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Review article

CURRENT CIRCULAR ECONOMY ASPECT IN VALORIZATION OF AGRO-INDUSTRIAL WASTE AS VALUE-ADDED PRODUCTS

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Abstract: Agro-industrial waste has been considered to be a good source for the production of biofuels. Apart from that, it has also proven to be a valuable source of high-value-added products. The conversion of agro-industrial waste into high-value-added products allows the whole process to be designed in line with the biorefinery and zero-waste circular economy concept, especially because all parts of agro-industrial waste can be utilised. The application of the circular economy to agro-industrial systems is spreading globally and is a response to the current unsustainable model of production and consumption based on resource depletion and increased demand. This review provides a more detailed understanding of the potential of the circular economy as a response to the need to reduce the environmental impact of agro-industrial waste in organic production and to promote a more sustainable agri-food industry.

Key words: *circular bioeconomy, agro-industral waste, lignocellulose, high-value products*

INTRODUCTION

Given the challenges of climate change, population growth, and urbanization, the circular economy is seen as a strategic approach to creating a more sustainable and resilient economic system. The circular economy concept became popular around 2015 as a sustainable and holistic approach to address environmental problems and economic inefficiencies associated with the traditional linear economic model. The linear economy follows a linear sequence of resource extraction, product manufacturing, and subsequent disposal as waste (Blasi, Verardi, Lopresto, Siciliano & Sangiorgio, 2023). This model had a side effect related to its negative impact on the environment, the exploitation of resources, and the generation of significant amounts of waste. On the contrary, the circular economy proposes a regenerative system in which materials are continuously recycled, reused, and repurposed, minimizing the need for new resource extraction. The aim is to create a closed-loop system that reduces waste and promotes the efficient use of resources. In this model, waste is not seen as an endpoint but as a potential resource that can be reintegrated into the production process (Blasi et al., 2023). As a potential member of the EU and a UN member state, Serbia has been committed to the concept of a circular economy. This strategy identifies the food and agriculture industries as one

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of the most critical targets for accelerating the circular economy's development, which can lessen the negative environmental impact of the sector (UN Economic Commission for Europe, 2022).

Due to global population growth, energy and resource consumption is increasing, which is also associated with high waste production. The problems of waste management and the associated economic, environmental, and social costs will, therefore, increase in the future (Chen et al., 2023). Figure 1 displays the various types of agro-industrial waste. This waste can be classified into agricultural and industrial residues, with agricultural residues further divided into field and process residues. Field residues refer to the leaves, stalks, seed pods, and stems left behind in the field after crop harvesting. On the other hand, process waste pertains to residues left after the crop is processed into valuable resources (Sadh, Duhan & Duhan, 2018).

Agricultural production in Serbia is increasing yearly and amounted to 820 million dinars in 2022. The main crops in Serbia are cereals, industrial plants, fodder crops, vegetables, fruits, grapes, medicinal and aromatic plants. As can be seen in Figure 2, the main contribution comes from cereals (40%) (Statistical Yearbook, 2023). In European countries, corn is widely produced while in Serbia, corn, wheat and barley are main contributors to cereals crop production. In this region, corn production overcomes domestic demand, therefore some amount is exported (Mihajlovski, Pecarski, Rajilić-Stojanović & Dimitrijević-Branković, 2021). Agriculture and food production in Serbia have an extensive tradition and they are one of the strongest points of the Serbian economy (Mihajlovski et al., 2021). Approximately 60 per cent of Serbia's agricultural land is used for cereal crop production. Due to this fact, cereals production leads to a large amount of waste coming from the agricultural side stream in Serbia (RAS, 2020). Based on the average harvesting index, approximately 40% of the total corn crop is waste while wheat straw waste makes up 50% of the total wheat crop mass (Petravić Tominac et al., 2020). Corn stover is identified as the most important potential source for low-carbon biomass production in Serbia, with the highest potential in the Province of Vojvodina, where it is estimated to be approximately 500,000 tons of dry matter (Martinov et al., 2015). About 80% of the corn stover and wheat straw are composed of lignocellulosic materials that contain cellulose, hemicelluloses, pectin, and lignin (Peng & Sun, 2010; Mihajlovski et al., 2021). In Serbia, there are approximately 2.3 million tons of dry mass crop residues available which could be used to produce valuable products. However, the waste treatment system is poorly organized and only 5-7% of it is estimated to be utilized.

