



Predicting gaseous emissions from small-scale combustion of agricultural biomass fuels



S. Fournel^{a,b,*}, B. Marcos^a, S. Godbout^b, M. Heitz^a

^a Department of Chemical and Biotechnological Engineering, Université de Sherbrooke, 2500 Université Boulevard, Sherbrooke, Quebec J1K 2R1, Canada

^b Research and Development Institute for the Agri-Environnement, 2700 Einstein Street, Quebec City, Quebec G1P 3W8, Canada

HIGHLIGHTS

- A prediction model of gaseous emissions from biomass combustion was developed.
- Gibbs energy minimization for chemical equilibrium characterization was applied.
- The model was validated and calibrated with data from literature.
- The model simulated the combustion of four dedicated energy crops.
- CO, CO₂, NO_x, SO₂ and HCl predictions were accurate.

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ABSTRACT

A prediction model of gaseous emissions (CO, CO₂, NO_x, SO₂ and HCl) from small-scale combustion of agricultural biomass fuels was developed in order to rapidly assess their potential to be burned in accordance to current environmental threshold values. The model was established based on calculation of thermodynamic equilibrium of reactive multicomponent systems using Gibbs free energy minimization. Since this method has been widely used to estimate the composition of the syngas from wood gasification, the model was first validated by comparing its prediction results with those of similar models from the literature. The model was then used to evaluate the main gas emissions from the combustion of four dedicated energy crops (short-rotation willow, reed canary grass, switchgrass and miscanthus) previously burned in a 29-kW boiler. The prediction values revealed good agreement with the experimental results. The model was particularly effective in estimating the influence of harvest season on SO₂ emissions.

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1. Introduction

The awareness and concern about depletion of fossil fuels, energy dependency and climate change have engendered the necessity to develop reliable, affordable and renewable energy sources. In this context, modern use of biomass is particularly attractive for reducing society's reliance on petroleum products and greenhouse gas emissions. In addition, biomass is readily available worldwide, especially in rural areas (Demirbas, 2005; Saidur et al., 2011). Therefore, the interest in using farm-grown solid biofuels as substitutes to fossil fuels for energy production has increased. Short-rotation coppices, such as willow and hybrid poplar, and perennial rhizomatous grasses, such as reed canary

grass, switchgrass and miscanthus, have been considered as the most promising dedicated energy crops (Lewandowski et al., 2003; McKendry, 2002).

Direct combustion is the most widely applied conversion method for biomass and still a relevant technique to provide heat for industries, homes and farm facilities where biomass boilers can replace oil- and gas-fired furnaces (Demirbas, 2005; Saidur et al., 2011). However, burning non-woody fuels in small-scale combustion systems stays a challenge in terms of ease of use, energy efficiency and atmospheric pollutants (Carvalho et al., 2013; Verma et al., 2011). Compared to wood, agricultural products usually contain less carbon (C) and hydrogen (H) and have higher contents in ash and critical inorganic elements such as nitrogen (N), sulfur (S) and chlorine (Cl). Concentrations of C and H impact the heating value of the fuel. Ash contributes to particulate matter emissions and to operational problems such as fouling and slagging which may disturb the combustion process, reduce boiler efficiency and lead to higher levels of compounds from an incomplete

* Corresponding author at: Research and Development Institute for the Agri-Environnement, 2700 Einstein Street, Quebec City, Quebec G1P 3W8, Canada. Tel.: +1 418 643 2380x611; fax: +1 418 644 6855.

E-mail address: sebastien.fournel@irda.qc.ca (S. Fournel).

combustion including carbon monoxide (CO) and some hydrocarbons. High amounts of N, S and Cl in agricultural biomass increase the emission of nitrogen oxides (NO_x), sulfur dioxide (SO₂) and hydrogen chloride (HCl), respectively. These gases can cause respiratory problems, acid rain, indirect increase of greenhouse effect in the atmosphere through ozone (O₃) formation, deposits and corrosion (Obernberger et al., 2006; van Loo and Koppejan, 2008; Werther et al., 2000).

Nevertheless, there is still insufficient information available regarding the combustion-related emissions of dedicated energy crops in current small-scale heating systems and their suitability to be burned in accordance of existing threshold values (Carvalho et al., 2013; Verma et al., 2011). Besides, the numerous varieties of solid biofuels complicate the establishment of reference values since previous studies (Obernberger et al., 2006; Vassilev et al., 2010) have revealed that gaseous emissions from biomass combustion differ significantly according to the properties of the fuel burned (e.g. chemical composition). From an experimental point of view, considering all the biomass possibilities would become a laborious and expensive work.

To overcome this situation, thermodynamic equilibrium models can become useful engineering tools to assess how fuel characteristics influence the exit gas composition (Baratieri et al., 2008; Puig-Arnavat et al., 2012; Sreejith et al., 2013). Actually, when the chemical composition of biomass and the equilibrium temperature are specified, thermodynamic models can simply predict the resulting emissions (Ranzi et al., 2011). The composition of the gas produced at thermodynamic equilibrium can be estimated using different approaches: kinetic/dynamic models (Gøbel et al., 2007; Ranzi et al., 2011), equilibrium constants (Gautam, 2010; Jarungthammachote and Dutta, 2007; Melgar et al., 2007; Zainal et al., 2001) or Gibbs energy minimization. The key advantage of the latter is that it has a more general application with predictive capability without requiring an extended set of data to train the model compared to kinetic/dynamic models and that it does not necessitate the selection of appropriate chemical reactions allowing the formation of products compared to direct chemical equilibrium computation (Baratieri et al., 2008; Néron et al., 2012).

