Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

An alternative method to determine the share of fossil carbon in solid refusederived fuels – Validation and comparison with three standardized methods



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ARTICLE INFO

Keywords: Fossil carbon Biomass content Refuse-derived fuels Balance Method Radiocarbon Method Selective Dissolution Method

ABSTRACT

Today different types of wastes are used as refuse-derived fuels (RDF) either in waste-to-energy plants or as fuel substitutes in energy-intensive industrial processes. In order to quantify their greenhouse-gas relevance (fossil carbon content), reliable and practical analytical methods are required, which allow differentiation between biogenic and fossil organic carbon. In the present paper, an alternative method to determine the fossil share in RDFs is examined and validated. The so-called "adapted Balance Method" (aBM) is applied to three different RDFs and the results are compared to three standardized methods, namely the Radiocarbon Method (14C-Method), the Selective Dissolution Method (SDM), and the Manual Sorting Method (MS). The aBM is based on the distinctly different elemental composition of water-and-ash-free biogenic and of fossil matter (TOX_{BIO} and TOX_{FOS}). Within the study, these compositional data are derived by manual sorting of the RDFs. The results show that the values obtained by the aBM are in excellent agreement with the results of the ¹⁴C-Method (considered as reference method). Mean deviations between the two methods of -0.9 to +1.9% absolute for the share of fossil carbon are found which are statistically insignificant. High trueness and reliability of the aBM can be expected, independent of the RDF type. In contrast, the reliability of the other standardized methods (SDM and MS) appears to strongly depend on the type and composition of the RDF. The results further indicate that the generation of RDF-specific data on TOX_{FOS} is important for the aBM if significant shares of polymers with comparably high oxygen content might be present in the RDF and if low uncertainties of the results (<3% relative) are required. The findings demonstrate that the alternative method has advantages compared to standardized methods with respect to reliability and/or costs.

1. Introduction

The utilization of waste materials as a secondary energy resource is increasing throughout most of the world. Today many wastes and waste fractions are used for energetic applications, a practice supported by national governments and the European Commission [1]. Policies on waste management and emission reductions as well as on the energy market have been established. The implementation of the Kyoto-protocol further stimulates the use of biomass-containing alternative fuels (e.g. Renewable Energy Directive [2], Directive on greenhouse gas emission trading [3], Landfill Directive [4], Chinas national emission trading System [5], Egyptian coal regulations [6]). Refuse-derived fuels (RDF) are being used in both, waste-to-energy plants and as a fuel substitute in energy-intensive industries. RDF in industrial processes are

https://doi.org/10.1016/j.fuel.2017.12.076

Received 17 August 2017; Received in revised form 15 December 2017; Accepted 18 December 2017 Available online 20 March 2018

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Abbreviations: A, ash content; aBM, adapted Balance Method; BM, Balance Method; C&I, commercial & industrial waste; MS, Manual Sorting method; MSW, municipal solid waste; n, number of samples; RDF, refuse-derived fuel; R^2 , correlation coefficient; SD, standard deviation; SDM, Selective Dissolution Method; SRF, solid recovered fuel; *TC*, total carbon in the water-free sample; TH, total hydrogen in the water-free sample; *TTK*_{wf}, total inorganic content of the respective element in the water-free ash; TN, total nitrogen in water-free sample; *TO*, total organic carbon; *TOH*, total organic hydrogen; *TON*, total organic nitrogen; *TOO*, total organic oxygen; *TOS*, total organic culphur; *TOX*_{BLDF}, total organic content of the respective element in the water-free sample; *TOS*, total organic culphur; *TOX*_{RDF}, total organic content of the respective element in the water-free sample; *TOS*, total organic culphur; *TOX*_{RDF}, total organic content of the respective element in the water-free sample; *TX*_{wf}, total organic content of the respective element in the water-free social carbon; *TOX*_{RDF}, total organic content of the respective element in the water-and-ash-free biogenic matter; *TOX*_{RDF}, total organic content of the respective element in the water-free sample; *X*_{wf}, total organic content of the respective element in the water-free sample; *X*_{wf}, biogenic mass fraction on water-free sample; *X*_{wf}, total content of the respective element in the water-free sample; *X*_{wf}, biogenic mass fraction on water-free sample; *X*_{wf}, biogenic mass fraction on water-free basis; *x*_{B,wf}, biogenic mass fraction on water-free basis; *x*_{B,wf}, fossil mass fraction on water-free basis; *x*_{F,TC}, share of fossil carbon; ¹⁴C-Method, Radiocarbon Method; %abs, absolute percentage; %rel, relative percentage

