parametric study and the product yield are described.

### **Flash & Fast Pyrolysis**

Early studies on decomposition (Rapid Pyrolysis) of cellulose were carried out by Peters et al. (1985). The effect of heating rate, temperature and solids residence time was studied. It was observed that 95% of cellulose decomposition takes place between 500<sup>0</sup>C and 750<sup>0</sup>C, heating rate being 1000<sup>0</sup>C/s. Beyond 750<sup>0</sup>C, the yield of char decreases. It is also important to note that tar yield goes through a maximum of 83% at 400<sup>0</sup>C and falls to 49% at 1000<sup>0</sup>C. This is because of secondary cracking of tar at higher temperatures. Beyond 750<sup>0</sup>C, the yield of char decreases from 6% to 3%. As the temperature increases beyond 900<sup>0</sup>C the yield of char again increases to 4%. This indicates that beyond 900<sup>0</sup>C re-polymerization reactions take place and results in increase of char weight. The result of cracking of cellulose between 300–600<sup>0</sup>C shows mostly oxygenated compounds like acetaldehyde. As the temperature goes beyond 600<sup>0</sup>C the yield of hydrocarbon gases increase indicating secondary cracking of tar. Moreover as the solid residence time increases, the amount of tar formed increases with temperature up to 800<sup>0</sup>C. Beyond this temperature, the solid residence time does not have any effect. Up to a temperature of 750<sup>0</sup>C and at short residence times, increase in heating rate decreases the amount of tar formed. This is because at high heating rate and short residence times, the de-volatilization of products is not complete and the products formed are swept away as soon as they are formed.

Quantitative information of pyrolysis on primary decomposition and volatiles secondary reactions is important. Rapid pyrolysis of sweet gum wood was done by Peters et al. (1995). The results show that decomposition of wood starts at 325<sup>0</sup>C. Sample weight loss increases with temperature and 93% of the sample weight is lost at 675<sup>0</sup>C. Most of sample weight loss takes place between 425<sup>0</sup>C & 625<sup>0</sup>C. Tar yield increases to a maximum of 55% around 700<sup>0</sup>C, then starts decreasing and stabilizes at 1027<sup>0</sup>C for a value of 46%. The decrease is attributed to secondary cracking of tar. Yield of gases increases steeply and stabilizes at 1027<sup>0</sup>C where tar reaches an asymptotic value. Increasing the temperature beyond 1027<sup>0</sup>C does not affect the yield of tar. This indicates that pyrolysis produces two type of tars, one thermally labile which decomposes completely and the other not so labile to temperature. CO and CH<sub>4</sub> evolution begins at 527<sup>0</sup>C and increases up to 927<sup>0</sup>C. This increase is in continuation to the decrease in tar yield indicating that CO and CH<sub>4</sub> are products of secondary cracking of tar. Similar trends are observed for ethylene, ethane and propylene indicating these too are products of secondary cracking of tar. In contrary CO<sub>2</sub> starts at 327<sup>0</sup>C and levels off at 680<sup>0</sup>C indicating that it is a product of direct decomposition of wood. Similarly chemical water, methanol, acetaldehyde etc are also products of primary cracking of wood.

This study proves that beyond a certain temperature the bio oil or the tarry fraction cracks to yield gaseous products. The kinetics of the secondary cracking was studied by Peters et al. (1989). He used a reactor in series to study the secondary cracking of primary pyrolysis products of woody biomass. The first of the reactor in series was maintained at a low heating rate and a low temperature of 450<sup>0</sup>C. This facilitated generation of tar. The tar was swept immediately as and when formed into the second reactor which was maintained at different temperatures between 600<sup>0</sup>C-800<sup>0</sup>C. The results were as follows. Primary pyrolysis at 450<sup>0</sup>C produces more than 50% (wt%) of tar, 18% of char and rest gas. Carbon dioxide is the main gaseous product followed by water and carbon monoxide. When the primary pyrolysis products are fed into the second reactor the tar started cracking and is reduced to 6 wt% at 800<sup>0</sup>C. Tar mainly cracks down to CO<sub>2</sub>, CO, CH<sub>4</sub> and low concentrations of other hydrocarbon gases like methane, ethane and propane.

