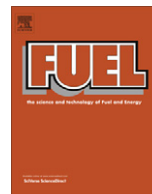


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Trace element behavior in the fluidized bed gasification of solid recovered fuels – A thermodynamic study

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HIGHLIGHTS

- ▶ A thermodynamic database on trace elements, combining data from several databases.
- ▶ Stepwise introduction of fuel ash components to eliminate unrealistic compounds.
- ▶ Validation of modeling data against experimental data from 1 MW_{th} pilot-gasifier.
- ▶ Critical review and comparison of own results against results from the literature.

ARTICLE INFO

Article history:

Received 28 May 2012

Received in revised form 2 October 2012

Accepted 3 October 2012

Available online 27 October 2012

Keywords:

Trace elements

Gasification

Thermodynamic equilibrium

Modeling

Solid waste

ABSTRACT

Gasification of biomass and recycled fuels is of particular interest for the efficient production of power and heat. Trace elements present as impurities in the product gas should be removed very efficiently. The objective of this work has been to develop and test thermodynamic models for the reactions of trace elements with chlorine and sulfur in the gasification processes of recycled fuels. In particular, the chemical reactions of trace elements with main thermochemical conversion products, main ash components, and bed and sorbent material are implemented into the model. The possibilities of gas cleaning devices in condensing and removing the trace element compounds are studied by establishing the volatilization tendency of trace element compounds in reducing gases. The results obtained with the model are compared with the measured data of trace elements of gasification experiments using solid recovered fuel as feedstock. Some corresponding studies in the literature are also critically reviewed and compared. The observed discrepancies may be attributed to differences in thermodynamic databases applied and experimental arrangements. The method of removing gaseous trace elements by condensation is already in use in the 160 MW_{th} waste gasification plant in Lahti, Finland.

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1. Introduction

Gasification of biomass and recycled fuels is of particular interest for efficient production of power and heat, and probably in the future also for the production of liquid biofuels and chemicals. In the city of Lahti in Finland, a 60 MW_{th} circulating fluidized bed (CFB) gasification plant, firing waste and biomass fuels, has been running since 1998 [1] in a combined operation with a 360 MW_{th} pulverized coal boiler. A new waste gasification plant with two 80 MW_{th} CFB gasification units has recently been commissioned and has been running for more than 1000 h [2]. Trace elements

as impurities in the product gas exiting the gasifier should be removed very efficiently [1,2].

The behavior of trace elements in fluidized bed gasification processes can be studied with thermodynamic equilibrium calculations. Details about that is given in the next Chapter.

The objective of this work has been to develop and test thermodynamic models of the reactions of trace elements with chlorine and sulfur in gasification processes of recycled fuels. In particular, the chemical reactions of trace elements with main thermochemical conversion products, main ash components, and bed and sorbent material are covered in the model. The main ash components are introduced stepwise to understand their effect in more detail. A separate database for the thermodynamic equilibrium calculations of trace elements is presented and used; this database combines data from several databases in the literature.

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The focus is on possibilities for flue gas cleanup with cyclones, filters or scrubbers used to condense and remove the compounds containing trace elements before further use of the product gas. The volatilization tendency of trace element compounds in reducing gases is established and the level of trace elements in the flue gases from different fuels is estimated. Finally, the results obtained by the model are compared with the measured data of trace elements in gasification experiments [3,4] using solid recovered fuel [5] as feedstock. The results are also compared with other corresponding studies reported in the literature. The idea of removing trace elements by careful condensation is already in practical use in some commercial waste gasification plants [1,2].

2. Combined database vs. previous work

For the work in this paper, five electronically available thermodynamic databases have been critically evaluated [3,6]. Data from all five have been used in the combined project database. The databases are:

- (1) The FACT database, Available through FactSage 5.2. CRCT, Centre for Research in Computational Thermochemistry, Ecole Polytechnique, Université de Montréal, Canada [7,8].
- (2) HSC Chemistry Version 5.11, Outokumpu Research Oy, Finland [9].
- (3) The GFE database, Denmark Technical University, Denmark [10].
- (4) The SGTE pure substance database. Scientific Group Thermo-data Europe, available through ChemSage, GTT-Technologies and FactSage [7,8].
- (5) Ivtanthermo for Windows, Glushko Thermocenter, Russia [8,11].

