Specialty types of waste paper as an energetic commodity

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Abstract. The collection and recycling rate of paper and paper packaging material has been on a rise. From 2010 to 2016 in Czech Republic, the recycled amount of all paper went up by 32%, while the share of energy use in waste paper utilization decreased from 5.5% to 3.8%. However, not every paper and cardboard product can be recycled, and some are rejected from the recycling stream. Recycling specialty types of paper with other grades of recyclable paper is often not possible and their production is not high enough for their separate recycling to be feasible. If material utilization is not feasible then within the waste hierarchy the next best treatment is their energy utilization. Therefore, this article evaluates selected types of specialty paper for their energy content. They were silicone coated papers, polymer coated papers, and paper cores. For all samples proximate, elemental and calorimetric analyses were determined and based on them stoichiometric combustion calculations were performed. Silicon coated papers fared generally well having small to reasonable ash content 1–10% and net calorific value from 15.10 to 17.10 MJ kg^{-1} on dry basis. Polymer coated papers had ash content around 6% and net calorific value from value from 16.29 to 22.98 MJ kg^{-1} on dry basis. With the exception of paper cores and self-copying paper, all evaluated paper types could be recommended as a component in refuse derived fuels. The least suitable samples were paper cores with nearly 20% wt. of ash and net calorific value 12.45 MJ kg^{-1} on dry basis.

Key words: non-recyclable paper, proximate analysis, stoichiometric calculations, heating value.

INTRODUCTION

Paper has the advantage of being both made from renewable resources as well as being able to be recycled to yield its constituent fibres (Venditti et al., 2000). Recycling waste paper normally means using it as a secondary raw material for the production of new paper (Voronych et al., 2016). Waste paper is converted back to fibres that are usually mixed with a portion of virgin fibres and new paper is produced from this mixture (Ekvall & Finnveden, 2000). Papers with functional layers are generally more difficult to recycle and they may not be suitable to mix with the most prevalent grades of sorted paper (Hess et al., 2001).

Paper fibres cannot be recycled indefinitely, but they can go through the process four to six times (Villanueva & Wenzel, 2007). After the fibres have been recycled, they are gradually deformed and shortened until the last possible product is manufactured e.g. paper cores or moulded pulp. Papers that are difficult to recycle or unprofitable for

recycling often contain specific chemicals that hinder the recycling process. This category includes waterproof papers, coated with PE foil, waxed, with adhesive layer, etc. These types of paper are for example silicone coated papers, polymer coated paper, self-copying paper and paper tubing. These types of paper may come from recycling bins or from industry. Recycling of these types of paper with other grades of recyclable paper is not possible. They may be used, for example, to make insulation materials (Vochozka et al., 2016). However, in a paper sorting line these papers will mostly be discarded and will generally be used in a waste-to-energy plant (Leyssens et al., 2014).

Therefore, it is advisable to find the optimal uses for these types of paper, including the energy or construction industry (Mucahit & Sedat, 2009). It is also possible to compost paper (Saludes et al., 2008; Alvarez et al., 2009; Torkashvand, 2009) or produce biogas (Teghammar et al., 2010; Steffen et al., 2016; Rodriguez et al., 2017).

One of the transformation possibilities when using waste paper as an energy source is briquetting, ideally with biomass (Kers et al., 2010; Gado et al., 2014). Briquetting has some advantages over direct combustion technologies, mainly increasing the energy density, avoiding dust emissions during handling and often enabling cleaner combustion in small combustion devices (Malaťák et al., 2017). Co-combusting of unwanted paper with biomass-based materials can be expected to produce emission benefits because this type of paper contains a low percentage of sulphur and a very low nitrogen content compared to biomass fuels (Boavida et al. 2003; Salvador et al., 2004; Leyssens et al., 2014).

According to the Ministry of the Environment of the Czech Republic (2017) the recycled amount of all paper and cardboard in Czech Republic went up by 32% from 2010 to 2016, while the share of energy use in waste paper utilization decreased from 5.5% to 3.8%. However, the energy use still amounted to 17,000 t.

In the view of these facts, present paper deals with the issue of using specialty papers as a source of energy. Main aim of experimental testing was to determine the energy potential of chosen types of paper and evaluate the suitability for combustion as a standalone solid fuel or as an additive to other wastes or biomass in production of derived fuels.

MATERIALS AND METHODS

The types of specialty papers
Samples of various specialty papers were evaluated by elemental and proximate analysis and based on them stoichiometric combustion calculations were performed. The samples were obtained from companies producing these papers.

