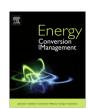
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Replacing conventional fuels in USA, Europe, and UK with plastic pyrolysis gases - Part I: Experiments and graphical interchangeability methods

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ABSTRACT

The development of effective recycling methods for plastic wastes is critical in terms of resource security and environmental conservation. In this work, we focused on the gaseous pyrolysis products of plastic wastes as alternatives to natural gas (NG) and propane. The pyrolysis of polyethylene (PE), polypropylene (PP), polystyrene (PS), poly(vinyl chloride) (PVC), poly(ethylene terephthalate) (PET), and their mixtures was carried out under isothermal conditions at 500, 700, and 900 °C. The potential for replacing NG and propane with the pyrolysis gases was assessed by graphical interchangeability methods for the first time. The pyrolysis gas compositions obtained from the mixed plastics and HCl-scrubbed gas from the PVC were deemed suitable NG alternatives in Europe and the USA. In addition, PE and PP pyrolysis gases showed the potential for replacing propane in the USA, whereas gases from PET showed the least interchangeability. Thus, the graphical interchangeability methods are promising for evaluating the potential of plastic waste pyrolysis gases as alternatives to NG.

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1. Introduction

Because of their outstanding properties, plastics are being widely used by an increasing number of people [1,2]. Consequently, in the second half of the 20th century (until 2013), the production and processing of plastics increased annually by ~9% on average, and there is no doubt that this increase will continue in future. In 2013, the production volume of plastics was almost 300 million tons, and is expected to reach 400 and 700 million tons by 2020 and 2050, respectively. China is the main producer of plastics (thermoplastics and polyurethanes) with a 24.8% share, followed by Europe (\sim 20%) and NAFTA (19.4%) [3,4]. The increased production has led to problems associated with waste plastics, with the EU [5] and USA [6] producing ~25 and 32 million tons of waste plastic per year (2012). In fact, in the USA, the share of plastics in municipal waste reached 13% in 2012, which is a dramatic increase compared to the 1% share in 1960 [6].

There are four possible methods of dealing with plastic waste: mechanical recycling, chemical recycling, use in power generation (incineration), and landfill disposal. The latter is the least favorable

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http://dx.doi.org/10.1016/j.enconman.2016.08.055 0196-8904/© 2016 Elsevier Ltd. All rights reserved. option because of the extremely long time required for waste degradation, with some wastes requiring hundreds of years [7,8]. Despite this, ~38% (9.6 million tons; worth 8 billion €) of waste plastic is subjected to landfill disposal in the EU [3], while 26% is recycled and 36% is used for power generation [4]. In the USA in 2011, the processing of plastic wastes by landfill disposal, recycling, and use in power generation reached 83.5%, 6.5% (9% in 2012), and 9.5%, respectively, while the consumption of plastics as an alternative fuel for cement production was 0.5% [6,9].

Pyrolysis, the thermal decomposition of organic materials in the absence of oxygen or air [10-12], can be used in chemical recycling and power generation [13-15]. This process is not oxidizing, in contrast to combustion [16,17]. Pyrolysis involves the transformation of specific raw materials (e.g., wastes) into gas, solid, or liquid products usable for power generation [18-24]. This process is also used, for instance, to transform various types of biomass and biological waste into bio-fuels [25,26] which currently represents a very significant transportation fuel and contributes to mitigating CO₂ emission [27,28]. Pyrolysis is a suitable alternative to incineration and landfill disposal because, after acquiring the energetically valuable products, the volume of the wastes is decreased, and the wastes' environmental impact is substantially reduced [29]. Moreover, some pyrolysis gases have high calorific values

Nomenclature correction factor of individual hydrocarbons Subscripts ď relative density component i Weaver coefficient F_i gas pyrolysis gas \hat{H}_S heat of combustion, (MJ m_N^{-3}) auxiliary factor 1 K_1 **Abbreviations** auxiliary factor 2 K_2 **EASEE** European Association for the Streamlining of Energy $S_{(W)}$ Weaver flame speed factor Exchange $S_{L,N}$ maximum combustion speed, $(cm s^{-1})$ EU m. EU mix gas type factor 1 $u_{(D)}$ IGU International Gas Union gas type factor 2 $\nu_{(D)}$ JP m. Japan mix V_t stoichiometric volume of combustion air, $(m_N^3 m_N^{-3})$ LPG liquefied petroleum gas WN Wobbe number, (MJ m_N^{-3}) NG natural gas WN' corrected Wobbe number, (MJ m_N^{-3}) PE polvethylene PET poly(ethylene terephthalate) Greek symbols рp polypropylene PS volumetric fraction, (%vol.) polystyrene Ω poly(vinyl chloride) density, $(kg m_N^{-3})$ **PVC TPA** terephthalic acid US m. USA mix

[30–34], and can help reduce natural gas (NG) consumption. Consequently, it is surprising that pyrolysis is only used for 0.3% of waste plastics [5].

