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Euro-CASE CAETS

EDITOR-IN-CHIEF'S WORD

Dear readers,

One of the tasks of our Academy is, among others, to familiarise the general public with the scientific research activities of our members, especially if these are related to their applications in industry.

In this issue Prof. Đurđica Ačkar, PhD, associate member of the Department of Bioprocess Engineering of the Academy, and full professor at the Faculty of Food Technology Osijek of the Josip Juraj Strossmayer University of Osijek, as the guest editor presents some of the possibilities and achievements in food industry especially related to the care of the quality management of waste as one of the necessities of sustainable modern civilization.

I believe that the contents of this issue will enable you to familiarise yourself with some aspects of the above-mentioned problem.

Editor-in-Chief
Vladimir Andročec, President of the Croatian Academy of Engineering

EDITOR'S WORD

Dear readers,

As in many other areas of contemporary engineering, environmental issues are of increasing importance in the food industry. To this end, recycling or reducing biological waste from food production has become a frequently addressed research topic during recent years.

By following this trend, it is my pleasure to present in this edition of Engineering Power a series of papers dealing with different aspects of by-product management in the food industry, representing excellent scientific research in the field conducted at the Faculty of Food Technology Osijek.

The Guest-Editor of this issue is Đurđica Ačkar, full professor at the Faculty of Food Technology Osijek, Josip Juraj Strossmayer University of Osijek, and associate member of the Academy in the Department of Bioprocess Engineering.

Editor
Zdravko Terze, Vice-President of the Croatian Academy of Engineering

FOREWORD

Environmental issues are deeply embroiled in the EU Strategy From Farm to Fork. Today, more than ever, the impact of food industry on the greenhouse gas (GHG) emission, water and soil pollution is protruding. It is estimated that more than 25% of GHG emissions results from food production. Therefore, food industry today faces the demand to shift from traditional "use-and-discard" approach to production that has at least neutral, if not positive environmental impact.

In recent years, many research objectives have been reuse, recycling or reduction of biological waste generated during food processing. Food industry by-products are no longer seen as a waste, but as valuable raw materials for further processing. The papers presented in this issue represent scientific research in this field conducted by different research teams at Faculty of Food Technology Osijek. As shown in the papers, the Faculty researchers deal with food industry by-products from different aspects: use in food, production of additives for bakery industry, extraction of bioactive components using green extraction techniques and use of by-products as substrates for microbial fermentation. More specifically, the overview of research of apple pomace, sugar beet pulp, and brewers' spent grain use in production of extruded snacks and modified flour; application of cocoa husk in chocolate production; as well as extraction of bioactive compounds from citrus peel, cocoa husk and tobacco waste using supercritical CO_2, subcritical water, high-voltage discharge-assisted extraction is presented in the first paper. It has shown how by-products from plant processing can be valorised through value-added food and pharmaceutical products. The second paper gives an example of valorisation of grape processing by-products by application of biorefinery concept, and gives an overview of "reuse-reduce-recycle" approach in the food industry. The third paper focuses on egg-shell, coffee spent grounds and brown onion skin in production of collagen, hyaluronic acid, enzyme immobilization, in addition to extraction of bioactive components. Combined, the three papers offer different approaches in the solution of food industry by-product disposal which may be applied in the industry single-handedly or combined. The last papers gives a different approach, showing that addressing waste disposal issues should be done in a broader range. It shows an example of educational activities involved in the municipal waste management, broadening Faculty's involvement in the environmental campaign.

Guest-Editor
Đurđica Ačkar, Josip Juraj Strossmayer University of Osijek, Faculty of Food Technology Osijek

CONTENT

Đurđica Ačkar, Jurislav Babić, Stela Jokić, Antun Jozinović, Borislav Miličević and Drago Šubarić

Transitioning to Zero Waste in Agro-Food Sector – Novel Solutions for By-Product Valorisation

Josip Juraj Strossmayer University of Osijek, Faculty of Food Technology Osijek, Franje Kuhača 18, 31000 Osijek, Croatia (dackar@ptfos.hr)

Abstract

Food industry is prompted to make a production eco-friendly and sustainable. Among other issues, by-products generated during processing received a great attention due to large quantities and valuable chemical composition – high contents of fiber, protein, and/or bioactive compounds. They are no longer considered as a waste, but as a valuable resource for food, feed and energy production. This article presents results gained through different projects conducted by the authors where solutions for the use of food industry by-products in food have been found. Brewer's spent grain, apple pomace, sugar beet pulp, oil press cakes, cocoa shell, citrus peel and even tobacco industry waste contain compounds that can be isolated and used in the form of extracts in food and other industries, but different ways of use of by-products "as such" are also shown on the examples of snacks, biscuits and chocolate, where not only problem of by-product disposal was resolved, but the nutritional values of these products were increased.
Although projects were conducted locally, the results and findings can be applied worldwide in order to resolve food industry by-product issues.