- Self-copying paper (SC) allows to make copies without the use of office equipment making it indispensable in some situations. It consists of two layers one of which is coated with microcapsules which burst under the pressure of a pen and colour the contacting layer.
- Greaseproof polymer coated paper (GPC) with low permeability for fats is used for food packaging or other packaging purposes. It is made from bleached sulphite and sulphate pulps.
- Silicone release paper (SR1) is white, smoothed, wood-free paper with one-sided silicone coating. It is used as release layer for sticky tapes, envelopes etc.

- Silicone release paper (SR2) was discarded release paper with the adhesive layer removed. It is coated on one side with a non-adhesive silicone layer. It is white, smoothed, wood-free paper.
- Silicone release paper (SR3) was the same as SR2 release paper with the adhesive sticker layer present.
- Silicone coated baking paper (BP) is a white, wood-free paper with silicone coating on both sides. It is designed to be hygienically safe at high temperatures, resistant up to 220 °C for single use.
- Packaging paper with one sided polyethylene coating (PEP) mostly used for production of paper bags. It is designed to protect packaged goods against dust, moisture and grease. According to the manufacturer, the unit weight of the paper is 70 g m^{-2} and of the coating 20 g m^{-2}.
- Paper cores (PC) are paper tubes of mostly cylindrical shape intended to hold strips of sheet material such as papers, foils, textiles, etc. The paper cores used were all spirally wound with layers glued together. There was a variety of sizes.

Material preparation and laboratory equipment

Prior to collection, all samples were kept indoors. After collection, the samples spent at least two weeks in laboratory climate (19–23 °C, relative humidity 40–55%). To produce analytical samples the papers were firstly shredded in an office paper shredder to 4 mm by 35 mm strips. The paper cores were shredded in a cutting mill Retsch SM100 on a 6 mm screen. For each material three 100 g samples were taken from the shredded materials and dried at 105 °C until constant weight for at least 24 hours to find the moisture in the original materials. The shredded samples were then milled in a rotor mill Fritsch Pulverisette 14 under 1 mm size to produce analytical samples.

The proximate analysis was performed in a thermogravimetric analyser (LECO TGA 701). The temperature programme first dried the samples at 105 °C to constant weight to determine the analytical moisture. Ash content was determined after burning the samples in oxygen at 550 °C until constant weight.

Elemental composition was analysed in an instrument LECO CHN628+S with helium as carrier gas to find carbon (C), hydrogen (H), nitrogen (N) and sulphur (S) contents. The analyser operates by analysing the flue gas from samples burned in oxygen. C, H and S are measured in infrared absorption cells; N is measured by a thermal conductivity cell. Oxygen was determined as difference from 100% of the sum of these elements and ash in dry state.

Gross calorific value was measured in an isoperibol calorimeter (LECO AC 600). The samples were pressed into pellets and burned in calorimetric bomb filled to 3 MPa. The reference temperature was 28 °C. The correction for the creation of nitric and sulphuric acid were not determined, otherwise the procedure and conversion to net calorific value was done according to ČSN ISO 1928:2010. At least three reliable results were acquired for all samples.

The results of the analyses were converted to the original state and to dry state of the materials.

Stoichiometric calculation

The theoretical amount of oxygen $O_{2,min}$ (m³ kg⁻¹) is based on the equation:

$$O_{2,\min} = V_m(O_2)\left(\frac{C}{M(C)} + \frac{H}{M(2\cdot H_2)} + \frac{S}{M(S)} - \frac{O}{M(O_2)}\right) \tag{1}$$

where $C, H, S,$ and O are contents of carbon, hydrogen, sulphur and oxygen in the sample (% wt.); $V_m(O_2) = 22.39$ m³ kmol⁻¹ is the molar volume of oxygen gas at normal conditions and $M(X)$ (kg kmol⁻¹) are molar masses of hypothetical species X that combine with O_2.

Where the theoretical amount of dry air L_{min} (m³_N kg⁻¹) is determined from the equation:

$$L_{\min} = O_{2,\min} \cdot \frac{100}{C_{atm}(O_2)} \tag{2}$$

where $C_{atm}(O_2) = 20.95$% vol. is volumetric concentration of oxygen in air.