The usage of primary energy from renewable sources is also low, at around 21%, and there is a very low level of awareness regarding sustainable development and the circular economy. This is due to the absence of an educational institution dedicated to the circular economy, as well as ineffective legislation (Mitrović, Radosavljević & Veselinov, 2017; Nesterovic, Djatkov, Viskovic & Martinov, 2023). Therefore, agricul-tural waste made up of lignocellulose material is the most cost-effective and readily available source that can be utilized for various high-value products. Several studies have highlighted its potential in producing such products (Maki-Arvela, Salmi, Holmbom, Willfor & Murzin, 2011; Rosenfeld et al., 2020; Arya, Rokes, Cahill & Lenka, 2021; Blasi et al., 2023). In line with the biorefinery and circular economy concept, this report focuses on utilizing all significant lignocellulose components to create more opportunities for future developments.

This review aims to provide an overview of the potential reuse of cereal waste, identify the streams of grain processing, identify all components from agro-industrial waste that are important for extraction, and finally, utilize the extracted target component from the streams and its applications. The analysis of structural constituents of agro-industrial waste has revealed that 80-85% of this material is present as lignocellulosic biomass. Cellulose, lignin, and hemicellulose are the primary components of lignocellulosic materials derived from plants.

Most lignocellulosic materials consist of 7-21% lignin, 20-38% hemicellulose, and 30-50% cellulose, which could vary significantly depending on the source (Pattanaik, Pattnaik, Saxena & Naik, 2019; Liu, Li, Feng & Cui, 2020). On the other hand, some agro-industrial wastes such as wheat bran, produced during the milling of wheat, have hemicellulose (20-40%) and protein (14-18%) as their main components, so

