

Acceptance Letter

Date: Sep 2016
Paper ID: ncCD2

Dear Author(s)

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Paper ID: **ncCD2**

It's our pleasure to inform you that the manuscript entitled 'Thermal degradation of household solid wastes with TGA: cellulosic and plastic materials' is accepted for publication in Vol. 160, Issue. 9 of SYLWAN journal (ISSN: 0039-7660) based upon the reviewers' positive comments on this paper.

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SYLWAN

7660-0039 :ISSN

0.263 :Impact Factor

(JCR Included-ISI)

Peer Reviewed Multidisciplinary Cited-Highly

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ISSN: 0039-7660

SYLWAN

.English Edition.



2014

[SYLWAN 158(4)]. ISI Indexed.

Printed in Poland

Thermal degradation of household solid wastes with TGA: cellulosic and plastic materials

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Abstract: Thermal behavior of household waste can be improved by knowing thermal degradation for the optimal design and operation of thermochemical process: pyrolysis, gasification or combustion. In the present study, Thermogravimetric analysis has been used for pyrolysis and combustion of waste plastic, cardboard, wood, and household waste of Abomey-Calavi city in Benin at different heating rates. For facilitating the laboratory study of household wastes, a model waste (MW) based on wood (88%), cardboard (7%) and plastic (5%) was developed. The model waste is based on the typological characterization of household solids wastes (HSW) and chemical assumptions.

The difference of mass loss between experimental and theoretical ones (calculated as arithmetic sums of those from each separated component) was used as a criterion of synergetic effect. The experimental results indicated that a low synergetic effect existed between plastic and cellulosic materials during the pyrolysis of MW in contrast to the effect observed in case of combustion.

Keywords: Thermogravimetric analysis, household waste, model waste, pyrolysis, combustion.

1. Introduction

Choice of waste management systems depends on decisions by city leaders as well as strategic structures related to the nature, quality and quantity of waste produced [1,2]. Waste management is a challenge that local authorities and researchers address using multidisciplinary approaches ranging from the humanities to exact sciences such as biology and engineering [1].

The level of development of a country has an impact on its waste management choices [1,3,2]. According to C. Riber et al., developed countries utilize various methods for waste management which give way to renewable energy forms and the emergence of new products such as compost [4]. In these countries, considerable investment is made to recycle waste for the benefit of agriculture [5,6].

The development of African cities inevitably leads to the accumulation of waste quantities increasingly important and centralized. Moreover, the use of plastic bags made from PVC (or Low Density Polyethylene: LDPE) is a scourge, these bags are not biodegradable, they accumulate in all locations of towns and in particular peripheries [7,8].

Studies on the characterization of African waste also show the presence of large amounts of organic matter, sand and various plastics in household waste. If organic materials can be used to produce compost under certain conditions, plastics are more difficult to manage because they are often contaminated and mixed with various inorganic materials (mainly sand), which does not allow for easy recycling.

Many researchers are widely being concerned with environmental friendly disposal of solid waste and biomass as a resource [9-12]. One of the promising ways for utilization of solid waste is pyrolysis. The pyrolysis process involves breaking down the molecular in the wastes under moderate pressure and temperature to produce liquid fuel [13]. Several authors have studied plastics pyrolysis and have demonstrated the potentialities of this technology to deal with the plastic wastes [14-17]. However, there are two constraints of the pyrolysis process [17]: first, recycled materials are not suitable for the whole



range of applications; and second, different components of wastes are not easily separated from the post-consumer stream.