The flash pyrolysis of Euphorbia Characias (Spurge) bagasse was carried out by Conti et al. (1994). The reactor was a fluidized bed reactor with sand particles and nitrogen gas to fluidize the bed. The heating rate, as a result of fluidization, has been claimed to be 10<sup>4</sup>°C/s and the residence time is 0.5s. Pyrolysis has been carried out at different temperatures between 400<sup>0</sup>C and 750<sup>0</sup>C. The observations are that between a temperature of 400<sup>0</sup>C and 550<sup>0</sup>C the oil yield continues to increase steeply and remains constant in the range 520<sup>0</sup>C – 550<sup>0</sup>C. Above 550<sup>0</sup>C the yield of oil starts decreasing and the gas yield increases. The product yield obtained from the above species is 45% at 520<sup>0</sup>C. Flash pyrolysis of mixed wood waste yields a maximum of 68% pyrolytic oil at 550<sup>0</sup>C ( Patrick and Williams, 1996). It has also been observed that concentration of poly aromatic hydrocarbons which are carcinogenic and mutagenic is very less in the range of less than 5 ppm. The concentration of PAH compounds increase as the temperature increase. But oils obtained from flash pyrolysis at a temperature of 550<sup>0</sup>C contain negligible amount of harmful PAH's. The oil obtained was homogeneous liquid with low viscosity.

The fast pyrolysis of Pterocarpus indicus (Narra) in a fluidized bed reactor was studied by Zhongyang et al. (2004). The parameters studied were temperature, particle size, feed rate and volatile residence time. A temperature of 500<sup>0</sup>C–550<sup>0</sup>C yields the maximum amount of bio-oil. At higher temperature the yield of gases increases. Meanwhile, particle size had no

obvious effect on products distribution when particle size was less than 1 mm indicating that resistance mass and heat transfer is negligible. It has also been observed that it is more effective to increase the feed rate than increasing reactor height to shorten volatile residence, so as to restrain secondary cracking. This is because shortening reactor height only acted on the dilute phase area which is at low temperature. However, with feed rate increase, the volatile residence time in the dense phase area which is at higher temperature would be cut significantly. The yields of bio-oil is observed to be around 56% for *Pterocarpus indicus* at 550<sup>0</sup>C. The same experiments were carried out with other biomass feedstock's like *Cunninghamia lanceolata* (China Fir), *Fraxinus mandshurica* (Japanese ash), and Rice Straw. *Pterocarpus indicus* faired better than all other feedstock's in terms of oil yield and low water content. In this study it has been said that more the ash content in the biomass species, less would be the yield of bio oil. It has been suggested that if the biomass is deashed the yield of products will increase.

The effect of deashing biomass species was studied by Piyali et al. (2004) Sugarcane bagasse was deashed by extraction with three different solvents – water, HCL and HF. The deashing with HF was found to be the most efficient of the three. The treated bagasse was then pyrolyzed in a pyrolyser at a maximum temperature of 500<sup>0</sup>C. It was observed that whereas the amount of total liquids (Oil + Aqueous) decreased, the amount of bio oil obtained increased in comparison to the untreated biomass. It shows that deashing shifts the equilibrium to producing more bio-oil and less of other liquids. The increase in bio-oil was to the tune of 10wt% for bagasse deashed with HF. The oil yield increases only if the percentage of cellulose and lignin increase in the feed. This shows that hemi cellulose & extractives does not contribute majorly to bio-oil. It has also been reported that the moisture content in the bio oil obtained from the deashed bagasse is less than that obtained from untreated bagasse. Moreover, higher the potassium, zinc and lignin content lesser is the amount of volatiles released and more the amount of char formed.

