PhD THESIS

Studies regarding the implementation of circular economy through integrated use of agricultural crops

(SUMMARY OF Ph.D. THESIS)

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INTRODUCTION

The worldwide exploitation of resources and energy exacerbated alongside the increasing demographics and excessive consumption coupled with negative environmental impacts, climate change, and depletion of resources. In order to become climate neutral by 2050, the European Commission developed a number of policies, under the European Green Deal, intended to enhance resource efficiency by advancing to a circular economy (European Commission, 2019). The new circular economy action plan is one of the fundamental components of the European Green Deal, it is focused on boosting sustainable products, decreasing the reliance on natural resources, and reducing waste production through the entire life cycle (European Commission, 2020).

Rural development is of fundamental importance in the transition to a circular economy, and agriculture is one of the key areas of influence, providing renewable resources convertible into value-added products. The EU promotes the production of energy from renewable sources, setting a target of 42.5% for 2030 (EU Directive, 2023).

Considering the established targets and the well-known worldwide situation regarding the finite availability of non-renewable resources and their significant environmental impact, more ecologically friendly and sustainable energy sources are needed. Therefore, transitioning to bioenergy might provide significant environmental, social and economic benefits. Bioenergy produced from biomass is one of the most significant alternative forms of energy, representing 55% of the renewable energy (IEA, 2023). It can be produced from a variety of raw materials, such as dedicated energy crops, wood biomass, municipal/urban wastes, and agricultural biomass waste.

Agriculture is an important sector with a great bioenergy potential. Among the agricultural biomass types, vine shoot waste from the annual pruning constitute a valuable renewable resource and its valorization can lead to rural development. It is highly abundant, as viticulture represents an important sector on international and national level. Vine shoot waste is mainly composed of cellulose, hemicellulose and lignin, rendering it convertible into biofuels, bioactive compounds, and various value-added products. Depending on the technology used, the conversion of vine shoot waste into biofuels has a great circular economy potential: reuse and valorization of waste, efficient use of resources, and generation of bioenergy. It is important to test and implement sustainable strategies and production processes to accelerate the transition from a linear to a circular economy, by promoting sustainable pathways.

Although, compared to fossil fuels, bioenergy is usually more favorable for the environment, the entire life cycle must be considered and analyzed. In this regard, life cycle assessment (LCA) evaluates the environmental impact of a production process in all its phases, starting with the extraction of raw materials, processing, manufacturing, distribution, until the final disposal. LCA provides a comprehensive overview of both processes and environmental concerns, it is an iterative and quantitative process, featuring numerous feedback loops connecting each life cycle stage.

RESEARCH OBJECTIVES

The thesis approaches the circular economy potential and its contribution to rural development through the integrated use of agricultural crops under sustainability constraints. The chosen topic for the case studies – bioenergy pathways from lignocellulosic biomass waste from viticulture – offers a spectrum of opportunities to tackle circular economy, sustainability and resilience from a complex adaptive system perspective, within a flexible methodological framework – the lifecycle assessment (LCA) – enabling the integration of various models to develop scenarios and predict their evolution to contribute to rural development.

The aim of the thesis was to investigate an alternative source of bioenergy for sustainable rural development by comparing two distinct pathways: bioethanol (Case study 1) and pellets/briquettes (Case study 2) production through the valorization of vine shoot waste using LCA, and to develop a replicable circular economy model for the use of vine shoot waste. To achieve the above-mentioned aim, the following specific objectives were set: (1) To analyze and characterize vine shoot waste to establish its suitability as a renewable source of bioenergy – a preliminary step for LCA; (2) To investigate the production pathways of liquid biofuel (bioethanol) and solid biofuel (pellets/briquettes) using LCA; (3) To perform a comparative analysis of the bioethanol and pellets/briquettes production pathways from vine shoot waste by identifying and quantifying their environmental impacts; (4) To investigate which of the two analyzed production processes better responds to the sustainable circular economy principles; (5) To develop a novel circular economy model called "Total ecosystem model" (TEM) that supports the conversion of agricultural biomass waste into bioenergy, suitable for the development of rural areas, based on the results of the vine shoot waste suitability as a renewable source of energy and on the LCA's outcomes.