Another type of waste that raises some environmental questions is biomass-derived waste. They accumulate on forest ground and cause forestry fires; their organic and energetic potential are wasted in this manner [18]. Pyrolysis of biomass has been studied to produce a liquid product with adequate fuel properties (called "bio-oil") [19]. However, some disadvantages of bio-oil obtained from biomass pyrolysis such as higher oxygen content, lower high-heat value (HHV), lower volatile, and delayed ignition time in engine severely hinder its use in practice [20,21]. Thus, it is necessary to explore how to improve the quality of the bio-oil. At present, there are two main pathways to upgrade the bio-oil: (1) High-pressure hydrogenation processing and catalytic cracking [22]; (2) Co-pyrolysis for the mixture of synthetic polymers and biomass [14-17]. The latter have received much attention in recent years because it provides an alternative way to dispose and convert synthetic polymers and cellulose (or lignin)-derived materials into high-value feedstock and specific chemical [23]. At the same time, the co-pyrolysis need not bring in high-pressure hydrogenation, so the production process is more safe. In addition, the plastics and biomass often co-exist in the municipal solid waste (MSW), and there is no need to add other material into the co-pyrolysis process. Therefore, the co-pyrolysis process of plastics and biomass has the broad application prospect, and H-transfer may also be involved in the co-pyrolysis under ambient pressure conditions [24].

As we know, biomass energy is one of the most important renewable energy, and its application becomes more and more significant for environmental protection. Compared with the other renewable energies, biomass is abundant in annual production, with a geographically widespread distribution in the world [25]. The co-pyrolysis of plastics and biomass is a very promising method to reduce the volume of waste and allow the recovery of chemicals and replacement of fossil fuels [14,26].

Combustion is a method of treatment and elimination of HSW allowing reduction of about 90% in volume and 75% of the mass of waste and complete destruction of bacteria [27-30]. It leads to the formation of slags which have the advantage of being valued, under certain conditions, in public works. It consists to burn the waste in an oven specifically adapted from temperature about 850 ° C by releasing the heat and steam, effluent gas, bottom ash and fly ash (3 - 4 %) [31,32].

Combustion is a complex phenomenon involving simultaneous heat and mass transfer with chemical reaction and fluid flow. Using an analogy to coal combustion, the combustion of biomass can be considered as a three stage process: devolatilisation to char and volatiles, and combustion of the volatiles and of the char [33]. The water content is significant and important and in some instances may dominate the biomass combustion process. Pollutant emissions generated by combustion are particular matter (PM), CO, HC, oxides of nitrogen and oxides of sulphur. Nitrogen oxides (NO_x) are some of the most important substances resulting from the combustion of fuels. Typical combustion gases contain two kinds of oxides: NO and NO₂. Other kinds of nitrogen oxides are N₂O, N₂O₃ and N₂O₅, but they do not play an essential role. Nitrogen oxides are, environmentally, the most harmful substances which is why researches study how to reduce NO_x emissions. This requires knowledge of the mechanisms of the formation and reduction of NO_x emissions based on the kinetics of chemical reactions [34]. Carbon oxide is a very harmful substance for living organisms, especially above permissible air emission standards. Carbon oxide, with its property of superseding the oxygen in the oxyhaemoglobin of the blood, forms bonds with the haemoglobin, generating carboxyhaemoglobin leading to suffocation. Sulphur dioxide is considered to be one of the fundamental air pollutants. SO₂ is assumed to be the only representative of sulphur oxides in combustion gases (at least 98 % of the total amount of sulphur oxides). During combustion some specific pollutants such as Cl₂, HCl, salts, dioxins and furans can occur. The emission of exhaust gases is a significant problem in the combustion of fuels. Numerical simulations using, e.g. CHEMKIN can model of biomass combustion to predict the amount of pollutants generated (NO_x, CO, SO₂) in the exhaust gases coming from boilers [35].



Pyrolysis and Combustion characteristics of biomass fuels have been studied widely using thermoanalytical techniques. The advantages of thermogravimetric analysis are its rapid assessment of the fuel value, the temperatures at which process starts and ends, and other characteristics such as maximum reactivity temperature, ash amount and total reaction time. Thermogravimetric analysis is very useful for studying the kinetics of pyrolysis and combustion processes [36,37,34,35,38-42]. Thermal methods such as TG, DTG and DTA have been used for studying a variety of areas of combustion. Thermal analysis (TA) determines a set of methods for study of the selected physical properties of the substance under the influence of temperature. Sometimes, simultaneously, the environment (pressure, atmosphere chemical composition) can be changed. Thermogravimetry (TG) is a technique which monitors the sample mass as a function of temperature or time when the sample is subjected to a controlled temperature program. Derivative thermogravimetric (DTG) is based on the rate of mass loss. DTG profiles make it possible to know, for example, the mass loss which is taking place at a temperature during the reaction process.