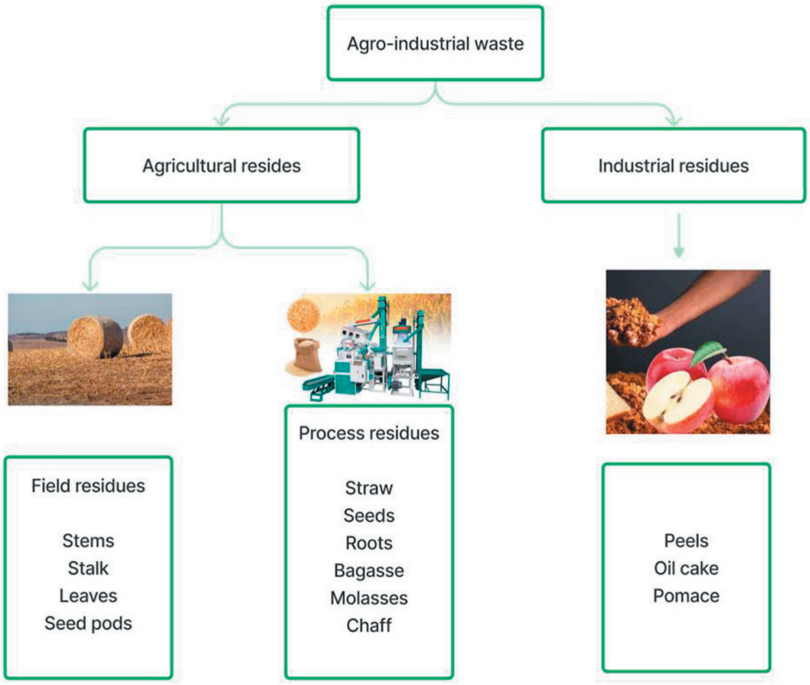

Figure 1. Classification of agro-industrial waste

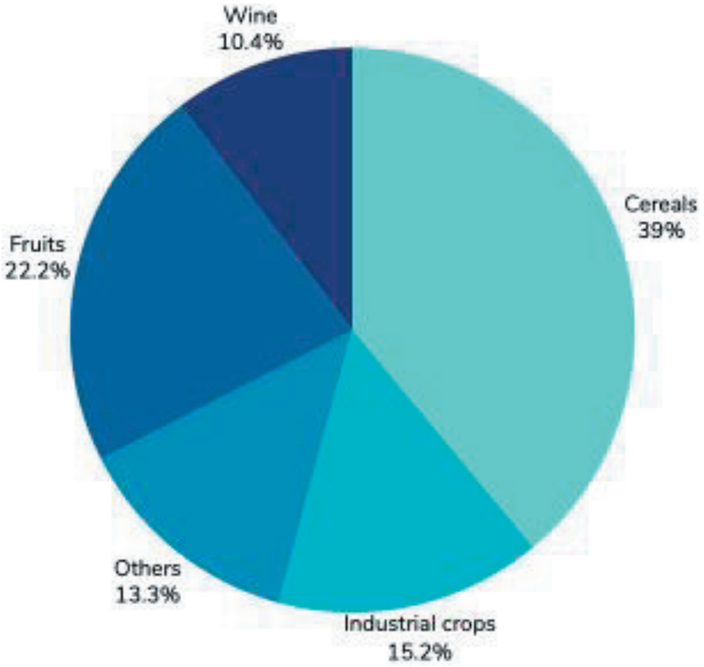

Figure 2. Agricultural goods output in Serbia (*Republički zavod za statistiku, 2023*)