Based on numerous studies, gaseous pyrolysis products are known to generally contain alkanes and alkenes (C_1-C_6) , hydrogen, carbon dioxide, and carbon monoxide in specific percentages (including zero) depending on the input materials and process conditions [35-37]. This article is focused on the feasibility of replacing traditional gaseous fuels with the gaseous products from the pyrolysis of the principal waste plastics (polyethylene (PE), polypropylene (PP), polystyrene (PS), poly(vinyl chloride) (PVC), poly(ethylene terephthalate) (PET)) and their mixtures. The potential for replacement is evaluated in terms of "interchangeability," which is defined as the ability to replace one gaseous fuel with another without requiring substantial design or conceptual changes to the burners or incinerators and without seriously impacting the efficiency, power output, operational safety, and emissions of the process [38-40]. The issue of interchangeability is not a new one, and a significant amount of work has been done in the last century in the USA, Europe, and the UK. In these cases, interchangeability was largely studied because of frequent transitions from coal gas to NG and vice versa [41]. Interchangeability was largely considered at the regional level, and the development of respective standards was thus quite independent [42]. However, these days, interchangeability has become quite practical on the global scale due to the depletion of extraction sites and increased international trade of NG and liquefied natural gas (LNG) [43,44].

Interchangeability methods are very important for evaluating fuel substitution, since different fuels possess different qualitative, physical, and chemical properties, and all incinerators are designed to be used under specific gas parameters. The interchange of two gases having different properties can lead to problems such as unstable incineration processes, incorrect flame dynamics, an increase in combustion air volume, self-ignition of the gas, and the increased emission(s) of CO, NO_x, C_xH_y, and SO₂ [45]. These undesirable effects can impact the operational efficiency, durability, and safety of the device, and cause malfunctions [46].

The pyrolysis gases from various materials, including wood chips [47], tires [48], lignin [49], PP and PS [50], plastic waste [51,52], tire/coal [53], sewage sludge [54], and paper [35], have been investigated by many authors. However, the literature does not discuss the potential for the substitution of fuels with gases

produced by the pyrolysis of plastics or plastic mixtures, which is surprising, considering the scale of the waste plastics problem and the suitability of pyrolysis as a treatment method.

Therefore, the objective of this study is to fill this gap by providing a standard technique for evaluating interchangeability and reporting new findings that may help reduce the volume of waste plastics and the consumption of NG. Since this is a broad issue, it is divided into two articles. The first part describes the substitution potential of pyrolysis gases based on the Wobbe number (WN), which represents the preservation of the power input of the incinerator upon changing the fuel gas, along with three graphical methods, each applicable in different parts of the world (the USA, Europe, and the UK).

2. Experimental

2.1. Materials

The five most widely used plastics, PET, PP, PE, PS, and PVC, were examined. All samples were acquired from Sigma Aldrich Inc. (USA), except for PET, which was prepared from bottle PET. The plastics were processed by pyrolysis, both separately and as mixtures. The base C, H, O, and Cl analysis of the individual input samples was done by the incineration method using a Micro Corder JM10 analyzer provided by J-Science Lab Co, Ltd. (Kyoto, Japan). The results are provided in Table 1. Data from the literature are provided for comparison in the right hand side of the table.

The mixtures of individual components were based on realistic waste plastic compositions in Japan [56], Europe [57], and the USA [9] (Table 2).

2.2. Pyrolysis experiments

All pyrolysis experiments were carried out using a tube reactor, as described in previous studies [58]. A minor difference was that the steam generator was removed and no catalyst was fixed in the lower furnace. In addition, when PVC was mixed in the sample, a 0.1 M NaOH trap was connected before the gasbag. The samples (500 mg) were filled in the hole-punched sample holders connected to the top of the reactor. The air inside the reactor was replaced with a helium flow of 200 mL min⁻¹, and the upper part

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