Review of

THE MATHEMATICAL MODEL OF OXY-FUEL COMBUSTION OF MUNICIPAL SOLID WASTE ON THE GRATE FURNACE INTEGRATED WITH CO2 CAPTURE

Doctoral Thesis in Environmental Engineering, Mining and Energy at the Silesian University of Technology Gliwice

Submitted by Paulina Copik

Under the supervision of Professor Andrzej Szlęk, Silesian University of Technology, and Dr. Mario Ditaranto, SINTEF Energy Research

Review by Associate Professor Corinna Schulze-Netzer, Norwegian University of Science and Technology, Norway

1. Evaluation of the published journal articles and written manuscript

The evaluation of the publications demonstrates both a high quantity and quality of scholarly output. With four papers as first and main author (60 – 80% contribution), three of which are published in a high-ranked journal (Energy, CiteScore 15.3, Impact Factor 9.0), the academic contributions are clearly impactful within the field. The perspective article "Waste-to-energy technology integrated with carbon capture – Challenges and opportunities", with over 100 citations, further highlights the research's influence and relevance to the scientific community. Additionally, the presence of nine presentations and posters at national and international conferences emphasizes strong engagement with both academic and professional audiences. Other scientific contributions leading to further publications showcase sustained research activity and collaboration at a high level. This is a robust record of scholarly achievement. The contributions are more detailed and assessed in the following.

The paper "Waste-to-energy technology integrated with carbon capture - Challenges and opportunities" offers a comprehensive examination of oxy-fuel combustion as a technology for capturing carbon dioxide emissions in waste-to-energy plants. It effectively addresses key questions regarding the challenges and opportunities of using oxy-fuel combustion for CO_2 reduction, the impact of municipal solid waste (MSW) composition on the efficiency and emissions of the process, and the primary research areas needed to optimize this technology for carbon capture and energy recovery.

In discussing the challenges and opportunities of using oxy-fuel combustion technology to reduce carbon dioxide emissions, the paper identifies the high energy consumption of air separation units, efficiency drops, and the need for sorbents to control heavy metal emissions as significant challenges. Despite these obstacles, the paper highlights the potential benefits, such as easier CO_2 capture and reduced flue gas volume, which leads to a better heat transfer to the boiler and a smaller flue gas cleaning system.

The composition of municipal solid waste is shown to play a critical role in determining the efficiency and emissions of oxy-fuel combustion. The authors explain how variations in MSW, such as moisture content and organic matter, can affect both combustion performance and the release of pollutants, emphasizing the importance of understanding waste composition in optimizing the process.

Finally, the paper identifies key research areas for further optimizing oxy-fuel combustion. These include the development of new air separation methods like chemical looping, the need for O_2/CO_2 ratio optimization, and adapting the process to use waste heat for auxiliary devices.

The paper could have been strengthened by more critically assessing the drawbacks, potential, and underlying assumptions. For example, according to the World Energy Outlook 2019 (International Energy Agency, 2019), carbon capture, utilization, and storage (CCUS) only accounts for 9% of potential solutions, while efficiency improvements contribute to 15% of energy savings. This suggests that any efficiency loss should be carefully avoided and more thoroughly evaluated. Additionally, certain statements are only plausible if waste-to-energy (WtE) plants are newly constructed. The paper does not address the long operational lifespans of these capital-intensive industrial systems. A more balanced comparison between post-capture technologies for existing plants and oxy-fuel combustion for new builds could have provided greater insight.

Overall, the paper provides a solid evaluation of oxy-fuel combustion technology, thoroughly addressing the proposed questions and contributing valuable insights to the field of waste-to-energy and carbon capture and is well received by the scientific community.

The papers on "Thermogravimetric and kinetic study of thermal degradation of various types of municipal solid waste (MSW) under N2, CO2 and oxyfuel conditions" and "A comparative study on thermochemical decomposition of lignocellulosic materials for energy recovery from waste: Monitoring of evolved gases, thermogravimetric, kinetic and surface analyses of produced chars" are here are aimed at one research aim, which is the characterization of waste fractions under oxyfuel conditions. An important baseline for oxyfuel combustion and modeling.