The effect of sweep gas flow rate was studied by Hasan (2002) in a fixed bed tubular reactor. Two sweep gas flow rates of 25 cc/min and 100 cc/min were employed. The feed was sunflower bagasse of diameter 0.85mm–1.8mm. The temperature range studied was between 400<sup>0</sup>C–750<sup>0</sup>C and heating rate employed was 300<sup>0</sup>C/min. Nitrogen was used to sweep the pyrolysis products. The results showed that the product yield was significantly influenced by the nitrogen flow rate. For a flow rate of 25 cc/min, the bio-oil yield increased from 29.64 at 400<sup>0</sup>C to 46.62 at 550<sup>0</sup>C. For a flow rate of 100 cc/min the bio-oil yield was marginally less at 46.43%, at 550<sup>0</sup>C. The results show that there is a slight decrease in oil yield at higher gas flow rate. However oil yield is maximum at around 550<sup>0</sup>C for all sweep gas flow rates.

In a similar study the fast pyrolysis of walnut was carried out by Ozlem et al (2004a). The effect of Temperature, heating rate and sweep gas flow rate was studied. Oil yield increased as the temperature increased from 350<sup>0</sup>C to 550<sup>0</sup>C and at temperature thereafter a decreasing trend is observed. It is consistent with the observation of Hasan (2002) in his study with sunflower bagasse. The heating rate had a significant effect on the oil yield. At a heating rate of 100<sup>0</sup>C /min and a max temperature of 550<sup>0</sup>C the oil yield was 27-28%, whereas on increasing the heating rate to 300<sup>0</sup>C /min the oil yield increased to 30-31%. Beyond this heating rate the yield of oil decreased. To establish the effect of sweep gas, a wide range of flow rates from 50 cc/min to 400 cc/min was used. The oil yield increased till 100 cc/min, but beyond 100 cc/min there was no significant effect of sweep gas. An opposite trend is observed for gas yield. Gas yield is less in the temperature range 350<sup>0</sup>C–500<sup>0</sup>C. It starts increasing steeply at around 700<sup>0</sup>C. Heating rate does not have a significant effect below

400<sup>0</sup>C, but beyond 400<sup>0</sup>C, increasing the heating rate affect the ratio of oil to gas production (Hasan, 2002; Ozlem et al., 2004a).

### **Slow Pyrolysis**

Slow pyrolysis of biomass is carried out at a heating rate of less than 100<sup>0</sup>C /min. The reaction mechanism and products formed are quite different from that of flash and fast pyrolysis. Many valuable products are formed during slow pyrolysis. The main product formed during slow pyrolysis is char and bio-oil. Char can be used as feed stock for combustion applications. Along with oil and char, at higher temperatures we get gas as a result of secondary cracking. Since we get a wide range of products a lot of interest is there these days for slow pyrolysis. The effect of temperature, heating rate and residence time are important for slow pyrolysis.

The effect of temperature and heating rate on slow pyrolysis of biomass was studied by Besler & Williams (1996). Heating rates between 5<sup>0</sup>C/min and 80<sup>0</sup>C/min was employed and temperatures between 300<sup>0</sup>C and 750<sup>0</sup>C were studied. Volatiles formed were swept with nitrogen as carrier gas and products were rapidly quenched. It was observed that as the heating rate increased the char formation decreased. Major gaseous products between 200<sup>0</sup>C and 400<sup>0</sup>C were CO and CO<sub>2</sub> whereas at higher temperature low concentration of hydrocarbon gases was also observed. As the heating rate was increased the amount of gases namely CO, CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub> etc increased. It shows that higher heating rate favors release of hydrocarbon gases. Similarly oil and aqueous yield increased with increase in heating rate. Temperature has a significant effect on product yield. Higher the temperature more is the product yield and less char yield.