STRUCTURE OF THE THESIS

The thesis provides background and state of the art information regarding circular economy and life cycle assessment, in general, and in the biomass-bioenergy framework, in order to justify the importance of the subject (State of the art). Further, in the second part (Original research), it presents the objectives, the experimental design and the laboratory methodology, the ISO standardized methodology for conducting LCA and the statistical methods used. Subsequently, it provides the obtained results and proposes a novel model for biomass waste use and creation of value-added products in a circular economy system that can contribute to rural development and it ends with conclusions and recommendations. The findings of the current research provide valuable information to policymakers in support of the circular economy implementation, as well as to different relevant stakeholders in viticulture in the pursuit of sustainability and rural development.

MATERIAL AND METHODS

The raw material consisted of the vine shoots of eight vine varieties collected immediately after the pruning operations. The pellets/briquettes were produced and provided by the Research Station of the "Ion Ionescu de la Brad" University of Agricultural Sciences in Iasi from vine shoot waste. Several methods have been applied to investigate the suitability of vine shoot waste as a renewable resource for bioenergy, to collect the data necessary to carry out the LCA to assess the environmental impact of the bioethanol and pellets/briquettes production process, and to build an efficient circular economy model suitable for rural development. The experiments were performed in triplicates. STATA version 15.0 (StataCorp, College Station, TX, USA) was used for statistical analyses. OriginLab version 2020b (Northampton, MA, USA) and Microsoft Excel were used for plotting graphs. A Monte Carlo simulation was performed on the results of the LCA to assess the uncertainty analysis of each impact category using SimaPro (version 9.0). The environmental impact of the bioethanol and of the pellets/briquettes production processes from vine shoot waste were studied using the conventional LCA approach (ISO 14040, 2006; ISO 14044, 2006).

RESULTS AND DISCUSSION

Results regarding the physico-chemical characterization of vine shoot waste to assess its potential integration into the circular economy

The raw material, namely the vine shoot waste, was chopped and dried prior to performing the experiments and then physico-chemically characterized following standardized procedures. The water content ranged between 7.74% and 8.28%, values similar with those obtained in literature. Regarding the ash content, the results varied from 2.71% to 6.41%, higher than in other types of agricultural biomass waste. The cellulose content ranged between 28.78% and 40.01%, that of hemicellulose between 16.87% and 27.45% and of lignin between 24.22% and 32.34%. The vine shoot waste varieties had a high carbon and oxygen content and a very low sulfur content. Concerning the vine shoot waste gross calorific value, the highest result was 17.488 MJ kg^{-1} dry basis, while the highest net calorific value was 15.941 MJ kg^{-1} dry basis. The aforementioned characteristics render vine shoot waste suitable for biofuels production, thus, for further investigation using LCA.

Results regarding the bioethanol production phases and characterization

The vine shoot waste varieties mix was converted into bioethanol under laboratory scale conditions, through autohydrolysis pretreatment, delignification, simultaneous saccharification and fermentation and distillation. In order to determine the most suitable pretreatment reaction, different conditions were tested, specifically 150°C, 165°C and 180°C for 10 minutes at a pressure of 60 bars. The most effective outcomes were recorded at 165°C: the highest cellulose and lignin content, and the most efficient hemicellulose separation. The delignification stage resulted in a high cellulose content and a very low lignin content. The SSF stage was performed by employing a 10% substrate loading at two temperatures, 37°C and of 45°C, for 24 h, 48 h and 72 h to assess and identify the best possible conditions. The bioethanol content was higher at 37°C. Following distillation, the presence of ethanol was confirmed by gas chromatography. The bioethanol produced by the conversion of vine shoot waste variety mix underwent physico-chemical characterization. All the analyzed parameters were within the limit values established by the SR EN 15376:2015 standard. The collected data serves a basis for further analysis employing the LCA approach.