The five databases have very different histories which is reflected in the set of data and the amount of compounds found in them. All have essentially the same set of data for gaseous C–H–O compounds, but data on gaseous trace element compounds and condensed species differ considerably. HSC is the largest with almost 4000 compounds, FACT and SGTE both comprise some 2500 compounds, whereas GFE and Ivtanthermo both contain around 1500 compounds. All databases are different. Related to this work, Talonen [12] reported a review of the thermodynamic data for trace elements (As, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sb, Ti, V, Mo, Zn) in various data compilations in the above mentioned databases 1.–5. It was also shown that the number of included compounds varies greatly in the databases and that the thermodynamic data also vary considerably for some species. This in turn will greatly affect predictions for the speciation of the elements. In their recent review, Lindberg et al. [13] conclude that one major limitation in using thermodynamic equilibrium modeling is the lack of comprehensive databases that contain the thermodynamic data of ash compounds and phases formed during the thermochemical conversion of fuels. In order to develop a fully consistent thermodynamic database including all phases and compounds for the trace elements, a thermodynamic evaluation of all available experimental thermodynamic data is needed. However, this would require a very extensive study, as all possible compounds of the trace elements with the elements C, H, O, N, Cl, S, Ca, Mg, Na, K, Si, Al, and some others, also should be included.

The idea of selecting and combining data from different databases, including also some data generated in this study, is different in comparison with the work reported previously in the literature. When reactions between main ash components and trace elements are considered, chemical compounds containing more than one metal play an essential role. Such compounds are,

for example CaBaC_{14} , PbOSiO_2 , and $\text{Ca}_3(\text{AsO}_4)$. The databases have been inspected for such combined compounds [3].

Usually in the literature of thermodynamic equilibrium modeling of gasification processes, one software (and database) is used, such as different versions of FactSage [14–19] or HSC [20]. The next paragraph contains a critical review of some selected references with respect to their relevance to the present work.

Kuramochi et al. [14] studied the behavior and emissions of gaseous H_2S and HCl in connection with the gasification of biomass fuels at temperatures of 673–1473 K. Based on a thermodynamic analysis, they proposed to add potassium-rich biomass to reduce HCl emissions to low levels acceptable for fuel cells. However, this observation was not validated against experimental measurements from a biomass gasifier. Increasing potassium in fuel feed can lead to other problems, such as agglomeration, in case of a fluidized bed. Wei et al. [15] studied the behavior and emissions of gaseous chlorine and alkali metals in connection with biomass gasification and combustion, under various process temperature and pressure conditions and air-to-fuel ratio. They were able to identify the main gaseous and condensed species between the elements Cl, K and Na, including chlorides and hydroxides. The fact that they did not find sulfides or sulfates among these species might be an indication that their calculation system was insufficient. The modeling results were not validated against experimental results. Bunt and Waanders [16] investigated the behavior of some trace elements (Hg, As, Se, Cd and Pb) in connection with a Lurgi-type fixed-bed gasifier, by using analysis results from residual ash samples, in comparison with FactSage modeled data. In their modeling approach, they divided the fixed-bed gasifier into drying, devolatilization and reduction zones, which were modeled separately using FactSage and the results were combined in post processing. The solid ash-containing samples from inside the gasifier were collected after shutting down the gasifier operation and cooling the gasifier. They achieved good agreement between model predictions and measurements, with the exception of As. Hg, Cd, Pb, As and Se were all found to be highly volatile, partitioning into the gas phase. Hg was found to be the most volatile element in the gas phase in the form of elemental Hg. They divided the behavior of different trace elements into three classes. Class I elements, such as Hg, As, Se, Cd and Pb are volatile (low boiling point). Class II elements, such as Cu, Mo, Ni and Zn are less volatile and partition between the ash and the gaseous phase, with condensation of vaporized species on the surface of ash particles as the gas cools. Class III elements, such as Ba, Co, Cr, Mn and V are the least volatile and remain in the ash.

Bunt and Waanders [17] also investigated the behavior of some other trace elements (Ba, Co, Cr, Mn and V) in a fixed-bed gasifier, by using data from residual ash samples and FactSage modeling. They found good agreement between most of the metals besides Cr, and good prediction of ash partitioning behavior in connection with gasification. The comparison of experimental data [17] of elements remaining condensed in ash against modeling is similar to the method reported in this paper for fluidized bed gasification. Porbatzki et al [18] studied the release of alkali metals, chlorine, sulfur and heavy metals in connection with gasification of wood, straw and miscanthus. They made modeling predictions using FactSage (version 5.5) and compared the results against measurements of gas-phase species from a laboratory-scale batch fluidized bed reactor. Besides some gaseous zinc above 900 °C, they did not observe any gaseous heavy metal species in the product gas, and the modeling results were in agreement with measurements. Such a “clean” gas could be possible because of using “pure” biomass fuels, in contrast with waste-type fuels such as those used in this paper. Yoshiie et al. [19] used a laboratory-scale drop tube to gasify coal particles and took samples of the ash particles from product gas after cooling and analyzed them for their Pb and Se contents.

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