The thermogravimetric and kinetic study thoroughly investigates the thermal degradation behavior of various waste materials under different atmospheric conditions, providing valuable insights into pyrolysis and char burnout processes. The research effectively answers key questions, including the influence of shifting from an inert to a gasifying atmosphere on thermal decomposition below 600°C and the impact of an O_2/CO_2 atmosphere on degradation patterns.

The study also provides detailed analyses of the thermal behavior of PVC under varying atmospheres and calculates crucial kinetic parameters for the pyrolysis of textiles, spent coffee grounds, and PVC. Furthermore, the research addresses the kinetic parameters for the char burnout of textiles and spent coffee grounds, as well as the activation energy of PVC char burnout in an oxidizing environment.

By comparing methods such as Friedman, Vyazovkin, OFW, and KAS, the paper evaluates the efficiency of different kinetic parameter calculation techniques, offering a comprehensive conclusion on which approaches are most effective. Overall, the research successfully addresses its objectives and contributes meaningful findings to the field of waste material thermal degradation.

The paper is methodologically well-designed, though the selection of waste representatives is not as straightforward. "Currently, spent coffee grounds have no significant market and are problematic for disposal, but their relatively high chemical energy content makes them a potentially valuable feedstock." However, food waste, according to EU strategies, is intended for biogas production in the future and is aimed to be excluded from incineration soon. Its high moisture content raises concerns about its suitability as feedstock for waste-to-energy (WTE) without intensive drying. "Even though textile is a natural polymer and PVC is synthetic," most modern textiles are actually made of or have partial shares of synthetic fibers. The paper could have been more specific about the types of feedstock and their abbreviations to avoid potential misinterpretations. The selection of PVC as a plastic for studying chlorine species is understandable, but PVC exhibits a staged release of volatiles, a feature not shared by other relevant plastics (PE, PP, PET, PS, etc.), which have larger shares in municipal solid waste (MSW). This makes the results less generalizable and harder to transfer to other plastic types.

The paper "A comparative study on thermochemical decomposition of lignocellulosic materials for energy recovery from waste: Monitoring of evolved gases, thermogravimetric, kinetic and surface analyses of produced chars" completes the picture by providing more details on the feedstocks.

The study effectively addresses all of its key research questions, providing a comprehensive analysis of the thermal decomposition of textiles (TEX) and spent coffee grounds (SCG) under varying atmospheric conditions. The research successfully explores how different atmospheres—namely inert (N_2), gasifying (CO_2), and oxy-fuel (O_2/CO_2)—affect the distribution of solid, liquid, and gaseous products during the thermal degradation of TEX and SCG.

Moreover, the study thoroughly investigates how these atmospheres influence the composition of the gaseous products released, delivering valuable insights into emissions behavior. The research also provides a detailed evaluation of the characteristics of the solid residue (char) produced under different pyrolysis conditions, along with the impact of factors such as heating rate and reactor type on the combustion performance of the resulting char.

Finally, the study addresses the variation of kinetic parameters, including activation energy and the pre-exponential factor, for char combustion across different pyrolysis conditions. Overall, this work successfully addresses its research objectives and contributes important findings to the field of MSW thermal treatment and resource optimization.

Together with the previous study, the feedstocks "TEX" and "SCG" are well characterized, providing valuable data that can be directly used by the research community for setting model boundary conditions or for validation purposes. The detailed analysis of gas evolution under different atmospheres provides valuable insights and represents a significant contribution to the research community. It is unfortunate that PVC could not be further analyzed, possibly due to the release of corrosive gases during decomposition. In this case, it would have been beneficial to use an alternative polymer feedstock to extend the analysis.

The paper "Simplified Mathematical Model of Oxy-Fuel Combustion of Municipal Solid Waste in a Grate Furnace: Effect of Different Flue Gas Recirculation Rates and Comparison with Conventional Mode" aims to develop a mathematical model to simulate oxy-fuel combustion of municipal solid waste (MSW). While the research addresses important topics, its overall contribution appears limited.