Results regarding the physico-chemical characterization of pellets/briquettes to assess their potential integration into the circular economy

The pellets/briquettes obtained from vine shoot waste were assessed according to international standards. The moisture content of both pellets and briquettes was under the 12% limit, that is regarded optimal for biomass combustion. The moisture content impacts the calorific value. The results regarding the calorific value are comparable with those from literature. The ash content plays a major role in combustion and it can have a detrimental influence on the burning process. The values ranged between 3.50 and 4.79% in pellets and between 2.09 and 3.27% in briquettes, being in the limits set by the standards. The obtained metal concentrations for pellets were situated under the limits set by the standard, except for the Cu content of three vine varieties, the increased values ranging between 25.50 and 36.10 mg kg^{-1}. In briquettes, the content of Cu exceeded the limit in all the varieties, all the other elements were under the limits. Sulfur and chlorine constitute important elements, due to their ability to cause the corrosion of the heating system and can be toxic to human health. Both elements, sulfur and chlorine, were present in the pellets and briquettes samples, with levels lower than 0.20% and 0.10%, respectively, set by the standards. Nitrogen concentrations were also below the limits in all samples, with the exception of one vine variety of pellets, with a value of 1.68%, exceeding the value of ≤ 1.5% for grade A pellets, but under the ≤ 2.0% limit of grade B. Carbon and hydrogen do not have preset limit values in the standards. The results revealed the potential of utilizing pellets/briquettes for bioenergy purposes, thus rendering them suitable for further analysis using LCA for assessing their potential to contribute to the circular economy.

Results regarding the life cycle assessment of bioethanol obtained from vine shoot waste (Circular economy case study 1)

The technological production process of bioethanol from vine shoot waste was analyzed using the life cycle assessment method. The analysis aimed to determine the environmental effects of a laboratory scale production technology that could be implemented in rural areas in order to contribute to rural development. The findings are useful for the circular economy rural development model (TEM). A functional unit of 1 kg bioethanol was defined and a system boundary of cradle to gate, that comprises all the processing stages and their related emissions. Due to the consumption of chemical reagents, to the high consumption of electricity and water, the LCA results indicate a significant contribution of the process of obtaining bioethanol in impact categories such as water consumption, ionizing radiation and freshwater ecotoxicity. The SSF, delignification and autohydrolysis stages have the highest share of causing impacts to the environment. The delignification stage contributes with over 50% to human non-carcinogenic toxicity and deficit of mineral resources impact categories, and the SSF stage to fine particle formation, freshwater eutrophication and ionizing radiation. The results provide insights for the development of circular economy models that contribute to the development of rural areas.

Results regarding the life cycle assessment of pellets/briquettes obtained from vine shoot waste (Circular economy case study 2)

The production pathway of pellets/briquettes obtained from vine shoot waste was evaluated using the life cycle assessment standardized methodology. The aim was to determine the extent to which the process contributes to the environmental impact categories. The findings are useful for the circular economy rural development model (TEM). The functional unit selected was 1 kg pellets/briquettes. Amongst the 9 processing stages, the highest influence was produced by dehydration, followed by pelletizing, baling, and chopping and grinding. The most affected environmental impact category was terrestrial ecotoxicity, thereafter global warming, fossil resource scarcity, ionizing radiation, human non-carcinogenic toxicity, water consumption, human carcinogenic toxicity, land use, and terrestrial acidification. These could be explained by energy production and consumption, fossil fuel extraction for the production of diesel used by the agricultural equipment, fertilizers manufacturing and use in the field, and various emissions to air, water and soil. The outcomes for the remaining impact categories were considerably lower. The highest overall damage incurred to the resources area of protection, followed by the ecosystems, and finally the human health. In this context, a comprehensive evaluation of the environmental implications of a production process using the LCA methodology enables a knowledge-based decision regarding which stages need to be improved and what can be done to mitigate the effects. The results provide insights for the development of circular economy models that contribute to the development of rural areas.