Detailed study on parametric effect of slow pyrolysis was carried out by Ozlem et al. (2004b) in two reactors viz Heinz's retort and a fixed bed reactor. The feed was Rapeseed. In the Heinz's reactor the experiments were carried out in the absence of sweep gas to establish the effect of temperature. As the temperature was increased to 700<sup>0</sup>C at a rate of 30<sup>0</sup>C /min the yield of char decreased and total conversion increased. A maximum oil yield of 41.4% was obtained at 550<sup>0</sup>C. The yield of gas and oil is maximum for a particle size of 0.85<d<sub>p</sub><1.25. On introducing sweep gas (nitrogen) at a flow rate of 100 cc/min the yield of oil increased to 51.7%. Further increasing the flow rate to 200 cc/min had no significant effect. But as the flow rate of sweep gas is increased beyond 200 cc/min the yield of oil starts decreasing.

The experiments were also carried out in a well swept fixed bed reactor for the same feed. A sweep gas flow rate of 100 cc/min–200 cc/min, particle size of +0.6–1.25mm and heating rate of 300<sup>0</sup>C/min was employed. The oil yield obtained was 68% for a particle size of 0.85<d<sub>p</sub><1.25 and temperature 550<sup>0</sup>C. It has to be noticed that for all similar conditions but heating rate between Heinz's reactor and fixed bed reactor, the oil yield and conversion varies. Heinz reactor was slow pyrolysis whereas fixed bed experiments were fast pyrolysis. The difference in yield shows that transport properties (mass and heat) are predominant in slow pyrolysis even in presence of sweep gas.

To understand the relationship between the composition of the raw material and released product a TGA study of Miscanthus Giganteus (Giant Chinese Silver Grass) and pine wood was carried out (Jong et al., 2003) at various heating rates between 10<sup>0</sup>C /min to 100<sup>0</sup>C/min. The maximum temperature employed was 900<sup>0</sup>C. It was observed that grass gives more char than wood. Moreover the yield of tar for wood was almost double as that of silver grass. This shows that volatiles yield of wood is greater then herbaceous material like straw and stalk.

Pyrolysis of *Euphorbia macroclada* (branched spurge) was studied by Feride & Hasan (2004) in a tubular fixed bed reactor. The temperatures employed were between 400<sup>0</sup>C and 700<sup>0</sup>C and the heating rate was 7<sup>0</sup>C /min and 40<sup>0</sup>C /min. It was found that the maximum yield of bio-oil was obtained at 550<sup>0</sup>C and increasing the temperature yields more gaseous products. The maximum amount of bio-oil obtained at 550<sup>0</sup>C was 24.1wt % at 7<sup>0</sup>C/min. The gas yield obtained was found to be 30.8 wt% at 400<sup>0</sup>C and 37.8 wt% at 700<sup>0</sup>C for the heating rate of 7<sup>0</sup>C /min. The char yield obtained for a heating rate of 40<sup>0</sup>C /min decreased from 29.7 to 24.1 wt% when the temperature was increased from 400<sup>0</sup>C to 700<sup>0</sup>C. Gas yield was 34.6 wt% at 400<sup>0</sup>C and 42.8 wt% at 700<sup>0</sup>C for the heating rate of 40<sup>0</sup>C /min. The overall conversion was 2-3% more at heating rate of 40<sup>0</sup>C /min than at 7<sup>0</sup>C /min. *Euphorbia macroclada* has less bulk density and hence the mass transfer restrictions were less pronounced than for coals.

Batch pyrolysis of municipal solid waste was studied by Williams & Besler (1992). The temperature of interest was between 300<sup>0</sup>C and 700<sup>0</sup>C. Heating rate employed was between 5<sup>0</sup>C /min and 80<sup>0</sup>C /min. The product yield at 720<sup>0</sup>C has been found to be 53% oil, 17% gas and 30% char. The yield of products suggest that MSW is a good candidate for pyrolysis. However no mention is made about the harmful gases that may evolve along with the main gaseous products.