The study's exploration of how the composition and distribution of oxidants affect combustion performance is a valid approach. However, the numerical procedure would have benefited from a more thorough literature review of existing models. Since the primary distinction of this model is the use of newly obtained oxy-fuel reaction rates, incorporating insights from other grate-furnace models could have enhanced the analysis. While the thesis builds upon using three representative feedstocks in this analysis, only one is employed (SCG).

Although the model is developed in MATLAB, it remains overly simplified, especially considering the complex nature of MSW combustion. The validation is inconclusive; the reported temperature profiles over the grate are neither representative nor realistic, and comparisons with existing literature data could have strengthened the findings. Additionally, the assumption of a constant primary airflow over the grate wind boxes may explain the observed temperature drop after Zone 2, as drying and burnout zones typically operate with much lower airflow rates. While the comparison against real operational data aims to validate the approach, it remains unclear how the simulations are processed to achieve the reported simulation values.

Furthermore, since the model is not sufficiently validated for conventional combustion, its predictive capabilities when transitioning to oxy-fuel combustion are questionable. The conclusions drawn do not rely on robust data and fail to provide meaningful insights that could directly impact the design and optimization of waste-to-energy plants.

Overall, while the paper offers a technical contribution, it falls short in addressing the broader complexities of MSW combustion and its potential to advance oxy-fuel technology in practical applications.

The following evaluation addresses the written manuscript. The introduction discusses climate change and the different impacts of various technologies. However, it appears that it was not updated compared to the paper from 2020. As this field is growing fast, new insight could have improved the introduction. The overall efficiency of waste-to-energy (WtE) with and without carbon capture and storage (CCS) or carbon capture and utilization (CCU) is examined. Nowadays, life cycle assessment (LCA) has become a standard tool for technology evaluation. It is expected that LCA-based arguments will be used when comparing the sustainability of technologies; see, for example, CEWEP Climate Roadmap 2022.

Overall, the structure of the introduction jumps between different topics in a non-logical order; for example, the author moves back and forth between the current status and the history of WtE. The aspects of WtE's operational time and capacity should be mentioned in the introduction. Post-combustion CCS is the most commonly applied technology, as existing plants can be retrofitted. The thesis seems to assume that a grate-fired plant can be converted into an oxy-fuel system. How plausible is that? Could large grate-fired furnaces be transformed into airtight oxy-fuel chambers? What about chemical looping and closed reactor concepts? Overall, a critical evaluation of whether oxy-fuel can be retrofitted or if it is a concept intended for newly built plants should be discussed in more depth.

Other important aspects also warrant discussion: What would this mean for countries with low installed WtE capacities versus those with maximum capacities? How expensive (economically, energetically, and environmentally) is the pure oxygen needed? Where would it come from?

The introduction should be revised more carefully to include more EU legislation, recent literature, and comparisons of technologies using normalized data rather than absolute numbers or randomly selected countries and their waste compositions (e.g., WtE plants per capita). Why is Poland compared to the USA? Other statements and examples lack proper references.

The chapter "Waste Incineration Integrated with Carbon Capture – Paper I" and corresponding papers discuss the benefits of CCUS only. For a well-rounded perspective, the drawbacks and challenges should also be discussed more critically. According to experts, the International Energy Agency plays only a small role (9%) in achieving net CO_2 reductions. Other measures, such as increasing efficiency, contribute more significantly and may be jeopardized by adopting CCUS. The importance and potential of alternative technologies to grate-fired furnaces, key arguments, and important aspects are highlighted in the published corresponding paper, while they are missing in the summary.

Regarding the experimental work, the methodology is not sufficiently explained or discussed. The methods aim to describe devolatilization using experimentally derived parameters. The employed heating rates are a maximum of 15 K/min. How transferable are these findings to solid fuel incineration, where actual heating rates are significantly higher? What is the potential of this method to predict the importance of considering changes in feedstock? What can we learn regarding emission formation? A comparison to state-of-the-art chemical models should have been performed, and these aspects should be discussed critically.

Overall, the summary in the manuscript does not capture all the necessary details and key findings of the study, and it does not reach the quality expected in published papers.