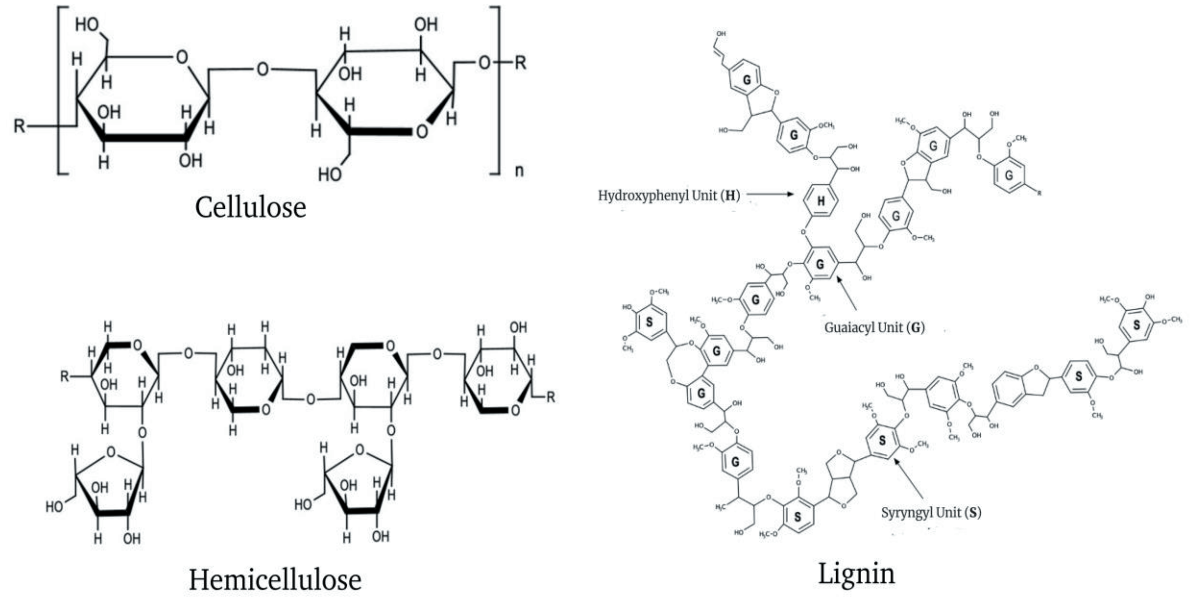

Figure 3. Lignocellulose structure

this lignocellulosic material is also a valuable source of plant proteins. Ferulic acid was often found to be the most crucial phenolic acid in lignocellulose. Hemicellulose forms covalent or non-covalent interactions with cellulose, b-glucans, and lignin, while ferulic acid forms most of the covalent cross-links of hemicelluloses via dimers and trimers.

The cell wall polysaccharides could primarily form hydrogen bonds between or non-specific surface interactions. Proteins form covalent bridges with hemicellulose via ferulic acid and tyrosine amino acid residues, forming a complex polysaccharide-protein matrix (Yilmaz-Turan et al., 2020).

Cellulose

Cellulose is the main structural component of the cell wall in higher plants, and its fibres provide strength to the wall. Its structure is depicted in Fig. 3. It is a natural homopolysaccharide composed of β-D-glucopyranose units connected by β-(1-4) glycosidic bonds (Anwar, Gulfraz & Irshad, 2014). The degree of polymerization of cellulose can vary among plant species, ranging from 925 to 5500 glucose units. Agricultural residues tend to have a lower degree of polymerization of cellulose, around 1000 (Hallac & Ragauskas, 2011). Plant biomass contains up to 50% of cellulose molecules that interconnect in their native state through intermolecular hydrogen bonds. Moreover, there is a significant tendency to form both intermolecular and intramolecular hydrogen bonds between cellulose molecules, increasing the rigidity and insolubility of cellulose in many organic solvents (Anwar et al., 2014). Cellulose molecules are organized into parallel bundles that join to form microfibrils in crystalline and amorphous regions.

Amorphous cellulose corresponds to regions where the bonds are weakened, making cellulose less ordered in its arrangement. Microfibrils build fibrils, and then cellulose fibres are formed. The fibrous structure and strong hydrogen bonds give cellulose high tensile strength, making the fibres insoluble in most solvents (Anwar et al., 2014).

In recent years, many studies have focused on the value-added potential of carbohydrates from agro-industrial waste and their application in the food, feed, pharmaceutical, and packaging industries (Fărcaș et al., 2022).

In the context of their crystal structure and cellulose nanofibers that can strengthen the plant stem, researchers have focused on isolating them from agro-industrial waste through mechanical and acid treatment to use as reinforcing fibres in biocomposites (Alemdar & Sain, 2008; Neto, Silvério, Dantas & Pasquini, 2013). As Table 1 shows, in Serbia, the most common practice is to utilize the cellulose component from corn waste through enzymatic hydrolysis to obtain bioethanol.

