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## EXPERIMENTAL INVESTIGATION ON THE HEAT OF COMBUSTION FOR SOLID PLASTIC WASTE MIXTURES

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### Abstract

The aim of this paper is to determine the heat of combustion of plastic wastes resulted from municipal solid waste, automotive shredder facility waste and building and construction waste. The plastic wastes have been separated by flotation technique using as flotation media: water, ethanol, their mixtures and magnetic fluid. Resulted fractions have been analyzed aiming to determine the most effective fraction from the heat of combustion point of view. The obtained results have been compared to those reported in the literature, with those calculated by oxygen consumption method and those proposed in this paper and calculated by weighted sum of combustion heat of components, aiming to allow its approximation for different fractions of polymeric wastes with known composition, avoiding the experimental measurements. Deviations of measured values of the heat of combustion from those obtained by theoretical calculation have been explained by the polymer degradation during their life cycle. The most effective fraction from calorific point of view is that containing polyolefins, but this fraction could be mechanically recycled. The present study evidenced that the heat of combustion of the plastic waste decreases after polyolefin extraction and the remaining density fractions can be effectively used for energy recovery of the plastic waste by incineration.

*Key words:* heat of combustion, oxygen bomb calorimetry, polyolefins, polymers waste, recycling

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### 1. Introduction

The main purpose of this paper is to determine the heat of combustion of plastic wastes coming from municipal solid waste, the automotive shredder facility and building and construction sector as mixture of polymers. The heat of combustion can decide the effectiveness of plastic wastes to be used as fuel for energy recovery as reported (Banyai and Fodor, 2014; Corabieru et al., 2014; EC-PWE, 2010; Ghinea et al., 2014; Hidalgo et al., 2014; Kocsis and Kiss, 2014; Luca and Ioan, 2014; Orlescu et al., 2013; Sarkady et al., 2013a, 2013b; Şchiopu and Ghinea, 2013; Simion et al., 2013).

The plastic wastes have been separated by flotation technique using as flotation media: water, ethanol, their mixtures and magnetic density separation (MDS) using a magnetic fluid (Rem et al., 2013). Resulted fractions have been analyzed aiming to determine the most effective fraction from the heat of combustion point of view. The results have been compared to those reported in the literature by Van Krevlen (1990), Walters et al. (2000) and Lechner (2005), with those calculated by oxygen consumption method (Babrauskas, 1992; Van Krevlen, 1990; Vilcu and Leca, 1990) and those calculated by weighted sum of combustion heat of components, aiming to allow its approximation for different

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fractions of polymeric wastes with known composition, avoiding the experimental measurements.

This study presents the effect of the polyolefins extraction from the polymeric wastes on the contaminants fractions heat of combustion, and it evidenced that the density fractions resulted could be effectively used for energy recovery of the plastic waste, by incineration.

## 2. Material and methodology

### 2.1. Material

The tested wastes investigated are covering the municipal solid waste source (WS1), the automotive shredder waste source (WS2) and building and construction waste source (WS3) as major contributors to plastic waste production. The source WS1 came from Romania, the source WS2 from Austria and source WS3 from France. Plastic wastes as were received as mixture of polymers are presented in (Fig. 1).

The plastic wastes have been separated by flotation technique using as flotation media: water, ethanol, their mixtures and magnetic fluid to separate different density fractions to extract polyolefins. The separation technique is presented in (Fig.2) and details are reported by various authors (Moldovan et al., 2012; Patachia et al., 2011; Vajna et al., 2010). The flotation process separates the polymer mixture in two fractions: float fraction with density smaller than 0.998 kg/m<sup>3</sup> and sink fraction with density bigger than 0.998 kg/m<sup>3</sup>. The float fraction is

processed with magnetic density separation and results three density fractions: polyolefin fraction with density between 0.880 and 0.964 kg/m<sup>3</sup>, light fraction with density smaller than 0.880 kg/m<sup>3</sup> and heavy fraction with density between 0.964 and 0.998 kg/m<sup>3</sup>. The plastic wastes from WS2 and WS3 came with polyolefin fraction already extracted. After separation the resulted fractions are dried to remove the moisture. Fractions composition has been determined by a combination between the image analysis, gravimetric method and FTIR spectroscopy by using a Perkin-Elmer BXII Fourier transform infrared spectrometer, equipped with an attenuated total reflectance (ATR). Polymer identification has been made by using Essential FTIR software data base, (eFTIR, 2013). The composition results were reported by Baltes et al. (2009, 2013), Patachia et al. (2010, 2011) for WS1, Vajna et al. (2010), Cazan et al. (2013) for WS2 and EC-PWE (2010) for WS3.