Minimization of the Gibbs free energy is mainly used for determining the chemical composition of reactive multicomponent closed systems under thermodynamic equilibrium (Néron et al., 2012). Therefore, a global minimum of the Gibbs free energy coincides with the stable equilibrium solution under constant temperature and pressure. The equilibrium problem is then solved as an optimization problem of a non-linear constrained system which must satisfy the restrictions of non-negative number of moles and stoichiometry (Rossi et al., 2011). Lagrange multiplier method is generally used to compute this constrained optimization problem (Baratieri et al., 2008; Jarungthammachote and Dutta, 2008). Even though this theoretical approach has some inherent limitations (necessity of establishing a list of chemical species expected in the product mixture and only suitable for processes whose duration is usually quite long with respect to the reaction timescale), it was judged as relevant for the prediction of product compositions in several operations and chemical processes such as biomass gasification (Altafini, 2003; Baratieri et al., 2008; Jarungthammachote and Dutta, 2008; Kalina, 2011; Néron et al., 2012; Puig-Arnavat et al., 2012; Rossi et al., 2009, 2011; Sreejith et al., 2013). This method can also be used for direct combustion of biomass (de Souza-Santos, 2010; Jarungthammachote and Dutta, 2008), but the applications have been only tested for coal and wood so far (Ross et al., 2002).

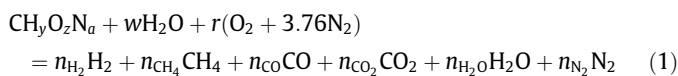
The objective of the present study is to develop a simple model for the prediction of gaseous emissions from small-scale combustion of agricultural biomass fuels. The model, once validated with published experimental and modeling results of other authors also

using thermodynamic equilibrium calculations, will provide the opportunity to rapidly assess the potential of dedicated energy crops to be burned in a sustainable way (respect of existing gas threshold values) as well as the influence of fuel properties.

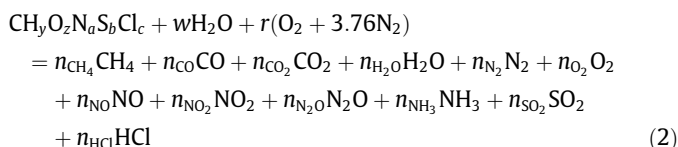
2. Methods: model development

2.1. Conversion processes, input data and assumptions

To develop the model, the chemical formula of biomass is defined either as CH_yO_zN_a or CH_yO_zN_aS_bCl_c. The former corresponds to a woody biomass with negligible S and Cl contents and is involved in the gasification reaction for simulation comparison with other gasification models as described in Section 3.1. The second term characterizes a general solid biofuel and is used in the combustion reaction for prediction of combustion-related emissions (Section 3.2). Both global gasification and combustion processes can be expressed by Eqs. (1) and (2), respectively:



and,



where *y*, *z*, *a*, *b* and *c* are respectively the mass fractions of hydrogen, oxygen, nitrogen, sulfur and chlorine per atom of carbon in the biomass, *w* is the amount of moisture per kmol of feedstock, *r* is the quantity of oxygen (O₂) supplied to the process per kmol of feedstock and *n_i* are the numbers of mole of species *i* (kmol). The molar biomass composition and the molar moisture quantity are obtained from standard ultimate and proximate analyzes. The variable *r* is calculated from the air-to-fuel ratios measured experimentally. All inputs on the left-hand side of Eqs. (1) and (2) are supposed entering the reaction system at 25 °C and 1 atm. On the right-hand side, the stoichiometric coefficients *n_i* are unknown and the model ultimately consists in evaluating them.

The resolution of the mole numbers *n_i* using the Gibbs free energy minimization approach is based on the following assumptions:

- The processes are long enough to achieve thermodynamic equilibrium. The degree of error introduced by this assumption is acceptable and its applicability is confirmed in literature (de Souza-Santos, 2010; Jenkins et al., 2011; Ragland and Bryden, 2011). Mass flows and average properties of input and output streams in the reaction system are therefore considered remaining constant.
- The products taken into account in Eqs. (1) and (2) are the main compounds formed during gasification and combustion processes (Jarungthammachote and Dutta, 2008; van Loo and Koppejan, 2008). Other gases such as hydrocarbons (C_xH_y) other than methane (CH₄) were found to be only present in insignificant fractions by some thermodynamic models (Baratieri et al., 2008; Ross et al., 2002). Consequently, they are assumed negligible.
- The gaseous products are all assumed to behave as ideal gases. This leads to insignificant errors because gasification and combustion reactions are usually conducted at high temperature (700–1000 °C) and low pressure (1 atm) (Gautam, 2010).
- All C and N in biomass are converted into gaseous form. Mass balances on C and N realized during combustion experiments by Brassard et al. (2014) revealed good agreement with

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