In Benin, (West Africa), few scientific studies have undertaken the management of household waste. The present study was undertaken as an extension of work of Topanou N et al who characterized the typology and size of waste particles, their physico-chemical characteristics, organic matter content and contamination by metal trace element to better determine the amount of solid waste produced and collected at the source in the district of Abomey-Calavi [43], a city of Benin which doesn't get an effective system of waste management.

In this work it is studied the pyrolysis and the combustion of the combustible household waste in the city of Abomey-Calavi using Thermogravimetric Analysis. Only the variations of the total mass of solid and of the cell temperature can be recorded in the course of time. The solid sample is assumed homogeneous in temperature and composition. In conclusion, the only input available to improve the experimental conditions is the programmed profile of the cell temperature.

The heterogeneous nature of household waste not facilitating their laboratory study, it was developed a model waste based on wood, cardboard and plastic as recommended by ADEME [44].

2. Experimental procedure

Plastic waste considered in the model waste of this study was the polyethylene (PE) because it is the only category of plastic that found in domestic waste in the Abomey-Calavi town. Because of advanced recycling of high density polyethylene (HDPE), low density polyethylene (LDPE: plastic bag black color) was only plastic waste used in this study. The plastic waste was cut into small pieces (approx. 1 mm²). The cardboard material used in this work is corrugated cardboard generally found in household waste from the city of Abomey-Calavi in Benin. The carton waste was also cut into small pieces (approx. 1 mm²). The wood materials used were wastes chip and sawdust.

The experimental work was carried out on a computerized thermobalance (NETZSCH STA 449 F3 Jupiter) using a high speed furnace which allows a heating rate faster than 500 °C min⁻¹. The thermobalance configuration gives a sensitivity of ±0.4 µg. It allows us to use small sample mass (10–50 mg) which is needed to ensure isothermal conditions in samples. In order to establish an inert atmosphere during all experiments, a controlled argon flow (carrier gas) (fixed at 200 mL min⁻¹, 273 K, 1 atm) sweeps the measurement cell that is purged for 20 min before starting the heating program. During experiments of pyrolysis, the argon flow is set to 20 mL min⁻¹. In combustion, argon is replaced by air. The initial dry mass of samples is about 30-50 mg.

Studies with TG instruments are typically conducted under isothermal conditions or non-isothermal conditions. Some experimental difficulties can exist with isothermal experiments i.e. a significant part of the reaction may occur during the setting of the experimental temperature at the beginning of the experiment; for instance, see [45] for more precision about this subject. One advantage of the isothermal conditions is the homogeneous sample temperature after the isothermal reaction temperature has been reached, whereas in non-isothermal mode, a temperature gradient in the sample can occur due to the



resistance of heat conduction in the sample and the resistance of outer heat transfer [46]. A way for compensating for this effect is to use low heating rates. Isothermal and non-isothermal methods have been widely used in the literature but papers comparing the results obtained with the two methods are rare [47]. Furthermore, it is not easy to compare the scientific studies published for a given product: the experimental conditions (equipment, technologies, operating conditions, sample conditioning, etc.) are often different and not always well known; moreover, the models, the data treatments and the computational methods are never the same [45].

Dynamic experiments were performed in this paper. For these tests, samples have been previously dried in an oven at 103 °C for 48 h.

Raw materials sample were taken and heated from room temperature to a final temperature of 900 °C and a residence time of 30 minutes at 900 °C. TG was performed in Argon and air atmosphere at the heating rates of 5, 10, 20, 30, 40 and 50 °C min⁻¹ under 20 mL min⁻¹ gas flow rate. Derivative thermogravimetric mass loss curve was plotted against temperature. It provides a range of temperature in which maximum thermal degradation of waste materials takes place. This mass is high enough that even small temperature gradients are observed in the particles. However, this effect is limited due to the position of the thermocouple is as close to the center of the sample.