Reported values for WS1 light fraction composition was 17% polyethylene(PE) and 83% polypropylene(PP), for heavy fraction a composition of 12% PE, 38%PP, 11%PS, 2%PET and 37%PVC. For WS2 the light fraction contains 9%PP, 23% polyurethane foam, 5% STPe, 55% cellulose and 8% epoxy adhesive, the heavy fraction contains 25% polyethylene, 15% polypropylene, 11% PET, 5% polystyrene, 10% nylon, 15% cellulose, 5% PVC, 10% EPDT and 4% PC. The composition of WS3 light fraction is 15% PE, 23% poly(1butene), 18% LLDPE, 15% polymethylpentene, 26% ethylene propylene rubber and 3% polyacril amide. The WS3 heavy composition is 56% PE, 24% PP, 14% polybutadiene and 6% construction adhesive.



Fig. 1. The test samples of plastic waste

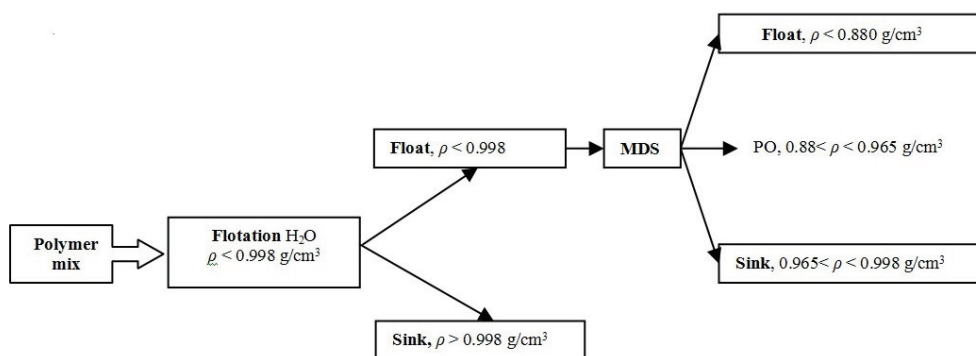


Fig. 2. The separation scheme

**Table 1.** Polyolefin percent in plastic waste mixture

Waste source	Polyolefin amount (%)			
	Light fraction		Heavy fraction	
	PE	PP	PE	PP
WS1	17	83	38	12
WS2	0	9	25	16
WS3	33	0	56	24

The size of shredded waste is about 4 mm or bigger which is not suitable for heat of combustion measurements. To determine of the heat of combustion of solid fuels, maximum grain size of 1 mm according ECS (2006) and homogeneity of mixture are necessary. The initial samples have been cooled with liquid nitrogen and grinded by using ultra centrifugal mill (Retsch ZM200, Retsch GmbH, Germany). The average sample size obtained after grinding was approximately of 0.5 mm. The samples are prepared dry and ash free and sample heat of combustion value has been measured in the absence of moisture and ash forming minerals. The tested samples are: mixture of polymers from WS1, WS2 and WS3, and light and heavy fractions resulted after MDS separation of waste.

It could be noted that PP and PE as polyolefin major fractions are present both also in lighter and heavier fraction of all the separated wastes as listed Table 1.

The lighter fraction of WS1 coming from urban plastic waste contains 100% polyolefins and 50% is found in the heavier fraction, the WS3 coming from building and construction waste contains 33% polyolefins in lighter fraction and 80% in heavier fraction and WS2 coming from automotive shredder waste contains 9% polyolefins in the lighter fraction and 41% in the heavier fraction. The samples WS2 and WS3 analyzed were received with form The polyolefin percent quantities in the light fraction can be explained by the presence of the PP and PE foam and bubble wrap and by the adherence of the air bubbles to PP and PE during the flotation process and magnetic density separation.