Results regarding the comparative analysis between the bioethanol and the pellets/briquettes production processes from a circular economy and rural development perspective

The comparative analysis was carried out to select the value-added product whose production process has the lowest environmental impact and the highest potential to contribute to circular economy and rural development. The results showed that from the possible products to be generated by the conversion of vine shoot waste into liquid (bioethanol) and solid (pellets/briquettes) biofuels, only the solid biofuels are recommended. The production process of 1 kg of bioethanol and 1 kg of pellets/briquettes from vine shoot waste was compared employing the life cycle assessment methodology using SimaPro and the ReCiPe midpoint method to establish the input to different impact categories on the environment. The results of bioethanol production at laboratory scale showed that the process required the consumption of chemicals, together with large quantities of water and electricity, all these having substantial negative environmental implications. Among the pellets/briquettes processing stages, dehydration had the highest contribution followed by pelletizing, especially on freshwater eutrophication, water consumption, human carcinogenic toxicity, fine particulate matter formation, ionizing radiation and global warming.

The production and use of pellets/briquettes have environmental, social, and economic positive impacts, contributing to sustainability and circular economy. From an environmental perspective, it would reduce greenhouse gas emissions, eliminate on field burning and make use of renewable energy. From a social and rural development perspective, new jobs would be created in the area, local supply chain and energy would be used, and new activity with added value would be established. Among the economic benefits, energy costs could be reduced, energy self-sufficiency could increase and it could reduce fossil fuel dependence, all these playing an important role in the implementation of the circular economy rural development model (TEM).

Results regarding the Total Ecosystem Model – circular economy rural development model based on life cycle assessment

The TEM was developed based on the outcomes of the physico-chemical analysis and of the LCA carried out during the research. The proposed TEM is based on the collaboration between entities forming a network and the way they regulate the processes to optimize resources management and to limit their consumption, according to sustainable environmental, social, and economic patterns. The proposed circular economy rural development model (TEM) could serve as a valuable instrument for supplying the required components to enhance the sustainability and circularity status of an ecosystem. It is intended for entities and researchers involved in decision-making processes related to the application of the circular economy principles, as well as to the social ecological systems balance, bringing together chemistry, ecology, environmental protection and rural development in an interdisciplinary effort. One of the primary requirements of the model is to accurately identify connection between economic, environmental and social processes, including the collaborating entities and their administrative techniques, alongside the system's feasible configuration.

The cooperation of the players and the resource's availability determines the cycling capabilities (circular economy) of the system and it is a condition for the self-maintenance and, implicitly, for the sustainability of the network. The cluster/community-level operations also determine overall economic and environmental gains. The circular economy is subordinated to the existence of the processes able to balance the consumed resource by re-creating it and making it available to the other production processes. The development of conceptual models and business model frameworks constitutes an essential part in scientific research, as well as in the decision-making process for stakeholders in the field.

CONCLUSIONS

The thesis enhanced the understanding of the circular economy concept and its principles, along with the importance of biomass in a circular economy and approaches for its effective implementation in a bioenergy context. An alternative source of bioenergy suitable for rural development was investigated and two production pathways were compared: bioethanol (Case study 1) and pellets/briquettes (Case study 2) through the valorization of vine shoot waste. Life cycle assessment was applied in the two case studies, proving to be a useful instrument in the evaluation of the environmental impact and sustainability of these pathways. A replicable circular economy rural development model (TEM) was developed based on the outcomes of the previously mentioned investigations.