Keywords: *Brewer's spent grain, apple pomace, sugar beet pulp, oil press cakes, cocoa shell, citrus peel, tobacco waste, extraction, snack, confectionary*

1. Introduction

Food industry is a large burden to the environment. It was estimated that app. 100 million tons of waste in food chain had been produced in EU-28 annually. In developed countries, 42% of waste is produced at the consumer level (in households), 39% in food production, 14% in food service and 5% in retail [1]. Food waste produces app. 3.3 billion tons of CO_2 annually, and food waste management requires significant resources, such as water, energy, labor, soil and money [2]. On the other hand, reduction of waste and by-product generation in food chain by ¼ globally could provide enough food to feed app. 870 million people, or 12% of current population [3]. These numbers indicate necessity for prompt reaction and major modifications and adjustments of food processing.

The framework was established in 2015, when United Nations published Sustainable Development Goals [4], which were adopted in national strategies worldwide. These goals need to be achieved by 2030. Therefore, any research directed towards lower use of toxic chemicals, efficient energy use, rational waste management and industry by-products is heartily welcomed on an international level. Both science and industry have been encouraged to seek solutions to utilize food industry by-products in such manner where firstly they would be used as food, followed by utilization in feed and finally in energy production.

As in other countries, waste disposal in Croatia represents an important national issue [5], so for the last couple of years researches focused on solving that particular issue are welcomed, especially those tackling the problem of different industrial waste.

Another important aspect is a high interest towards possible use and application of different by-products, especially from the food industry. These by-products often contain large quantities of bioactive components, vitamins, minerals, etc. and could be a highly valuable raw material for the food processing [6]. Producing food/products with added value in terms of health preservation and protection, improved consumers' diets and promotion of good health and nutrition is prioritized by the European food and drink manufacturers association.

Consequently, by-product utilization has become one of the fastest growing area of research because it represents a cheap and nutritiously highly valuable raw material, and by using it in subsequent industrial processes more efficient waste management is achieved, fully in line with the UN's Sustainable Development Goals. Also, economic efficiency of a particular production process is increased, because along with the reduction of waste, added value is achieved.

As a result, the price of thus produced food should decline, and better usage of resources could benefit targeting zero hunger in the World.

Additionally, the industrial innovation and responsible production are important drivers of today's research focused on the area of sustainable development. The emphasis has been also put on the potential of a number of new, innovative techniques, so called green extraction techniques and their possibility for application on various industrial by-products. Advantages of these techniques compared to conventional extraction include shorter processing time, without or with minimal organic solvent utilization, under significantly lower temperatures, which consequently improves the quality of the final product, higher energy efficiency, higher yield of extracted components and environment preservation.

Following this strategy, our research group decided to take part in seeking solutions to "return" by-products into food, at least partially, and by employing the "think globally – act locally" approach. By now, three major and several minor research projects of our team have been targeting by-products from local food industry in Croatia, eg. brewery, sugar production, apple processing, edible oil (from different raw materials) and chocolate production with two major goals: reducing food waste at processing level and enrichment of food products with nutritionally valuable components.

In the following text, the major aims and results of (still ongoing) research that have been collected by now are reviewed.

2. Application of food industry by-products in development of functional and environmentally friendly extruded food products and additives

The project of the same name was funded by Croatian Science Foundation under the number HRZZ-IP-2013-11-1321. It lasted from 2014 to 2018 and it was focused on utilization of apple pomace, brewer's spent grain, sugar beet pulp and defatted oil press cakes (pumpkin, hazelnut, camelina, hemp) in production of modified flour, directly expanded products (flips) (Fig. 1) and cookies.

Corn grits were selected as the base material for the extrusion process, since this is commonly used raw material in snack production and corn flour has high glycemic index, making it less and less popular with consumers. Snacks and cookies were target products because they are widely consumed by all population groups, from children to elderly, and are energy dense, with low nutritive value. All by-products listed above are rich both in fiber and bioactive compounds and could reduce the caloric value of cookies and flips, while increasing fiber, vitamin and/or mineral contents. The review of composition and beneficial effects of some food industry by-products was given in detail by Jozinović et al. [7].