Hemicellulose

Hemicellulose is a non-cellulosic polysaccharide of the plant cell wall and represents a significant renewable energy source. Hemicelluloses constitute 15–40% of the dry mass of lignocellulosic materials and are the second most abundant polysaccharide in plants (Terrett & Dupree, 2019). Although scientists have highlighted their potential and applications, hemicelluloses have not been extensively applied at the industrial level. However, as the shortage of energy sources has become an everyday problem, along with environmental issues related to petroleum products, the demand for healthy food and alternative medicine has shifted scientists' attention from cellulose, which is actively used in commercial products, to other polysaccharide components of the cell wall, such as hemicelluloses (Egüés, Sanchez, Mondragon & Labidi, 2012). Therefore, using hemicellulose from agro-industrial waste after using cellulose and its conversion into other valuable products is expected to be more consistent with the zero-waste circular economy aspect. While the basic structure of cellulose is the same in all plant species, the composition of hemicellulose can vary between sources and within the plant itself, depending on the part (root, seed, leaf). Unlike cellulose, hemicellulose is often a branched, amorphous structure (Svärd, Brännvall & Edlund, 2015). Covalent and hydrogen bonds often interconnect hemicellulose polymers. They also bind to other components of the cell wall. They are hydrogen-bonded to cellulose, and through aromatic esters, they bind to lignin, thus establishing a connection between these two cell wall components. Hemicelluloses can have a diverse structure because they are composed of various monosaccharide units, pentoses (D-xylose and D-arabinose), and/or hexoses such as D-glucose, D-mannose, D-galactose, with xylose as the dominant sugar. In addition, uronic acids (D-glucuronic, D-galacturonic, and methylgalacturonic) may be present (Houfani, Anders, Spiess, Baldrian & Benallaoua, 2020). Their classification is based on the dominant sugar present in the polymer structure. They are classified as xylans, mannans, and xyloglucans. Xylan is the primary type of hemicellulose present in herbaceous plants, while the mannan type occurs to a lesser extent. The structure of xylan is represented in Fig. 3. Hemicelluloses have many physiological effects, such as inhibiting oxidative stress reactions and preventing cardiovascular disease and diabetes type 2. Also, it was found that this effect consequences with their structural characteristics (Chen et al., 2021). Previous studies demonstrated that ferulic acid attached to hemicellulose structure can affect the antioxidant ability of hemicellulose. In addition, it was noticed that cross-linking modified hemicellulose structure could be resistant to vito digestion, which leads to maintaining antioxidant and antidiabetic activities (Li et al., 2021). From an ecological and cost-effective production perspective, using hemicellulose from biomass as a starting source for prebiotic synthesis is preferred. XOS are prebiotic, functional food components that have anti-inflammatory, immune-modulatory, anti-cancer, and antioxidant effects on biology. As Table 1 shows corn cobs are the potential source of xylooligosaccharides (XOS). In addition, the European Commission has certified that XOS made from maize cobs is safe for ingestion by humans (Ristović et al., 2023).

Lignin

Lignin is a complex polymer and the most prevalent non-carbohydrate component of lingocellulosic biomass, providing strength to the cell wall and hydrophobicity that protects polysaccharides from microbiological degradation. Lignin constitutes 15-40% of the total structure and represents an amorphous, heterogeneous, branched polymer whose structure is depicted in Fig. 3. Composed of phenylpropanoid units derived from coniferyl, sinapyl, and p-coumaryl alcohols as monomers, it contains aromatic and aliphatic constituents (Yoo, Meng, Pu & Ragauskas, 2020). The basic units of lignin are connected by ether bonds, and the number of units in the polymer can vary depending on the source. Lignin is intertwined and interconnected with other macromolecules in the cell wall, linked to cellulose and hemicelluloses by covalent, ester, and hydrogen bonds, as well as structural proteins, providing strength to the cell wall. The content of hydroxyl groups in the lignin structure (aliphatic, aromatic, or total), in addition to the mentioned bonds, also influences the resistance of lingo-cellulosic biomass (Terret & Dupree, 2019; Houfani et al., 2020; Yoo et al., 2020). To solve the enormous waste problems, but also to meet the requirements of the circular economy, the use of lignin, which is a by-product of biomass, is an important target to increase the profitability of biofuel production from biomass. Table 1 shows that lignin from corn stover by-products has great potential for its antioxidant and antimicrobial application, evaluated as part of the integrated biomass-to-biofuel process. An alkaline approach was used for the extraction of bioactive lignin from biofuel residues, but extraction conditions were found to have a strong influence on these properties of lignin extracts (Dong et al., 2011). Lignin, as a residue from bioethanol production is possible to use for the generation of electric and thermal energy needed for the process (Martinov et al., 2015).