3. Results and discussions

3.1. Model of household waste of Abomey-Calavi city

Household wastes have a variable composition depending on the season and the places of their production. In the case of a region or a city, find a representative composition of the waste requires campaigns characterization of the waste according to the seasons, to obtain a statistically valid model. The latest campaigns characterization waste from the city of Abomey-Calavi were performed by Topanou N et al [43]. Table 1 shows the results of these campaigns.

Table 1 : Typological characterization of household solids waste in Abomey-Calavi

	Dry Season	RainySeason	Average
Fermentable	48,64	53,50	51,07
Sand	34,50	19,64	27,07
Cardboard Paper	1,14	1,76	1,45
Plastics	2,69	2,75	2,72
Glass	1,50	1,45	1,47
Metal	1,04	1,44	1,24
Textiles	2,28	2,79	2,54
Others	8,23	16,67	12,45

The composition of model waste reflects the fuel from household waste for the thermochemical treatment.

- fermentable and greens wastes are chemically treated as wood;
- cardboard, papers, textiles, and sanitary textiles are treated as cardboard;
- plastics and special wastes are treated as plastics;

On the basis of these assumptions, the model waste (MW) of the city of Abomey-Calavi is composed of **88% wood, 7% cardboard and 5% plastic** according to the mass fraction.

3.2. Fuel analysis

Before thermogravimetric analysis, materials were analyzed to determine the main properties affecting thermal conversion. The proximate and ultimate analyses of the studied samples were determined (Table 2 and Table 3). The properties including volatile matter, ash, and moisture content and chemical composition were tested in accordance with French standards (NF EN 14774, NF EN 1510, 14775, NF EN 15148 and NF EN 14918). The moisture content was determined by successive weighing of a sample



placed in an oven at 103°C until achieving constant mass. The proximate analyses were performed on the TG apparatus in described in section 2. The results, similar with those found in the literature are presented in Table 2 and Table 3.

Table 2 : Proximate Analysis of wastes plastic, cardboard, wood and model waste

	Plastic	Cardboard	Wood	MW Experimental	MW Calculated
Volatil Matter (%)	97,72	81,93	80,43	80,21	81,40
Fixed Carbon (%)	0,71	7,45	14,2	13,72	13,05
Ash Content (%)	1,57	10,62	5,37	6,07	5,55
Moisture Content (%)	1	6,7	9,6		8,97

Table 3 shows the values of elemental analysis of model waste (MW) determined from elemental compositions of wood, plastic and cardboard found in this study and in agreement with the literature [48-50]. The analysis of Table 3 shows that the model waste of the city of Abomey-Calavi is mainly composed of Carbon, Hydrogen and Oxygen. However, there is the presence of traces of chlorine atoms which are linked to the process of manufacturing plastic bags and in particular the fact that additives are added. It also appears clearly that the composition of the waste model and the wood are very similar.

Table 3 : Elemental Analysis of Model Waste (MW)

Sample	C%	H%	N%	O%	S%	Cl%	Ash %	Total (%)
Wood	44,79	5,07	0,18	37,05	0,04	0,00	0,88	88
Cardboard	3,06	0,40	0,01	3,15	0,01	0,00	0,37	7
Plastic	3,69	0,58	0,01	0,24	0,01	0,14	0,34	5
MW (%)	51,54	6,04	0,19	40,43	0,06	0,14	1,59	100

3.3. Pyrolysis study

As has been stated in previous works on pyrolysis of lignocellulosic and polymeric materials [51-54], dynamic experiments carried out with constant heating rates present some advantages over isothermal experiments. One of these advantages is the possibility of obtaining results in a larger temperature range and more representative of industrial process. Furthermore, these experiments allow the study of the influence of the heating rate β on the thermal decomposition process. Taking this into account, experiments at different β between 5 and 50 °C min⁻¹ have been carried out.

Fig.1 shows Derivative Thermogravimetric (DTG) curves of different wastes materials at 10 °C min⁻¹. The corrugated cardboard and the wood decompose thermally within relatively narrow temperature regions between 200 and 400 °C and causes a more important mass loss as found in the work of David, C and Al [55] and Han, B and Al [56] with a temperature range of degradation of 300-450°C and 200-400°C respectively. Moreover, the decomposition of plastic takes place between 400 and 600°C as it has already been established in some work [57-59]. The DTG plot of MW shows two step of decomposition. The first decomposition is carried out between 200 and 400°C as found for cardboard [60] and wood. The second decomposition takes place between 400 and 500°C as found for LDPE [7]. These results show that there would no synergetic effects between biomass and PE blends. It would be nice to verify this assumption later in the study.