The theoretical amount of dry flue gases $v_{fg,min}$ (m³ kg⁻¹) is based on the equation:

$$v_{fg,\min} = \frac{V_m(CO_2)}{M(C)}\cdot C + \frac{V_m(SO_2)}{M(S)}\cdot S + \frac{V_m(N_2)}{M(N_2)}\cdot N + \frac{C_{atm}(N_2)}{100}\cdot L_{\min} \tag{3}$$

where $V_m(X)$ (m³ kmol⁻¹) are the molar volumes of flue gas components; $C_{atm}(N_2) = 78.05$% vol. is the concentration of N_2 in air.

The theoretical amount of emission concentrations of $CO_{2,max}$ (m³_N kg⁻¹) is based on the equation:

$$CO_{2\max} = \frac{M(C)\cdot C}{V_m(CO_2)\cdot v_{fg,\min}}\cdot 100 \tag{4}$$

Volumetric amounts of combustion products:

$$v(CO_2) = \frac{V_m(CO_2)}{M(C)}\cdot C + \frac{C_{atm}(CO_2)}{100}\cdot L \tag{5}$$

$$v(H_2O) = \frac{2\cdot V_m(H_2O)}{2\cdot M(H_2)}\cdot H + \frac{V_m(H_2O)}{M(H_2O)}\cdot W \tag{6}$$

$$v_{N_2} = \frac{V_m(N_2)}{M(N_2)}\cdot N + O_{2,\min}\cdot\frac{C_{atm}(N_2)}{C_{atm}(O_2)} \tag{7}$$

where W (% wt.) is the moisture content in the fuel.

Adiabatic combustion temperature t_a (°C) assumes stoichiometric amount of combustion air and all released heat to be kept in the combustion products. Adiabatic combustion temperature is expressed as:

$$t_a = \frac{NCV}{v_{fg,\min}\cdot c_{sp}} \tag{8}$$

where NCV is the net calorific value of the fuel (kJ kg⁻¹); c_{sp} is the specific heat capacity of the flue gas (kJ kg⁻¹ K⁻¹).

Adiabatic combustion temperature in excess of air t_a (°C) is used to compare different fuels at the same combustion conditions given by excess air coefficient n, fuel enthalpy and combustion enthalpy. In this case, the chosen value was $n = 2.11$. The theoretical combustion temperature is given by the equation:

$$t_t = \frac{NCV + Q_p + Q_{air}}{V_{fg} \cdot c_{sp}} \tag{9}$$

where Q_p is the enthalpy of the fuel (kJ kg^{-1}); Q_{air} is the enthalpy of the combustion air (kJ kg^{-1}); V_{fg} is the volume of flue gas (m^3_N kg^{-1}). Since the specific heat capacity of flue gas is dependent on temperature, the combustion temperatures were determined using iterative calculation. Calculations start using expected temperature and are iterated until two consecutive results differ by less than 0.001 K.

RESULTS AND DISCUSSION

The results of the elemental analysis of the paper samples (see Table 1) show a high concentration of ash, especially in PC, where the ash content was 19.69% wt. in the dry matter. Such a large amount of ash significantly reduces the calorific value. High contents of ash have detrimental effect on the combustion process as well as the operation combustion plant, e.g. clogging of parts of incinerator by ash (Malaťák & Passian, 2011). The ash contents were generally high compared to biomass fuels, e.g. herbal biomass is 7.8% wt. (Vassilev et al., 2010; Thy et al., 2006) and pure wood biomass averaging 0.5% wt. (Tao et al., 2012). In this regard, only GPC. SR2, BP and PEP with ash content around 1% correspond roughly to the amount of ash in wood biomass (Gürdil et al., 2009). In general, high ash contents lead to high concentration of particulate matter in flue gas (Niu et al., 2016). With plant biomass it has been shown that the amount and composition of fly ash can be affected by operational parameters of a combustion device, e.g. the excess air amount (Bradna et al., 2016; Bradna et al., 2017).

Table 1. Proximate, elemental and calorimetric analysis of specialty papers, values are given in original and dry state