Understanding the mechanism of pyrolysis of MSW's is important because MSW's are heterogeneous and there might be various reactions like re-polymerization or interaction between products that may alter the product yield and composition. To analyze the possibility of interactions between species, co-pyrolysis of PVC with cellulose as a model for municipal solid waste was studied by Peters et al. (1995). Characterization of MSW is difficult but roughly consists 65% organic material (cellulose, lignin etc), 15% polymeric material and 20% inorganic materials. To understand the behavior of MSW, 100% PVC, mixture of PVC and straw in various weight fraction and two samples of municipal solid waste was pyrolyzed. The results show that weight loss characteristics of PVC were different from straw. Straw showed a much higher rate of weight loss at high temperature. A mixture of PVC and straw showed a much different behavior than the parent material alone. This acts as evidence that there is significant interaction between polymeric and cellulosic material and hence the weight loss characteristic differs. Moreover the Chars obtained from PVC was found very less reactive in comparison to the char obtained from Straw.

The summary of pyrolysis carried out at different conditions, feeds used and the products obtained is summarized based on the above study. It can be observed from table 1 that the pyrolysis of pure cellulose yields more than oil and gas to the tune of 90% of original biomass sample. Also it can be observed that char yield at 1000<sup>0</sup>C was more than that at 750<sup>0</sup>C indicating polymerization reactions take place only above 750<sup>0</sup>C. Moreover from the table it can be observed that flash pyrolysis at high temperatures produces high gas yield and low char yield.

## CONCLUSIONS

1. Pyrolysis of biomass is the best candidate to meet the energy requirements of the future. To make the process more efficient and affordable the parametric effects affecting the reaction has to be well understood. The following are few results that can be summarized based on the study.
2. Under rapid pyrolysis, Cellulose and sweet gum wood loses 95% weight between 500<sup>0</sup>C and 750<sup>0</sup>C. Whereas for the case of slow pyrolysis the decomposition takes place between

450°C and 700°C. There is a lateral shift in temperature of primary decomposition with heating rate.

3. Tar yield (gas + oil) is 83% for cellulose whereas it is only 53% for sweet gum wood, clearly indicating that more the cellulose content more the volatiles yield. The same observation can be made with other products as well that are listed in the table.
4. Ash makes a major difference in the products yield. It is clear that deashing the feed increases volatiles yield.
5. Oil yield is observed to be maximum in the range of 500°C–550°C. Above this temperature the yield of gas increases. Oil contains mainly oxygenated aromatic substances.
6. Rapeseed is the most favorable feed that gives high yield of oil, because of its high cellulose content. Other feeds like sunflower bagasse, Pterocarpus indicus also give high yield of bio-oil and can act as good candidate for bio-oil production.
7. The mode of pyrolysis determines the product yield and composition. Heating rate along with residence time influences the product yield and composition of the products.
8. Fluidized bed pyrolysis yields more volatiles as a result of high heating rate obtained in comparison to batch pyrolysis or fixed bed pyrolysis.

**Table-1: Comparative between of various biomass Feed.**

S. No	Pyrolysis Type	Feed	Temp °C	Heating rate °C/min	Particle Size (cm)	Gas yield wt%	Oil Yield wt%	Char yield wt%	HHV of oil MJ/kg
1	Flash pyrolysis	Cellulose	1000	6 x 10 <sup>4</sup>	2x6x0.01	46.97	49.12 (Tar)	3.91	-
2	Flash pyrolysis	Cellulose	750	6 x 10 <sup>4</sup>	2x6x0.01	36.76	59.92 (Tar)	3.32	-
3	Fast pyrolysis	Sunflower Bagasse	550	300	0.085-0.18	13	46.62	34	32.9
4	Fast	Walnut Shell	500	300	0.06–0.125	15.6	31.1	27.5	31.4
5	Fast	Pterocarpus indicus	500	-	<0.1	28	55.7	16	-
6	Fast	Rapeseed	550	300	0.06-0.125	13	68	13	39.4
7	Slow pyrolysis	Misacantus Giganteous	900	100	<0.05	-	81.9 (Tar)	18.1	-
8	Slow Pyrolysis	Deashed Sugarcane Bagasse	500	-	-	14.7	47.9	12.4	23.2
9	Slow Pyrolysis	Euphorbia macroclad	550	7	0.047	18	24.1	26	34.5

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