The thesis' findings regarding the implementation of the circular economy principles by the integrated use of agricultural crops, namely vine shoot waste resulted from viticultural activities, enables the drawing of the following conclusions:

- Based on the physico-chemical characterization of the raw material (vine shoot biomass waste), the lignin content (24-33%) and the considerable calorific value (15.94 MJ kg^{-1} dry basis) demonstrate its potential for bioenergy production.
- In bioethanol production, the pretreatment method is an essential element of the process, the most effective autohydrolysis temperature was found to be at 165°C, a reaction time of 10 minutes and a pressure of 60 bars.
- The physico-chemical characteristics of the bioethanol resulted from the conversion of the vine shoot waste varieties mix are in line with the values established by the SR EN 15376:2015 standard.
- The LCA used for the calculation of the environmental impact of the two processes, through the valorization of vine shoot waste, revealed that, compared to the bioethanol production process, the pellets/briquettes production exhibited a smaller contribution to the evaluated environmental impact categories, being a more environmentally friendly and sustainable process.
- The main responsible stages in the case of bioethanol production were the delignification, generating high freshwater and terrestrial ecotoxicity due to the large quantities of water, chemicals, and energy consumed, and the SSF process, which

contributes to the formation of fine particulate matter, freshwater eutrophication, water consumption, global warming, human toxicity, and terrestrial ecotoxicity.

- In the pellets/briquettes production, human toxicity, fossil resource scarcity, and freshwater ecotoxicity were the most significant impacts, with the dehydration and the pelletization stages playing the most important role.
- The production of value-added products in the form of pellets/briquettes from vine shoot waste using a densification process represents an efficient and sustainable alternative to fossil fuel.
- The efficient use of vine shoot waste reduces the consumption of wood while avoiding deforestation, instead of dedicated crops, it reduces land use, and it provides a solution faced by vineyard owners regarding the management of large amounts of biomass wastes.
- The use of agricultural biomass waste as a source of renewable energy in the form of pellets/briquettes for heating systems has the potential to support rural development by generating local employment opportunities, improving rural ecosystems, and reducing dependency on non-renewable resources.
- The proposed circular economy rural development model - Total ecosystem model (TEM) - developed based on the results obtained by using the LCA approach to assess the suitability of the two production pathways focuses on the partnership of entities building a network and the manner in which they manage the processes to improve the management of resources and to reduce consumption, with implications on environmental, economic and social dimensions. TEM can be a useful tool for providing the elements needed to improve an ecosystem's circularity and sustainability in rural areas.

ORIGINALITY OF THE RESEARCH

The research carried out throughout the thesis represents an in-depth study on the implementation of circular economy by the integrated use of agricultural crops, such as grape vine, through the valorization of vine shoot waste. The thesis integrates the concept of circular economy and its principles within the field of bioenergy, bioeconomy and sustainability through the valorization of an alternative source of bioenergy, namely lignocellulosic biomass. It investigates and determines the optimal pretreatment, delignification and simultaneous saccharification and fermentation parameters for bioethanol production from vine shoot waste. The research shows that these parameters can be perfected to achieve maximum bioethanol yield, making vine shoot waste a promising source for biofuel production. This is the first attempt at national level to assesses in parallel the entire technological flows involved in the production process of bioethanol and pellets/briquettes from vine shoot waste. The thesis provides a complex comparative analysis from a physico-chemical and an environmental point of view, by utilizing LCA as a sustainability assessment tool. It provides new data regarding the production and use of value-added products, particularly bioethanol and pellets/briquettes, obtained from the vine shoot waste generated in large quantities by viticultural activities. The development of a novel circular economy model called "Total ecosystem model" (TEM) that brings together ecological, social, and economic dimensions in order to integrate a sustainable, homogenized approach in rural areas. The thesis demonstrates the functionality of the biomass to bioenergy concept throughout the valorization of vine shoot waste into pellets/briquettes instead of bioethanol. This approach not only reduces waste, but also provides a sustainable source of energy in the form of heat, leading to sustainable rural development. The thesis revealed information regarding the potential of employing pellets/briquettes obtained from vine shoot waste for energy purposes at local level (farm and household heating systems), contributing to circular economy and sustainability of rural areas. The results provide essential information to researchers, policymakers and various viticulture stakeholders, supporting the implementation of circular economy and sustainability.

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