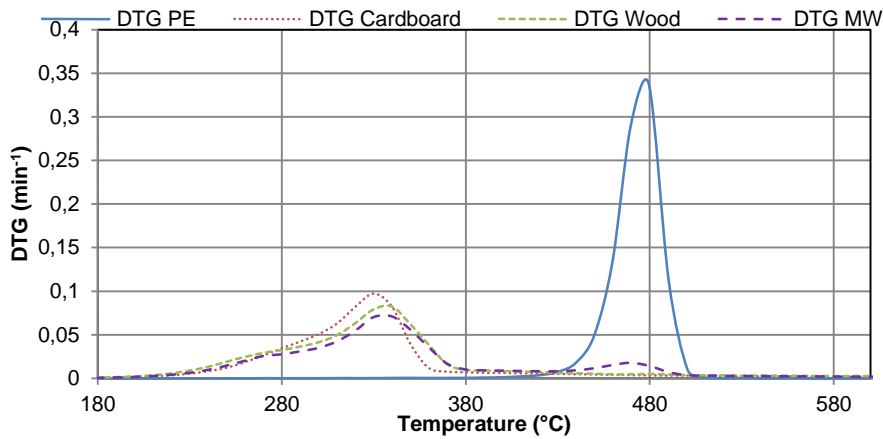


Fig.1DTG of Wastes Materials at 10°C.min⁻¹

DTG curves of wood and MW at different heating rates (Fig.2) were shifted to higher temperature due to the heat transfer enlarging with increasing heating rate as it was found in previous studies on LPDE [7] and cardboard [60]. Similar effect of various heating rates on DTG curves for HDPE pyrolysis has been obtained by A. Aboulkas [61] and S. Kumar [59]. It is also the case for more authors who studied the thermal degradation of polyethylene and cellulosic materials [62-64,58,65].

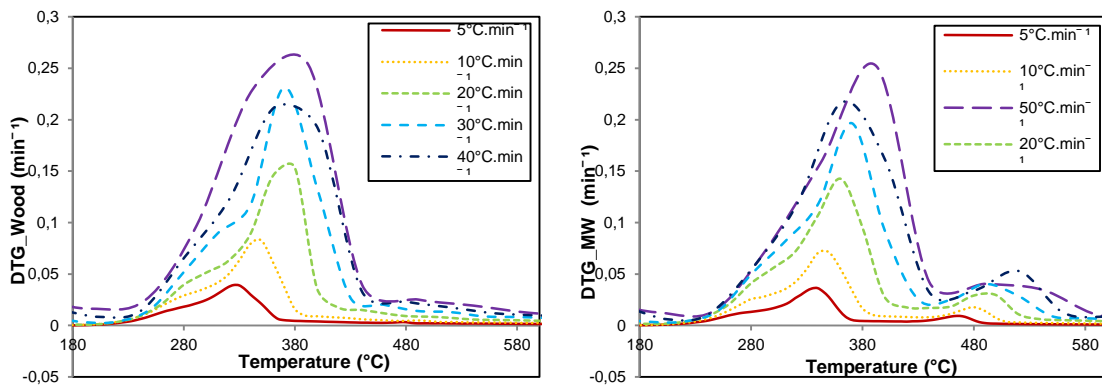


Fig.2 DTG plots of wood and MW at different heating rates

To further illuminate the synergetic effect between the plastic and biomass (wood and cardboard) samples, we defined the difference of mass loss as (ΔW):