2. Detailed comments on the main manuscript.

a. List of publications

1) some doi addresses are missing the hyperlink

b. Nomenclature

1) E Energy activation, J/mol – should be "Ea Activation energy, J/mol "
2) k reaction rate, 1/s – the unit of the reaction rate depends on the order of the reaction
3) Greek letters – all properties here must have units as well

c. Introduction

1) Figure 1: the x-axis is not readable, and the figure should be improved following scientific standards
2) "Another serious global issue, municipal solid waste (MSW)" – why is only municipal waste a problem and not industrial and commercial waste?
3) Figures 2 and 3: As two countries are compared here, it should be considered to present them also in one Figure to make the conclusions for the reader more apparent.
4) Page 4: "As can be observed, at the end of the last century, landfilling was the most common method of waste disposal, while the amount of waste that was treated by other methods of disposal was negligible." This is a consequence of waste legislation by the EU and on a national level. The reason for trends should be highlighted rather than only described. There have been active laws since 1992, and the waste directive has been in place since 2008.

5) Page 5: For the statement that the EU sees WtE skeptical, needs references as this is a very strong statement
6) Page 6: "Currently, incineration involves controlled combustion of waste in the air atmosphere to produce ash, flue gas, and heat used to generate electricity. " The definition must be more precise, as we have plants that produce electricity or heat or both (CHP).
7) Figure 7 is misleading, and the conclusions drawn "As can be observed, most waste incineration plants are located in Western Europe, especially in Germany, France, and Italy. A large number of the WtE plants are also located in Scandinavia and Great Britain." are not correct. Italy is not a leading country in WtE and hence has huge development needs. The same is true for the UK, which has just started to increase its WtE capacity. Not the absolute number of plants but capacity per inhabitant or waste production are relevant numbers. See here https://link.springer.com/article/10.1007/s12649-018-0297-7
8) Page 10: "There are at least 20 BECCS projects worldwide, comprising various technologies, for example, ethanol plants in France, Brazil and Sweden, biomass combustion and co-firing in Japan, two pulp and paper plants in Sweden, biomass gasification in the US, biogas plant in Sweden as well as incineration plants in the Netherlands, Norway, and Japan." These projects deserve credit and references.
9) Page 11: "Therefore, introducing renewable energy certificates or a negative emission refunding system by the EU would significantly influence the economic viability of these technologies." It is unclear whether this is the author's statement or taken from somewhere else.
10) Page 12: "Many studies indicate that oxy-fuel combustion should be the most energyefficient, cost-effective as well as ecological of the available carbon capture technologies." – Which studies are these? References are needed.

d. Objectives of this study

1) The objectives need to be more clearly defined. First, there is one main objective; later, the text says "the objectives". In between research questions, no objectives are discussed.

e. Waste incineration integrated with carbon capture – paper I

1) Page 17: Check the numbers – do 400 000 t of waste lead to 400 000 t of CO2 (oxygen makes up 32/44 kg/mol)? Further, a consistent number format should be used, 400,000 versus 400 000.
2) Page 18: "that the 30% oxygen content in the CO2/O2 atmosphere" on which basis? (mass, mol or volume).
3) Page 18: "The results indicated that the direct replacement of nitrogen by carbon dioxide affects weight loss rates and the reactions occurring above 600 °C, while at lower temperatures CO2 acts as an inert atmosphere." This statement needs clarification. When CO2 acts as inert below 600°C - what does this mean and opposed to what? N2 would also act inert in this temperature range, does the author mean "as a diluent"? Further the processes above 600 °C should be stated and explained.
4) Page 18: "The authors also noticed that blends of materials can hinder the burnout and char-CO2 reaction." What are these reactions?
5) Page 18: "from oxygen-enriched combustion technology." Why not use terms like lean and rich? Define oxygen-enriched combustion technology compared to the standard classification.

f. **Thermogravimetric and kinetic analysis of the waste material under different atmospheres – II paper**

1) Page 21: What about other state-of-the-art techniques, such as the more flexible surrogate approach established by the CRECK group? A full state-of-the-art description is expected before deciding on a particular approach, which seems not flexible and depends on the individual measured reaction rates. Again, the drawbacks of the chosen approach should be discussed in more depth.

g. **Experimental investigation of waste decomposition in various conditions – III paper**