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utilized to save costs for fuels, to reduce natural resource consumption and to lower the amount of greenhouse gas emissions associated with the production process [7–9]. Due to the CO_2 emission trading scheme in place in Europe [10], lowering fossil CO_2 emissions by utilizing biomass-containing RDFs is of economic relevance for cement manufacturers. CO_2 emissions stemming from the biogenic matter are regarded as carbon neutral, whereas CO_2 from fossil matter is climaterelevant.

The prime example for the utilization of RDFs is the cement industry, which represents one of the most material-, energy-, and CO₂intensive industries [11,12]. Within the European Union, more than 34% of thermal energy demand in the production process of cement plants is already provided by RDFs [13]. In some European countries the substitution rate of primary energy carriers has already reached a level of above 50% – e.g. Austria 76.1% (2015) [13], Germany 64.6% (2015) [14], Belgium 60% (2011) [15], Switzerland 53.7% (2014) [16].

Compared to fossil fuels, RDFs exhibit a much broader variation in composition, which strongly depends on the waste utilized for RDF production. Variations can easily appear due to different types of commercial and industrial waste, municipal solid waste, waste collection scheme, or seasonal variations in waste generation. In order to check the quality of solid RDFs with respect to their composition (e.g. calorific value, biomass content, heavy metal contents) and their associated environmental impacts, reliable and practical methods are required. Both, waste management companies generating RDFs and industries utilizing these fuels require tools for reporting and documentation purposes. With respect to the determination of the climaterelevant share in solid RDFs, three methods are described in the standard EN 15440:2011: the Manual Sorting method (MS), the Selective Dissolution Method (SDM), and the Radiocarbon Method (¹⁴C-Method) [17]. Furthermore, the Balance Method (BM) has recently been published in the Standard ISO 18466:2016 [18] and has also been recognized by the United Nations Framework Convention on Climate Change (UNFCC) as an approved methodology to determine the fraction of fossil carbon in waste [19]. Table 1 summarizes the available methods with respect to their applicability to solid RDF samples ("prior combustion") and/or to gaseous samples ("post-combustion"). Possible outcomes (parameter) of the methods are listed.

Only the BM and ¹⁴C-Method are applicable for a "post-combustion" analysis in the flue gas, whose representative sampling is considerably easier compared to the solid waste material, as the latter is much more heterogeneous. The BM is able to monitor the fossil share in the input of waste-to-energy plants in real-time [20-22]. This is possible by using operating data of the waste-to-energy plant, usually making additional sampling and analysis campaigns redundant. Yet, characterizing the waste and RDFs prior to combustion requires solid samples to be analyzed. This is possible by means of the MS, SDM, and ¹⁴C-Method and by means of a recently adapted version of the BM (adapted Balance Method). The adapted Balance Method (aBM), which relies on the analysis of the elementary composition of the RDF in the laboratory, has recently been successfully applied to artificially produced RDFs [23,24]. Some methodological and economic benefits compared to the laboratory-based standardized methods (SDM, MS, ¹⁴C-Method) have been identified so far; i.a.:

- In contrast to SDM and MS, the uncertainties of the aBM results are statistically derived. They are propagated from the uncertainties of the input parameters [24]. By comparison, the methodological constraints of the SDM (unselective dissolution) can lead to significant in-accuracies which are difficult to detect and quantify [1,25]. As MS is greatly affected by the knowledge of the sorting person and available facts about the waste compounds, the uncertainties of this method can hardly be calculated [26,27]. Despite the high analytical precision of the ¹⁴C-Method, uncertainties for this method are introduced by the choice of a ¹⁴C-reference value [28,29].
- The aBM is far less time- and cost-intensive than MS or the 14C-

Table 1

Available methods to determine the fossil share in solid refuse-derived fuels; indicating possible output parameter of the methods and the applicability prior to combustion or as a post-combustion method (measurement in the flue gas).

Method applicable	Parameter			
	Fossil/ biogenic <u>mass</u> share in wt%	Fossil/biogenic - share of <u>carbon</u> in wt% - share of CO₂- emissions in wt% - CO ₂ -emission factor	Ratio of <u>energy</u> from fossil/ biogenic sources in %	<u>Plastic</u> content in wt%
prior-combustion	MS, SDM, aBM	(MS) ³ , SDM ¹ , ¹⁴ C-Method ² , aBM	(MS) ³ , SDM ¹ , ¹⁴ C-Method ² , aBM	MS, SDM, aBM
post-combustion	BM	¹⁴ C-Method ² , BM	¹⁴ C-Method ² , BM	BM

aBM – adapted Balance Method, BM – Balance Method (ISO 18466:2016), SDM – Selective Dissolution Method (EN 15440:2011), MS – Manual Sorting (EN 15440:2011), 14 C-Method – Radiocarbon Method (EN 15440:2011).

 1 SDM: Carbon/energy-based parameters (share of carbon, CO₂-emission factors, ratio of energy) can only be derived when the carbon/energy content is known in the RDF and in the dissolution residue.

 2 $^{14}\text{C-Method:}$ Energy-based parameters (ratio of energy, energy-related CO_2-emission factor) can only be derived when the energy content is known in the fossil and biogenic matter.

 3 MS: Carbon/energy-based parameters (share of carbon, CO₂-emission factors, ratio of energy) can only be derived when the carbon/energy content is known in the fossil and biogenic matter in each compound.

Method. The ¹⁴C-Method can only be employed by a limited number of laboratories which are equipped with the appropriate instruments (around 64 laboratories in Europe) [30]. Costs for aBM and SDM are expected in a similarly lower range than MS and ¹⁴C-Method [24,30].

- High trueness and precision of the aBM was found recently when defined mixtures of biogenic (e.g. cardboard, paper) and fossil materials (plastics) were investigated (deviations from the theoretical value below 4.5% rel; precision of $\pm 3\%$ rel) [23]. This is better than reported for SDM and MS [27,31,32].
- Contrary to most standardized methods, the aBM can provide a range of parameters with reference to the fossil/biogenic content in RDFs, including the key parameters identified in Table 1. Different parameters may be required depending on the interested party (RDF producer, RDF user, authority, etc.) or the reporting obligations (CO₂-emissions, energy produced, etc.). Thus, when choosing a method one needs to be aware of the potential parametrical limitations of these methods. For example, the ¹⁴C-Method does not provide information on the biogenic or fossil mass share in the RDF, whereas from sorting (MS) the share of fossil carbon cannot directly be derived. After sorting, additional information or analyses on the carbon content of the different RDF compounds are necessary. The same applies also to the share of biogenic energy, which is only directly derivable from aBM results.

Finally, during a recent interlaboratory comparison, the SDM was applied to determine the ratio of biogenic carbon in RDF samples (output of mechanical biological plant). The results showed that from the values generated by 12 laboratories only 30–70% were within the tolerance limits of two standard deviations [33]. This rather poor result again indicates that routinely applied methods can be unsatisfactory and that the development of alternative approaches is justified.

The objective of the study presented is to examine the aBM as an alternative method for determining the fossil carbon (and mass) share in solid RDFs. Following the promising results with artificially produced

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