Fig. 1. Extruded corn snack without by-products, and with brewer's spent grain, sugar beet pulp and apple pomace

The first part of research included production of directly expanded snack (flips) in a laboratory single-screw extruder, enriched with apple pomace, brewer's spent grain and sugar beet pulp. Since all these by-products contain large quantities of water, they were dried in laboratory oven with forced air circulation at 60 °C and milled to pass through 2 mm sieve. Dried and milled by-products were added to corn grits in ratio of 5, 10 and 15% d.m. and moisture of mixes was set to 15%. Control sample was pure corn grits with 15% moisture. After equilibration, samples were extruded (at temperature profile: 135/170/170 °C, with 4:1 screw and 4 mm die) and dried at ambient conditions [8].

Although apple pomace changed the color of extrudates significantly compared to corn extrudate, it did not reduce expansion and increase hardness unlike other two by-products. Additionally, although hard, extrudates with sugar beet pulp were well expanded. The main fiber in brewer's spent grain, according to literature, is cellulose, and apple- and sugar beet pulp are significant sources of pectin. This led us to believe that pectin has an important role in expansion. Apple pectin has good gelling properties, unlike sugar beet pectin and this difference may have caused the difference in the hardness.

To explore this hypothesis, we prepared mixtures of corn grits with sugar beet pulp and brewer's spent grain with the addition 0.5 and 1% of commercial pectin and extruded them at the same conditions. The resulting extrudates were well expanded and had desirable hardness, with satisfactory results for sugar beet pulp at level of 0.5% pectin, whereas for brewer's spent grain 1% was needed [8]. This confirmed that pectin indeed has an important role in expansion. We assume this may be attributed to pectin ability to reduce fracture of cell walls by increasing their extensibility along with emulsifying and stabilizing effect on fats and proteins. The fact that sugar beet pulp pectin only contributed to expansion and not hardness may be attributed to the presence of large number of acetyl groups, its relatively small size and low average molecular weight, due to which it also has poor gelling properties.

The fiber analysis revealed significant increase of total fiber content in extrudates by addition of by-products, with the most pronounced effect of brewer's spent grain on insoluble and sugar beet pulp on soluble fiber. Snacks with 5% of by-products can have a health claim "source of fiber", while snacks with higher content of by-products can carry a claim "rich source of fiber". This is very important from the nutritional point of view and is significant improvement of otherwise nutritionally undesirable products [9].

Trained sensory panel evaluated samples with lower content (5%) of by-products only slightly lower than control sample, and, although samples with larger contents of by-products (10 and 15%) received statistically significantly lower grades, they were still in the acceptable range [8]. Here, we have to emphasize that snacks were not additionally aromatized nor salted, which would increase their palatability and overall sensory score.

The second part of the project was focused on press cakes from oil production, specifically hazelnut, camelina, pumpkin and hemp. Considering that in the region around Osijek (Croatia) there are many small oil producers that produce cold pressed oils from afore mentioned raw materials, firstly, we conducted cold pressing of selected oils in a laboratory scale. Since significant amount of oil remained in the press cakes after the cold pressing and this would cause problems with sliding of material inside the extruder, press cakes were additionally defatted using supercritical fluid extraction (more about this process in the chapter below). Defatted oil press cakes were added to corn grits in 3, 6, and 9% (much less than afore mention by-products due to the negative impact on aroma in higher amounts). Pectin was added (1%) and expanded snacks were prepared. The effect on properties of extruded snacks was similar to above, with high sensorial acceptability [10].

Additional research involved exploring the potential of supercritical CO_2 extrusion in a single screw extruder (supercritical extrusion is normally conducted in twin-screw extruders). All defatted press cakes were added in the same manner as previously described, only without pectin. The hypothesis was that CO_2 would enable expansion at the exit of the extruder, however, this did not happen. Extrudates were not expanded at all, but pores were more evenly distributed and finer than in classically produced pellets. This led us to try secondary expansion of pellets and excellent results were achieved by microwave process [10]. The benefit of supercritical extrusion in production of secondary expanded products is that no puffing agents are needed, unlike with traditional process.

The third part of research was the production of modified flour and cookies enriched with extruded flour. For this part of the research, we focused on apple pomace and brewer's spent grain, based on the fact that apple is normally used in bakery and confectionary products and consumers are well adjusted to its taste, and brewer's spent grain is a source of ß-glucan, which has a positive effect on cholesterol level and postprandial glucose in blood according to EFSA health claims [11,12].