### 2.2. Measurement of the heat of combustion

Both the initial waste samples and the obtained fractions have been submitted to calorimetric analysis. After drying, the samples were weighed using precision analytical balance of 0.1 mg (Kern & Sohn ABJ 220-4M model). Each sample weight was at approximately 1 gram in order to get a accepted temperature rise of 2 liters of water in the oxygen bomb jacket of 2.0-4.0°C for calorimetry measurements according to standard (ECS, 2006) at ambient temperature of 25°C.

The higher heat of combustion was measured in an oxygen bomb calorimeter model XRY-1C, Plain Jacket Oxygen Bomb Calorimeter, Shanghai Luheng Instrument co, with data acquisition software. For each tested sample were measured the mass of probe and the ignition wire mass. The bath

temperature was monitored and recorded during burning process. The calculus is made using the Regnault-Pfaundler method as described in (RNS, 1995; ECS, 2006; GNS, 2000) using recorded temperature values.

### 2.3. Calculation of the heat of combustion

The following methods were considered in the view of the calculation of the heat of combustion:

a) The oxygen consumption method (Walters et al., 2000) considers that the wide range of organic compounds including polymers have the same heat of combustion per gram of diatomic oxygen consumed. The empirical quantity used is  $E=13.10\pm0.78$  kJ/g- $O_2$  and authors have found that the estimation of the heat of combustion for 49 pure polymers, using the Eq. (1) is about  $\pm 4.4\%$ , where  $Q_c$  is the heat of complete combustion of the sample with all products in gaseous state,  $n_{polymer}$  is the number of moles and  $M_{polymer}$  is the molecular weight of the polymer repeating unit,  $n_{O_2}$  is the number of moles of  $O_2$  consumed in balanced thermochemical equation,  $M_{O_2} = 32$  g/mol is the molecular weight of diatomic oxygen and the quantity  $r_0$  is the stoichiometric oxygen-fuel mass ratio.

$$Q_c = E \cdot \left( \frac{n_{O_2} \cdot M_{O_2}}{n_{polymer} \cdot M_{polymer}} \right) = E \cdot r_0 \quad (1)$$

b) The proposed as second method to calculate heat of combustion is described by the Eq. (2) using literature data for polymers as a mass weighted contribution of the heat of combustion of pure polymers that could be find in a density fraction, where  $\%_w polymer$  is the mass ratio of each fraction in the mixture and  $Q_{c,polymer}$  is the heat of combustion of each pure polymer.

$$Q_{c,mix} = \sum(\%_w polymer \cdot Q_{c,polymer}) \quad (2)$$

The reported values from literature for the heat of combustion of polymers are listed in Table 3. The values used in Eq. (2) for the heat of combustion of pure polymers are represented as bold values in Table 2, and were selected considering experimental determination of them. The overall mixture heat of combustion was considered as a mass weighted contribution of the heat of combustion of density fractions, without considering the interaction between them.