1) Page 29: "oxidising with 90/10 vol.% CO2/O2" does this correspond to the 30% O2 needed as discussed earlier? If not an explanation and discussion on why this was chosen is necessary.
2) Page 29: "Each test was repeated to assess the reproducibility of the experimental results." This statement says nothing: How many times and how were the statistics assessed?
3) Page 32: "In both atmospheres, the maximum peak of CO is around 0.15 mmol/min and appears at 380 °C for Tex and 0.50 mmol/min at 350 °C for SCG, while the CH4 peak height is of 0.03 mmol/min and occurs at 550 °C for Tex, and 0.19 mmol/min at 560 °C for SCG. However, in the gasifying atmosphere, the second peak of CO can be distinguished above 700 °C with a height of 2.5 mmol/min (Tex) and 5.8 mmol/min (SCG), which results from the secondary reactions, such as Boudouard reaction of char with CO2." This description must be linked better to the figures by referencing the figure number and tables (a or b).
4) Figures 14 to 16 are not referenced in the text.
5) Page 31: "Because of the occurrence of reactive oxygen," this should be "presence" instead of "occurrence".
6) Page 33: "The results showed that with the increasing the heating rate, higher Ea can be observed. It can be explained by the fact that slow pyrolysis developed significantly greater porosity compared to the fast devolatilization process." This needs further explanation and reference to confirming literature.

h. **Mathematical modelling of the oxywaste incineration – IV paper**

1) How are the experimental results used in the model approach?
2) Page 35: "In the last of a cycle which PhD thesis is composed of" needs reformulation, not an English sentence.
3) Page 35: The term "grate" stands for the moving metal. Do you mean "fuel bed" instead of grate? These labels should be added to Figure 17.
4) Equation 5.3 and 5.6. For the mass flow, the dot is used to indicate the time derivative to stay consistent. This should also be done for the other time-dependent variables.
5) Page 38: "many researchers» according to the references by 2. Statements like this need a substantial amount of references or should be removed.
6) Page 38 and all occurrences: there is no space between a number and °C: https://www.nrel.gov/comm-standards/editorial/temperature.html#:~:text=Use%20a%20degree%20symbol%20(%C2%B0,between%20the%20number%20and%20K.
7) Page 39: "and causing the growth of the temperature." Temperature "rises".
8) Page 39: What is "the waste char" compared to "char"?

9) Figure 18: How reasonable are temperatures of 2500 K in the freeboard? The average temperature measurements in the literature are much lower.
10) Page 39: "with lambda (λ) equal to 0.52" WtE plant is always run in a staged concept, slightly rich over the fuel bed and lean in the burnout zone and overall lean. Which values in 0.52?
11) Page 40: "As can be observed, " where? a reference to a figure is missing.
12) Figure 19: What causes the jump at 2 m distance in the figures? Why should temperatures drop like this in the middle of the chamber? The figures are very hard to read, and different line styles need to be used.
13) Figures 19 c and d. How does the distribution in the air cases that are used for validation look like? WtE plants are not operated like this. There is a profile with very little in the first and last wind box. Besides that, the figures do not provide any information. A number in the text would be more useful.
14) Page 41: "As can be seen, the higher ratio of the flue gas recirculation, the higher the usage of oxygen» And then the oxidizer is adjusted to keep the same level of O2. This statement does not make sense considering the operation of a plant.

3. Final conclusions

The presented PhD thesis is firmly situated within the disciplines of environmental engineering, mining, and energy engineering, and it meets the standards expected of a doctoral dissertation. This is largely attributed to the robust contribution of the experimental work and the significant relevance of the initial focus on oxy-fuel technologies for waste-to-energy plants. The numerical work demonstrates a commitment to advancing the field and showcases the technical potential for future contributions. Given the larger focus on experimental work, it is suggested to shorten the thesis title to "~~THE MATHEMATICAL MODEL OF~~ OXY-FUEL COMBUSTION OF MUNICIPAL SOLID WASTE ~~ON THE GRATE FURNACE~~ INTEGRATED WITH CO2 CAPTURE".

With this, I recommend accepting the thesis and proceeding with the awarding process.

Podpisała Corinna Schulze-Netzer

Trondheim, 07.11.2024