Firstly, corn grits were extruded with the addition of by-products in levels 15, 30 and 45%. Extrudates were milled and used in cookie production as a substitute for 5, 10 and 15% of wheat flour. The physico-chemical analyses revealed that such produced modified flour may be used in the production of cookies enriched with fiber up to 15% level, where all samples may carry a health claim "source of fiber", and some even a claim "rich source of fiber", without significant alterations in physical and sensory characteristics of cookies [13].

Encouraged by excellent results in this project, we decided to further explore potential of food industry by-products for extraction of bioactive components.

3. Application of innovative techniques of the extraction of bioactive components from by-products of plant origin

The project of the same name was funded by Croatian Science Foundation under the number HRZZ- 2017-05-UIP-9909. It started in 2018 and will last until 2023 and is focused on production of the extracts rich in bioactive components from selected by-products using different innovative green extraction techniques. Their application increases the extraction yield, higher content of bioactive components in the extracts, higher quality of the extract, and resource savings in time, solvents, etc. [14]. Each of the above mentioned extraction techniques has its advantages, so it is necessary to examine the process parameters of each extraction procedure and to compare them with conventional extraction methods to obtain a true insight about the impact of a particular method on the content of the bioactive components in obtained extracts.

Cocoa shell, tobacco industry waste and citrus peel are only some of the potentially valuable by-products that can be successfully used in the production of extracts rich in bioactive components and are therefore selected as raw materials in the project.

3.1. Focus on cocoa shell

During the processing of cocoa beans (*Theobroma cacao* L.), the cocoa shells, comprising at least 10% weight of cocoa fruit, is discarded. The cocoa shell is mainly used in animal feed preparation, fertilizers or as a fuel. So far, the cocoa shell has been the main source of dietary fiber [15], but new studies have also shown the content of some other valuable bioactive components such as phenolic compounds (Fig. 2). Phenolic components of this potential raw material have an antioxidant effect and protect cells from oxidative stress. Cocoa shell is also a potential source of proteins [16], theobromine, theophylline, caffeine [15] and water-soluble pectins [17]. Theobromine is a potential diuretic, stimulates kidney circulation and alleviates elimination of harmful substances in the urinary system [18]. Caffeine, theobromine and theophylline have a similar effect on the body such as stimulation of the nervous system, stimulation of the heart and bone muscles and muscle relaxation, but with different intensity. During the fermentation of cocoa beans, these compounds migrate into the cocoa shell, making it inadequate for use in animal feed. The relatively low fat content and low concentration of soluble sugars are an advantage of this potential raw material and make it a useful ingredient for functional energy-reduced foods [18]. This matrix is still insufficiently explored especially in Republic of Croatia and therefore in this project special attention will be given to research on this by-product.

Fig. 2. Bioactive components in cocoa shell and their potential health effects

3.2. Focus on tobacco industry waste

During tobacco production in the industry, large quantities of waste (leaves, parts of leaves) that cannot be recycled are produced. This waste is hazardous and represents a significant environmental problem [19]. Tobacco leaves (*Nicotiana tabacum*) contain pyridine alkaloids, primarily liquid nicotine [20]. Nicotine has a wide range of applications, including pharmaceutical, chemical, industries, as well as in the tobacco industry as the primary additive in cigarette manufacturing. It has antimicrobial and insecticidal activity, which is why it is used as a natural insecticide [21]. By reducing the nicotine level in the polluted solid to the level below the legal threshold using different extraction techniques, it would be possible to change the classification of waste from “hazardous” to “special” waste, which could then be simply removed as urban waste or further used as a starting material for extraction of bioactive components [19]. In addition, tobacco leaves contain carbohydrates, proteins, high concentrations of organic acids, glucosides, phenolic compounds, flavonoids, solanesol, and since waste is actually a parts of leaf of different granulation, all of the components are also found in the tobacco waste (Fig. 3) [21].

is rutin. Its potentially positive effect is manifested in reducing capillary, swelling and bruising and because of that it is used to treat venous insufficiency and to improve microvascular blood flow so tobacco waste can be an efficiently source for rutin extraction for the pharmaceutical industry [32]. This matrix is still insufficiently explored especially in the Republic of Croatia and therefore special attention to research on tobacco waste is given in the project.