**Table 2.** Reported heat of combustion for pure polymers

<i>Polymer</i>	<i>Heat of combustion (J/g)</i>	<i>References</i>
PE	44600	Babrauskas (1992)
	<b>47740</b>	Walters et al. (2000)
	46400	Lechner (2005)
	47195	NIST (2013)
PP	42660	Babrauskas (1992)
	<b>45800</b>	Walters et al. (2000)
	44000	Lechner (2005)
	45799	NIST (2013)
PS	42500	Babrauskas (1992)
	<b>43650</b>	Walters et al. (2000)
	41600	Lechner (2005)
PA	<b>31400</b>	Lechner (2005)
PET	23220	Babrauskas (1992)
	<b>24130</b>	Walters et al. (2000)
	21600	Lechner (2005)
PVC	17950	Babrauskas (1992)
	<b>19000</b>	Lechner (2005)
	18000-19000	NIST (2013)
PU foam	<b>31600</b>	RNS (2010)
STPe	<b>32600</b>	Smirnova et al. (2010)
	33900	RNS (2010)
Cellulose (wood)	<b>17000-20000</b>	RNS (2010)
Epoxy adhesive	34930	Babrauskas (1992)
	<b>31370</b>	Walters (2000)
	31700	RNS (2010)
Modified cellulose	<b>20000</b>	RNS (2010)
EPDT	<b>40600</b>	RNS (2010); Chuang (1997)
PC	31000	Babrauskas (1992)
	<b>31300</b>	Walters (2000)
	30700	Lechner (2005)
PB	45000	Lechner (2005)
	<b>45334</b>	NIST (2013)
LLDPE	<b>44600</b>	Walters et al. (2000)
	46500	Lechner (2005)
PMP	<b>47496</b>	NIST (2013)
EPR	<b>33900</b>	RNS (2010)
PAM	22425	Vatani et al. (2007)
	<b>21901</b>	Smirnova et al. (2010)
BR	<b>44200</b>	NIST (2013)
	44213	CCD (2013)
	44250	EPST (2005)
C&R adhesive	<b>14000-19500</b>	RNS (2010)

2.4. Calibration

As is mentioned by Walters et al., (2000), and described in (RNS, 2010; ECS, 2006; GNS, 2000), before determining the calorific value of tested samples the oxygen bomb calorimeter must be calibrated. For calibration a standardized benzoic acid sample is used. The heat of combustion of standard benzoic acid used is  $H_e = 26454$  J/g as is given by Parr Instrument CO. The weighted benzoic acid mass of the sample is  $m_e = 1.0018$  g. To initiate burning a 0.1 mm nickel chromium alloy wire is used as standardized fuse wire from Parr Instrument Co. with a given heat of combustion of 5861.52 J/g. The heat correction for the wire and cotton burning resulted is  $Q_{wc} = 131.36$  J. The final temperature recorded during calibration from the main stage is  $t_f =$

20.453 °C, and the initial temperature from the main stage is  $t_i = 18.405$  °C. The thermal capacitance of the calorimeter,  $W$ , it is determined using Eq. (3).

$$W = \frac{H_e \cdot m_e - (Q_{wc})}{(t_f - t_i)} = 12875 \text{ J / K} \tag{3}$$

2.5. System setup

According to standard procedure the weighted sample is placed inside a calibrated adiabatic bomb calorimeter with 1 mL of deionized water. The ignition wire is connected to the electrodes in the pressure vessel and placed in contact with the sample. The bomb is then assembled, sealed and purged twice by pressurizing to 0.4MPa with pure oxygen to evacuate the air. Before determination it is

pressurized with pure oxygen to 3.0MPa and placed inside a bath containing 2 liters of water in an insulated jacket. Temperature rise of the water is measured by using a precision sensor with  $10^{-3}$  K temperature resolution. The equilibrium temperature of the bath during to the test is recorded by the data acquisition system at every 30 seconds. Three replicates are performed for each polymer mixture sample. The results recorded during the experiment are the values of temperatures for before period to reach adiabatic conditions, main burning period and after burning period.

### 3. Results and discussion of calorimetric analysis

The results obtained for mixed plastic waste with standard deviation are presented in Table 3, where  $Q_{hi}$  is higher heat of combustion and  $Q_{lo}$  is lower heat of combustion. Results of calorimetric analysis of remained fractions after extraction of polyolefins from samples are presented in Table 4. Each result is the average of at least 5 determinations.

As it can be seen in Table 3, the higher heat of combustion of waste, before extraction of polyolefin fraction, has a deviation of values of 2% for WS1 compared to WS3, of 6.4% variation for WS1 compared to WS2 and of 9.1% for WS3 compared to WS2.

The higher heat of combustion after extraction of polyolefin fraction from WS1 and WS3 waste decreases with the increase of the fraction density for sample (Table 4). For WS2, the light fraction density has a smaller heat of combustion than the heavier fraction. That could be explained by considering the reduced polyolefin percent identified in WS2 light fraction density of 9%.