	W % wt.	Ash % wt.	C % wt.	H % wt.	N % wt.	S % wt.	O % wt.	GCV MJ kg^{-1}	NCV MJ kg^{-1}
Self-copying p. (SC)	5.30	13.36	37.34	4.96	0.17	0.05	38.82	14.33	13.12
dry state		14.11	39.43	5.24	0.18	0.06	40.99	15.13	13.99
Greaseproof p. (GPC)	6.16	1.04	42.79	5.48	0.02	0.06	44.45	16.49	15.14
dry state		1.11	45.60	5.84	0.02	0.07	47.36	17.57	16.29
Silicone p. 1 (SR1)	5.38	9.40	39.03	5.35	0.05	0.12	40.67	15.46	14.16
dry state		9.94	41.25	5.65	0.05	0.13	42.98	16.33	15.10
Silicone p. 2 (SR2)	6.51	0.59	42.01	5.48	0.00	0.04	45.37	16.54	15.18
dry state		0.63	44.93	5.87	0.00	0.04	48.53	17.69	16.41
Silicone p. 3 (SR3)	5.20	9.41	42.97	5.49	0.03	0.03	36.88	17.40	16.08
dry state		9.92	45.32	5.79	0.03	0.03	38.90	18.36	17.10
Baking p. (BP)	6.10	1.19	41.27	5.57	0.00	0.06	45.80	16.60	15.23
dry state		1.27	43.96	5.94	0.00	0.06	48.77	17.68	16.38
Packaging p. (PEP)	6.04	0.75	52.14	7.27	0.00	0.08	33.71	23.16	21.44
dry state		0.80	55.50	7.74	0.00	0.08	35.88	24.67	22.98
Paper cores (PC)	5.54	18.60	34.24	4.02	0.20	0.08	37.32	12.64	11.63
dry state		19.69	36.25	4.25	0.21	0.08	39.50	13.38	12.45
Office paper – dry state*		13.17	37.73	4.84	0.06	0.04	44.15	13.49	12.44
Printed paper – dry state*		21.31	36.86	4.49	0.09	0.03	37.22	13.77	12.80
Cardboard – dry state*		12.17	41.34	5.12	0.14	0.05	41.18	15.46	14.34

W – moisture; GCV – gross calorific value; NCV – net calorific value; *values from (Balada et al., 2016).

The moisture content of the samples in their original state is low and averages to 5.82% by weight. In biomass such low moisture can only be kept after it has undergone further treatment, for example pelletizing (Malaťák & Bradna, 2017).

The amount of carbon, as the main carrier of calorific value was mainly influenced by the high ash concentration. In samples of SC and PC, the carbon content in dry matter was less than 40% wt. On the other hand, in PEP it reached up to 55.5% wt.

The resulting heat and calorific values of the materials correspond to the proportion of combustible matter. Above all, a large amount of ash reduced the net calorific value of PC to 12.45 MJ kg^{-1} in dry matter. Samples of SC, GPC, SR1, SR2, SR3 and BP achieved net calorific values comparable to wood biomass (Vassilev et al., 2010). The highest net calorific value of all samples was found in PEP with 22.98 MJ kg^{-1} in dry matter. Compared to ordinary recycling paper grades, i.e. discarded office paper, printed magazine paper and cardboard (Balada et al., 2016), the presently studied papers, except for PC and SC papers, are more favourable for energy use both in heating value and ash content.

Sulphur content in a fuel has a direct effect on the formation of H_2SO_4 and therefore also on the service life of the combustion device. Someshwar (2015) shows that addition of sulphur containing fuel may lead to increase in HCl concentration in the flue gas. On the other hand, the presence of SO_2 in flue gas reduces the corrosion of metals by alkali metal halides (Paneru et al., 2103). Highest sulphur concentrations of 0.13% wt. in dry matter were determined in SR1. Sulphur concentrations in other samples correspond to wood (Vassilev et al., 2010). Relatively higher contents of nitrogen were found in PC. However, the maximum concentration of 0.21% wt. in dry matter corresponds to usual levels in plant biomass (Vassilev et al., 2010).

Table 2. Stoichiometric volumes of combustion air and gaseous products, adiabatic temperatures under stoichiometric and excess air conditions

	L_{min} m^3_N kg^{-1}	$v_{fg,min}$ m^3_N kg^{-1}	$v(CO_2)$ m^3_N kg^{-1}	$V(H_2O)$ m^3_N kg^{-1}	$V(N_2)$ m^3_N kg^{-1}	CO_{2max} % vol.	t_a °C	t_t °C
Self-copy. p. (SC)	3.33	3.30	0.69	0.75	2.60	21.00	2,310	1,068
Greaseproof p. (GPC)	3.77	3.74	0.79	0.84	2.94	21.24	2,350	1,089
Silicone p. 1 (SR1)	3.53	3.48	0.72	0.80	2.75	20.80	2,360	1,088
Silicone p. 2 (SR2)	3.67	3.64	0.78	0.84	2.86	21.39	2,410	1,115
Silicone p. 3 (SR3)	4.04	3.95	0.80	0.84	3.15	20.18	2,360	1,092
Baking p. (BP)	3.61	3.59	0.77	0.84	2.82	21.34	2,450	1,132
Packaging p. (PEP)	5.43	5.21	0.97	1.10	4.24	18.57	2,380	1,093
Paper cores (PC)	2.86	2.87	0.64	0.63	2.24	22.12	2,350	1,095