$$\Delta W = W_{MW \text{ Experimental}} - W_{MV \text{ Calculated}} \tag{1}$$

$$W_{MW \text{ Calculated}} = (x_1 W_1 + x_2 W_2 + x_3 W_3) \tag{2}$$



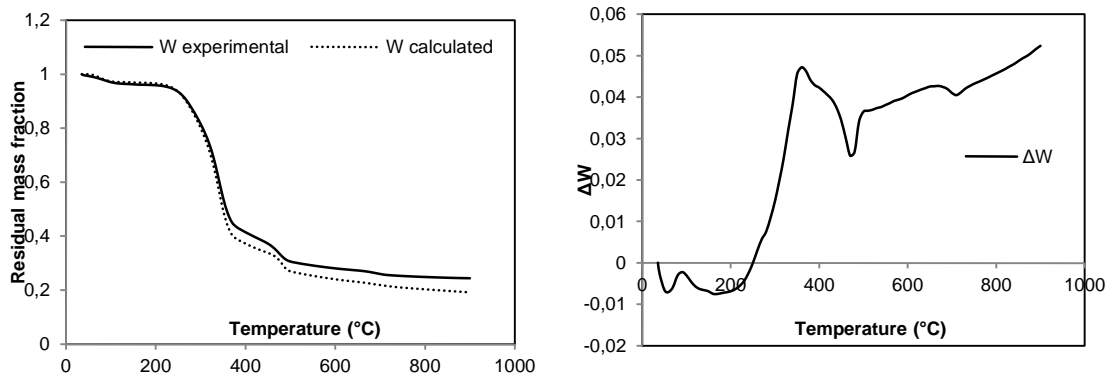


Fig.3 Curves of synergetic effects versus the temperature (°C)

x_i is the fraction of each material in the blend, and W_i is the mass loss of each material in the same operational conditions. Obviously, ΔW describes the “extent” of synergetic effect during the process of co-pyrolysis between plastic and cellulosic materials.

Fig.3 shows the significant interaction, i.e., the synergetic effect between PE and cellulosic materials. Before 300°C, LDPE softened and had only a slight impact on the decomposition of the blend in the first stage $\Delta W < 1\%$. ΔW becomes important after 300°C and varies between 1 and 5%. This behavior was probably due to the presence of chlorine atoms which are linked to the process of manufacturing plastic bags and in particular the fact that additives are added. Indeed, the HCl released from LDPE can act as an acid catalyst to promote dehydration and coke generation in biomass pyrolysis and inhibit the fracture of cellulose molecular chains [66-68]. These results show that the synergetic effect between cellulosic materials and plastic is very low.

3.4. Combustion study

Fig.4 shows Derivative Thermogravimetric (DTG) curves of different wastes materials at 10 °C min⁻¹. The cardboard and the wood decompose thermally within relatively narrow temperature regions between 200 and 500 °C in two stages: a first stage between 200 and 350°C causes a more important mass loss and a second stage corresponding to the second peak appearing between 350 and 500 °C. Comparison with the results of pyrolysis study (degradation between 200 and 350 °C) shows that the first peak resulting from the degradation of combustion corresponds of the pyrolysis during which the volatile species are produced. The second phase of the degradation then represents the combustion phase of the char obtained in the first phase. The lower second peak and the temperature range of cardboard compared to wood are explained by the difference between their fixed carbon values (see Table 2).