Proteins

Nowadays, researchers are very interested in turning cereal by-products into a viable protein source that can meet the world's protein needs (Fărcaș et al., 2022). Among the many protein-rich cereal by-products used in the industry, brewer's grains, rice bran, wheat bran and corn bran are suitable for the production of hydrolysates that can exhibit biological activity. The bioactive peptides that can influence human health consist of 2–50 amino acids and are divided into exogenous and endogenous categories.

Table 1.
The main lignocelluloses composition of different agro-industrial waste and their application

Source	Waste	Waste generation	Component	Procedure	Properties/Application	References
Soy	Hulls	Soybean oil extraction	Cellulose	Acid hydrolysis	Great potential as reinforcement in nanocomposite preparations.	Neto et al., 2013
Wheat	Straw	Wheat milling process	Cellulose	Chemical and mechanical treatment	Reinforcement fibers in biocomposite applications	Alemdar & Sain., 2008
Corn	Stover	Harvesting	Lignin	Alkaline extraction	Antioxidant	Dong et al., 2011
Corn	Stover	Harvesting	Lignin	Alkaline extraction	Antimicrobial	Dong et al., 2011
Wheat	Bran	Wheat milling process	Hemicellulose	Alkaline extraction+enzyme	Antioxidant, antidiabetic	Li et al., 2021
Triticale	Brain	Wheat milling process	Hemicellulose	Extraction+hydrolysis	Antioxidant, antidiabetic	Chen et al., 2021
Wheat	Straw	Wheat milling process	All components	-	Biosorbent	Božić et al., 2021
Sunflower	Husk	Peeling sunflower seeds	All components	Acid modification	Biosorbent	Radenković et al., 2022
Corn	Stover	Harvesting	Cellulose	Hydrolysis	Bioethanol	Mihajlovski et al., 2021
Corn	Stover	Harvesting	Cellulose	Hydrolysis	Bioethanol	Ilić et al., 2022
Corn	Cobs	After harvesting and grains removing	Hemicellulose	Chemical extraction and hydrolysis	XOS as prebiotic	Ristović et al., 2023
Corn	Cobs	After harvesting and grains removing	Cellulose	Chemical pretreatment and hydrolysis	Reducing sugars for bioethanol production	Mladenović, Grbić, Đukić-Vuković & Mojović, 2022
Brewers′ spent	grain	Brewing industry	Cellulose and hemicellulose	Enzymatic hydrolysis	Bioethanol	Ilić et al., 2022

Table 2.
The phenolic and protein structures of lignocellulose from various agro-industrial wastes and their utilisation

Source	Waste	Waste generation	Component	Procedure	Properties/Application	References
Triticale	brain	Wheat milling process	Phenolic	extraction	Antioxidant	Hosseinian & Mazza, 2009
Triticale	straw	Wheat milling process	Phenolic	extraction	Antioxidant	Hosseinian & Mazza, 2009
Triticale	leaves	Harvest	Phenolic	extraction	Antioxidant	Hosseinian & Mazza, 2009
Oat	bran	Oat milling process	Phenolic compounds	Ultraosound-assisted extraction	Antioxidant	Călinoiu & Vodnar, 2019
Rice	bran	Rice milling process	Free phenols Bound phenols	Ultrasound-assisted extraction	Antioxidants in cosmetic formulation	Guerrini et al., 2020
Rice	bran	Rice milling process	Protein	Microwave-assisted extraction	Antioxidant	Phongthai, Lim & Rawdkuen, 2016
Brewers′ spent	grain	Brewing industry	Protein	Alkaline extraction	Bioactive ingredients for incorporation into conventional and functional food	Connoly et al., 2013
Rice	bran	Rice milling process	Protein hydrolysates	Enzyme-assisted extraction	Antioxidant and food additive	Cheetangdee & Benjakul, 2015
Wheat	Bran	Wheat milling process	Protein	Alkaline extraction	Excellent functional properties for applying in the filed of food and supplements	Alzuwaid et al., 2020