The heat of combustion for the sample with density smaller than  $0.88 \text{ g/cm}^3$ , coming from WS1, calculated by using Eq. (2) with composition of 17%PE and 83%PP, taking into account the measured values reported by Walters et al. (2000) for heat of combustion for PE and PP, respectively ( $Q_{c,PE} = 47740 \text{ J/g}$  and  $Q_{c,PP} = 45800 \text{ J/g}$ ) (Table 2), lead to a mixture's heat of combustion of  $Q_{c,mix} = 45959.27 \text{ J/g}$ . By using Eq. (1) for the same composition sample, the resulting mixture heat of combustion from oxygen consumption method, with  $r_0 = 3.42193$  is  $Q_{c,mix} = 44683.4 \text{ J/g}$ . The results obtained by using the Eqs. (1-2) are in a good agreement with the present study measured value of higher heat of combustion of  $Q_{hi} = 44824 \text{ J/g}$  (Table 5), with an absolute relative error of -0.32%, respectively of 2.53%. Table 5 presents the measured values of the heat combustion of the initial mixtures, calculated with Eqs. (1 and 2) and relative errors reported to measured value in this research.

**Table 3.** Results of calorimetric analysis of the initial plastic waste

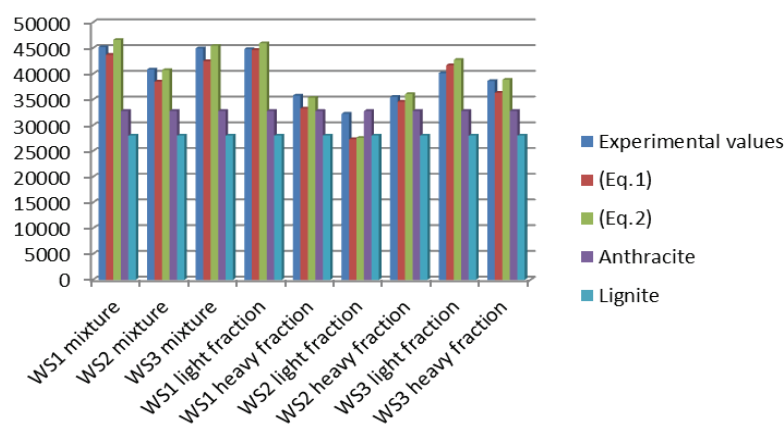
Source	$Q_{hi} \text{ (J/g)}$	$Q_{lo} \text{ (J/g)}$	StDev (%)
WS1	45202	44649	±0.24
WS2	40863	40323	±2.38
WS3	44979	44427	±1.37

**Table 4.** Results of calorimetric analysis of plastic waste fractions after extraction of polyolefins

Source	Density fraction ( $\text{g/cm}^3$ )	$Q_{hi} \text{ (J/g)}$	$Q_{lo} \text{ (J/g)}$	StDev (%)
WS2	< 0.88	32257	31757	± 1.39
WS2	0.964-0.998	35565	35051	± 3.15
WS3	< 0.88	40161	39629	± 0.96
WS3	0.964-0.998	38639	38110	± 2.1
WS1	< 0.88	44824	44272	± 2.50
WS1	0.964-0.998	35796	35281	± 4.03
WS1	> 0.998	23257	22793	± 4.09

**Table 5.** Calculated and experimental values of heat of combustion

Source	Density Fraction ( $\text{g/cm}^3$ )	$Q_{hi} \text{ (J/g)}$	$Q_{c,mix} \text{ (J/g)}$	Relative error	$Q_{c,mix} \text{ (J/g)}$	Relative error
		Present study	Eq. (1)	%	Eq. (2)	%
WS2	mix	40863	38500.07	5.34	40749.48	-0.19
WS3	mix	44979	42494.98	5.52	45449.66	-1.05
WS1	mix	45202	43741.61	3.23	46608.96	-3.11
WS2	< 0.88	32257	27313.23	15.33	27582.01	14.49
WS2	0.964-0.998	35565	34599.49	2.71	36081.95	-1.45
WS3	< 0.88	40161	41678.76	-3.78	42732.39	-6.40
WS3	0.964-0.998	38639	36343.16	5.94	38852.96	-0.55
WS1	< 0.88	44824	44683.4	-0.32	45959.27	2.53
WS1	0.964-0.998	35796	33269.54	7.06	35366.40	1.20
WS1	> 0.998	23257	28209.07	-20.51	29479.13	-26.75