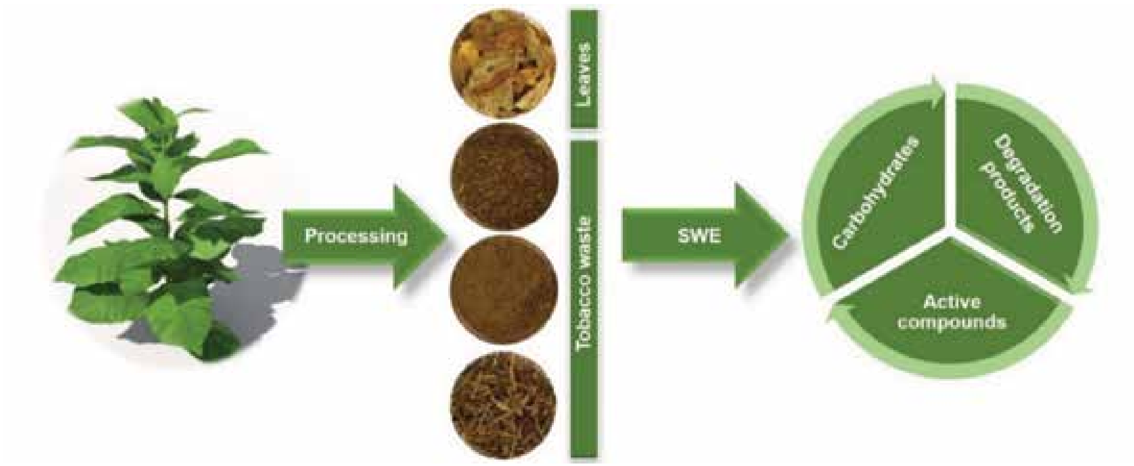

Fig. 3. Tobaco waste extraction (one example using SWE)

In tobacco, a high amount of solanesol is found. It is the terpene alcohol, consisting of nine isoprene units that play an important role in plant interactions with their environment, and it is a key intermediary for the pharmaceutical synthesis of supplements and ubiquitous drugs [22]. It has antioxidant, antibacterial, anti-inflammatory and anti-septic activity [23]. It is industrially important because it represents the starting material for coenzyme Q10 and vitamin K analogues [24]. Since coenzyme Q10 is present on the market in the form of dietary supplements to relieve pain caused by migraines [25], has protective role in Parkinson disease and other neurodegenerative diseases, a positive effect on regulation of blood pressure and glycemia in type 2 diabetic patients [26], solanesol demand is on the rise.

Clorogenic acid is used as an additive in beverages, food, cosmetics as well as in medical substances. In addition, it has antibacterial and antiviral properties and is a natural antioxidant and anticancer agent [27]. Caffeic acid is one of the natural phenolic compounds widespread in plant materials, and recently, pharmacological studies have shown that caffeic acid has antioxidative [28], anti-apoptotic [29], antidepressive [30] and anti-cancer effect [31]. Tobacco industry waste is a potential raw material for producing of caffeic acid. One of the most important flavonoids found in tobacco

3.3. Focus on citrus peels

One of the main by-products of citrus peel is essential oil. In our project, essential oils are produced by different extraction techniques from selected types of different citrus peels (special Croatian varieties) which have not yet been explored. Along with aroma compounds, citrus peel is also rich in phenolic components, such as phenolic acids and flavonoids. The most common flavonoids present in the citrus peel are hesperidin, naringin, narirutin and neohesperidin [33]. Hesperidin is found in citrus, as well as in peel in high concentrations, especially in sweet orange and lemon [34] and possesses different properties, such as positive effects on vascular or cardiovascular system [35], protective effect in case of exposure to radiation [36], anti-inflammatory [37], anticancer [38], antimicrobial [39], antioxidant [40] and a negative impact on fertility [41]. Hesperidin can be used as a dietary supplement primarily in combination with other components such as vitamin C. Also naringin, a flavonoid found in higher concentrations in the citrus peel is determined [33]. Naringin has a number of positive effects on human health and life, such as antioxidative [42], anti-inflammatory [43], anticancer [44], gastroprotective function [45], positive effect on cardiovascular disorder [46], diabetes complications, bone diseases [47] and allergy [48].