L_{min} – Stoichiometric volume of combustion air;, $v_{fg,min}$ – Stoichiometric volume of flue gas; $v(CO_2)$ – Stoichiometric volume of CO_2; $v(H_2O)$ – Stoichiometric volume of H_2O; $v(N_2)$ – Stoichiometric volume of N_2; CO_{2max} – Stoichiometric concentration of CO_2 in dry flue gas; t_a – Adiabatic temperature in stoichiometric combustion; t_t – Adiabatic temperature in combustion with excess air.

The elemental contents affect not only the calorific value, but also the behaviour during combustion. In particular, high content of fuel oxygen will affect the stoichiometric consumption of combustion air and the total amount of flue gas produced (Malaťák et al., 2017). The resulting stoichiometric analysis (see Table 2) shows large differences in these properties, especially between PEP and PC. These samples show higher stoichiometric need for combustion air and therefore higher quantities of flue gas

compared to other samples. The values determined for PEP and SR3 are comparable to e.g. wood biomass (Malať ák et al., 2013). On the other hand, PC was on the opposite end with low stoichiometric volumes due to low carbon content. These large differences in stoichiometric values of air consumption and flue gas production affect the selection and adjustment of the combustion plant depending on their representation in the fuel stream. Large differences were also found at the maximum carbon dioxide concentration, where PEP is at 18.57% vol. CO_2 and PC 22.12% vol. CO_2. The resulting stoichiometric parameters have a major impact on the actual efficiency of combustion plants (Malať ák & Bradna, 2017) and on the total exhaust gas concentration. Similar results were obtained when evaluating the potential of waste biomass in (Brunerová et al., 2017).

Table 2 gives the results of the adiabatic combustion temperatures under stoichiometric and excess combustion air conditions. The highest adiabatic stoichiometric combustion temperature was determined for BP and SR2 samples at 2,450 °C. Since this temperature is limited to stoichiometric amount of air, the adiabatic combustion temperature with excess air is more useful for practical comparison. The highest and lowest values were determined for BP ($t_t = 1,132$ °C) and SC ($t_t = 1,068$ °C), respectively. Higher combustion temperature has a positive effect on complete combustion and therefore also on lowering CO emissions (Eskilsson et al., 2004), but on the other hand higher combustion temperature increases NO_x emission production (Díaz-Ramírez et al., 2014). The problem of lower combustion temperatures may in practice be amended by co-combustion. There have been positively evaluated tests of firing pellets made of cardboard paper and wood sawdust (Leyssens et al., 2014) where the results showed that such pellets lead to a very good combustion and boiler efficiency. Boavida et al. (2003) showed that co-combustion of paper with coal and plastics improves the process of combustion increasing the combustion temperature and improving emissions of CO, CO_2 and NO_x.

CONCLUSIONS

The use of specialty types of waste paper for energy recovery e.g. in refuse derived fuel can be recommended as long as material recovery is not effective. In practice, according to EN 15359:2011 most of tested types of paper are suitable for addition to refuse derived fuels up to class 3 without detrimental influence judged by their net calorific value above 15 MJ kg^{-1} and ash content sufficiently low. Especially good were silicone release paper 2, greaseproof paper, baking paper and polymer coated packaging paper which had both high calorific value and low ash content. On the other end were paper cores and self-copying paper which have calorific value corresponding to class 4, although they could still be mixed for example with biomass to produce class 3 refuse derived fuel. Sufficiently low chlorine content is another demand on refuse derived fuel. As chlorine content was not analysed low level was only assumed, although in theory it could pose some issue with chlorine-bleached papers.

The limiting factor for direct incineration, especially in smaller combustion devices, is the high percentage of ash in some of the specialty paper types. In pressed fuels made solely from paper the unburned remains often effectively prevent the combustion of the inside portion of the pellet or briquette. A large amount of ash, the highest being 19.69% wt. in the dry matter, can cause major problems during combustion, e.g. fouling of the combustion equipment, failure of the fuel to be burn out, etc. For a complete assessment

of the effect of ash on combustibility, it would be necessary to determine its composition and the ash fusion temperatures as e.g. some inorganic species will react beneficially with components of flue gas or, on the other hand, some will melt below combustion temperatures causing the necessity of increased maintenance. To improve combustion properties, it is advisable to burn paper together with plant biomass.

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