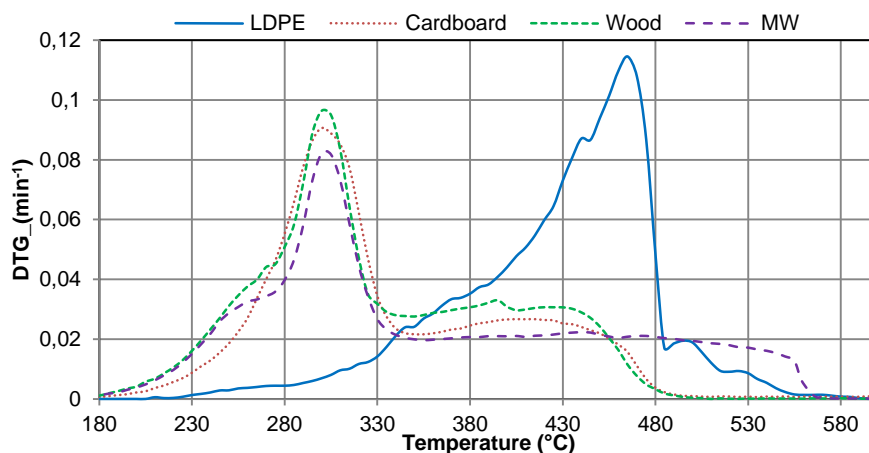


Fig.4 DTG of Wastes Materials at 10°C.min⁻¹



Combustion of the plastic takes place between 200 and 600 ° C and has only one peak of degradation between 430 and 480 ° C. Combustion of plastic starts at lower temperatures than the pyrolysis (between 400 and 600 ° C). It is therefore acceptable that the combustion of MW is carried out between 200 and 600 ° C as shown in Fig.4. The peaks observed show that there is a possible synergy between the plastic and cellulosic materials in the case of combustion.

Fig.5 shows the synergetic effect between PE and cellulosic materials obtained with Eq.(1) and Eq.(2). Before 200°C, LDPE softened and had only a slight impact on the decomposition of the blend in the first stage $\Delta W < 1\%$. ΔW becomes important after 200°C, varies between 0 and 15% and sweep degradation range of the plastic. The interaction between the plastic and the cellulosic materials is not negligible in combustion in contrast to the effect observed in pyrolysis (0 to 5%). The presence of plastic slows combustion of the model waste by increasing its temperature range of and duration of thermal degradation. This behavior is probably not only due to the presence of chlorine atoms but also by the presence of oxygen.

The synergistic effect is significant between the plastic and cellulose materials, it is a priori possible to carry out the combustion of MW from 200 ° C.

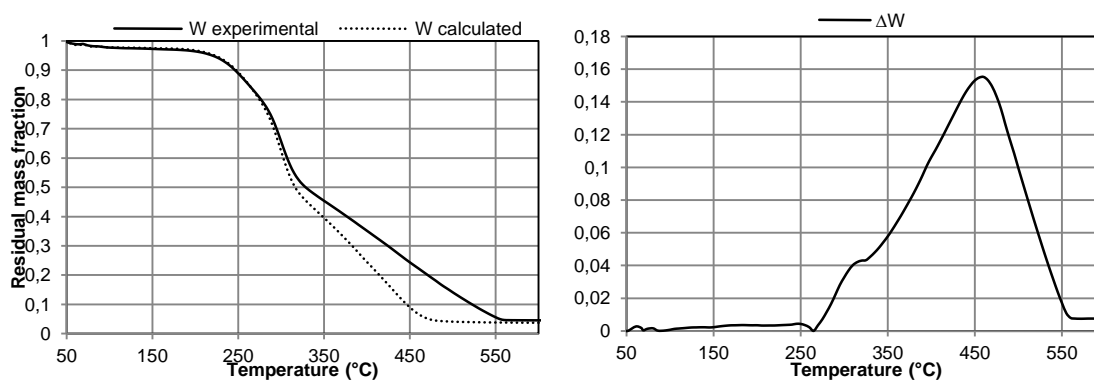


Fig.5 Curves of synergetic effects versus the temperature (°C)

4. Conclusion

Dynamic experiment showed that the pyrolysis of model waste of the city of Abomey-Calavi takes place in two temperature ranges: 200-400°C and 400-500°C for the cellulosic materials (wood and cardboard) and Plastic (LDPE bag used in Benin) respectively. In the other hand, the combustion of cellulosic materials takes place in two stages between 200-500 °C which illustrate the pyrolysis of raw material and the combustion of the char obtained. The combustion of plastic and model waste were carried out from 200°C. The synergetic effect in combustion is important in contrast to the effect observed in pyrolysis.

In conclusion, these results show that the pyrolysis of household waste of Abomey-Calavi and other waste composed of cellulosic materials and LDPE is not feasible at the temperatures below 400°C whereas its combustion may be occurred from 200°C. However, what would be the consequences on emission of pollutants?

Acknowledgments

This work could not have been performed without financial support of CPER "Région Lorraine" for the equipment and instrumentation, financial support of Labex Arbre. The authors wish to thank these Organizations for their support.



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