Protein isolation methods from plant by-products are attractive to researchers because they are cholesterol-free, low in saturated fatty acids and abundant. Moreover, it has been reported that bioactive peptides from plant proteins could be used as healthy and useful ingredients for the development of functional foods (Gençdağ, Görgüç & Yılmaz, 2020). In this article, we have reviewed the protein structures with antioxidant activity obtained from agro-industrial wastes by alkaline and alternative physical and enzymatic treatments that affect the nutritional and functional properties of the obtained proteins (Cheetan-gdee & Benjakul, 2015; Alzuwaid, Sissons, Laddomada & Fellows, 2020).

Phenolic compounds

Natural antioxidants are biological compounds found primarily in a variety of plant sources. Antioxidant compounds have also been extracted from agro-industrial waste and are often used as additives in the food industry, cosmetics, phytopharmaceuticals or health products due to their bioactive properties. Various extraction methods can be used to obtain antioxidants from agro-industrial waste. However, as shown in Table 2, most of these antioxidants from by-products are related to the phenolic structure (Hosseinian & Mazza, 2009). The predominant phenolic acid in cereals is ferulic acid, which accounts for up to 90% of the total phenols, while other phenolic acids are vanillic, syringic and p-coumaric acids (Hosseinian & Mazza, 2009). The phenolic content can vary in cereals, but its distribution also depends on the part of the cereal in which the phenols were found.

For example, a higher phenolic content was generally found in the bran than in the endosperm (Fărcaș et al., 2021). In addition, the antioxidants extracted from by-products represent an environmentally friendly and economical source of bioactive compounds with remarkable health benefits that can improve the nutritional profile of products in the food industry (Connoly et al., 2013).

CONCLUSIONS

Based on the literature investigated, the use of lignocellulosic biomass for value-added applications in Serbia remains mainly limited to biofuels. Although Serbia possesses the fundamental prerequisites for bioethanol production, its share on an industrial scale is very limited. Also, the production of bio-products from biomass is not a new concept, but its introduction and implementation are a major challenge for developing countries. Factors such as limited public awareness of the benefits of renewable energy, a lack of financial resources and the absence of a legal framework hinder Serbia's ability to effectively address sustainable development issues. However, as a candidate for membership in the European Union, Serbia is committed to aligning itself with EU policies and initiatives to promote the production and use of renewable energy sources.

Furthermore, the abundant and unused agro-industrial waste offers natural solutions that can promote sustainable development on a local and global scale. In the future, secondary and tertiary processing of agricultural waste needs to be explored as it offers immense potential and available compounds that can be used in all sectors of industry, including pharmace-utical, food, beverage and chemical indus-tries, which not only benefits the bioeconomy but also ensures sustainability in our society.

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VALORIZACIJA AGRO-INDUSTRIJSKOG OTPADA KAO POTENCIJALNOG IZVORA PROIZVODA SA DODATNOM VREDNOŠĆU SA ASPEKTA CIRKULARNE EKONOMIJE

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Sažetak: Poljoprivredni i industrijski otpad često se smatraju potencijalno dobrim sirovinama za proizvodnju biogoriva. Pored toga, ovi materijali mogu biti potencijalni izvori hemikalija i proizvoda sa dodatnom vrednošću. Primena cirkularne ekonomije u poljoprivredno-industrijskim sistemima širi se globalno i odgovor je na trenutni neodrživi model proizvodnje i potrošnje zasnovan na iscrpljenju resursa ali i njihovoj povećanoj potražnji. Ovaj rad pruža detaljnije razumevanje potencijala cirkularne ekonomije, kao odgovora na neophodno smanjenje uticaja poljoprivrednog i industrijskog otpada na životnu sredinu i promovisanje održivije agro-prehrambene industrije.

Ključne reči: *cirkularna bioekonomija, poljoprivredni otpad, lignocelulozni materijali*

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