**Fig. 3.** The heat of combustion (J/g) of test samples of plastic waste and the values calculated by using Eq. (1) and Eq. (2) compared with 2 sorts of coal

For WS2 samples before and after extraction of polyolefins it could be noted that the heat of combustion of the plastic mixture is higher than the heat of combustion of light fraction with 25% and higher than of heavy fraction with 15%. The relative error resulted for WS2 by comparing to measured and calculated values shows for mixture, an estimation of 5.34% with Eq. (1) and of -0.19% with Eq. (2) and for heavier density fraction of 2.71% and -1.45%. An interesting result is for WS2 lighter fraction which has relative errors of 15%. The relative errors of 21-27% has found also on estimation of WS1 density fraction bigger than 0.998 kg/m<sup>3</sup>.

For WS3 samples results show that the calorific power of the mixture is higher than calorific power of light fraction with 10% and higher than calorific power of heavy fraction with 15%. The absolute relative error for estimation of heat of combustion of WS3 is of 6%. The best estimation is made by the Eq. (2). For WS1 samples the calorific power of the mixture is higher than that of the light fraction with 1% and higher than that of the heavy fraction with 48%, That could be explained by considering the reduced polyolefin percent identified in WS1 heavy fraction density of 50%, meanwhile the light fraction have 100% polyolefins. The absolute relative error for estimation of heat of combustion of WS1 is of 5%. The best estimation is made by the Eq. (2) with 3% mean relative error.

The (Fig. 3) shows that the Eq. (2) estimates very good the experimental values for heat of combustion for WS1, WS2 and WS3 plastic mixtures, for WS1, WS2 and WS3 heavy fractions, since Eq. (1) give a good estimation for WS1 light fraction and closer values for the rest.

The lighter and heavier fractions of tested plastic waste have a heat of combustion bigger than 35.0 MJ/kg for WS1, 32.0 MJ/kg for WS2 and 38.0 MJ/kg for WS3, and greater than anthracite coal heat of combustion of 32.8 MJ/kg, or lignite coal heat of combustion of 28.0 MJ/kg, which assures their

effectiveness to an energy recovery system (Kittle, 1993).

Regarding the method used to calculate heat of combustion of plastic waste with known composition avoiding the experimental measurements has found that the method of weighted sum of combustion heat of components is a choice. This result is very useful in industrial management of plastic waste to design a plastic incineration plant which requires to calculate the estimated heat of combustion of waste and to evaluate the amount of energy recovered considering the large variety of polymers involved in plastic mixtures. Before incineration the plastic waste needs to be washed to remove organic compounds, metals and ash, to be dried to remove moisture, as additional steps. These steps determine for real plastic waste an increasing of the energy recovered by incineration and a method to reduce the amount of plastic wastes from nature.

#### 4. Conclusions

The calorimetric analysis shows that the mixed plastic wastes can be used as fuel because of high heat of combustion comparing with coals. The presence of polyolefins as the PE and PP both in the lighter and heavier fractions of wastes as composites, the heat of combustion of these fractions will be higher, due to the higher polyolefinic heat of combustion. The study shows that the extraction of polyolefins from the mixed plastic waste the heat of combustion decreases for all kinds of wastes. Even after extraction of polyolefins, the heat of combustion of residues is still greater than those of different sorts of coals and these wastes could be effectively used for energy recovery from the plastic waste, by incineration.

The weighted sum method proposed in this paper for calculation of the heat of combustion with known composition of polymers in the mixture has found to be a good method to estimate the heat of combustion of the plastic waste without additional experimental determinations.

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