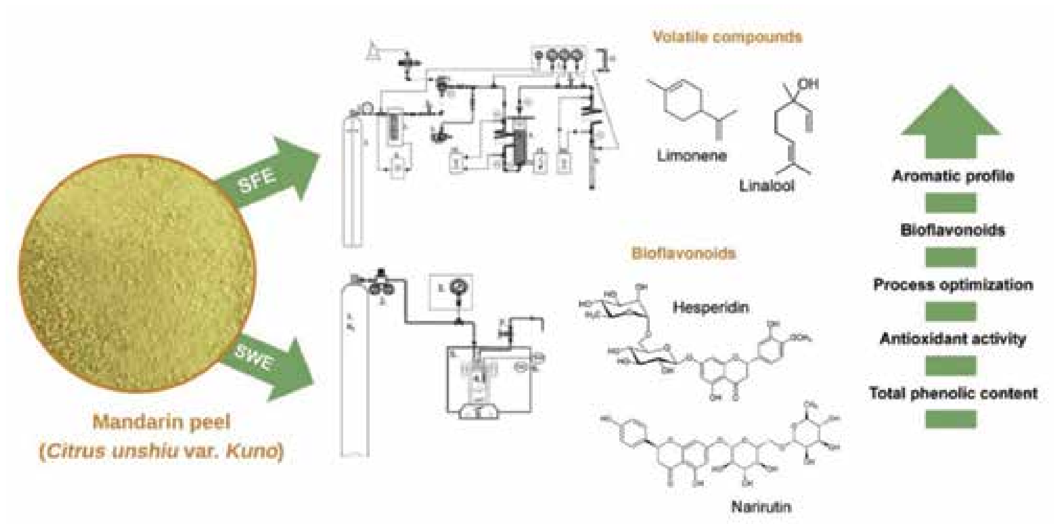

Fig. 4. Mandarin peel extraction (one example using SWE and SFE)

The innovative green extraction techniques in this project are applied in the isolation of different bioactive components from selected by-products with finding the potential applications in other industries. One example is given in the Fig. 4 for the extraction of bioactive compounds from mandarin peel variety Kuno using two different extraction techniques: SFE for the isolation of nonpolar volatile compounds and SWE for the extraction of polar bioflavonoids.

4. Cocoa shell as a raw material in food production

With good results of the previous projects and chocolate factory on hand, we tried to further valorise cocoa bean shell. Namely, the shell represents up to 20% of cocoa bean [49] and is discarded prior or after the roasting of cocoa beans. It has application in animal feed, for mulching in agriculture, biofuel production and as an adsorbent. Despite this, it still represents a major load for the environment due to limitations: low bulk density makes it very light and wind spreads it easily abroad; it contains methyxantines that are harmful for some animals and have to be extracted before inclusion into feed; polyphenols inhibit microflora and microbial biofuel production is limited [50]. Therefore, we tried to put it "back into food". Firstly, we continued with extrusion experiment and added it to corn grits in the ratio of 5, 10 and 15% d.m. and expanded it directly as described above (Fig. 5). Obtained extrudates were darker and harder than control sample, but acceptable for consummation, with increased polyphenol content [51].

Fig. 5. Extruded corn snack with added cocoa shell.

Further investigation is oriented towards chocolate and chocolate-like products within an ongoing project "Application of cocoa husk in production of chocolate and chocolate-like products (UIP-2017-05-8709)" financed by Croatian Science Foundation. Current scientific researches in the chocolate technology largely deal with polyphenol content in the products [52], influence of growing conditions and origin of cocoa beans [53], cocoa bean processing [54] and chocolate processing [55] on polyphenol and/or aroma profile and increase of polyphenol content in chocolate by addition various additives of plant origin, rich in polyphenols [56].

Cocoa shell has also been in focus of scientific research as a source for extraction of polyphenols, theobromine, dietetic fibre, pectin; in preparation of milk beverages, enzymes, as an adsorbent in water purification and biogas production substrate. Furthermore, it was used to enrich cookies and bread, as well as in production of muffins enriched with cocoa husk fibre. Cocoa husk polyphenols were antioxidants in frying oil and cooked beef [10,15]. As far as members of this research group are aware, scientific researchers have not dealt with cocoa husk application in chocolate.

Cocoa husk is not used in current chocolate production processes above 3% since it influences aroma (high content of polyphenols), high fibre content makes it coarse and difficult to reduce particle size and if used in quantities exceeding 3%, it makes problems with viscosity.

Firstly, we tried to explore if HVED (High voltage electrical discharge) treatment would be a beneficial technique in decontamination and disintegration of the husk. We came to some interesting findings, from polyphenol content [49], where we determined that virtually all caffeic acid is extracted into water during HVED treatment, unlike (+)-catechin, (-)-epicatechin, (-)-epicatechin gallate, 10 – 90% of which was retained in the husk. Also, we determined reduction of the contents of acrylamide and HMF in cocoa husk to quantities below the limit of quantification, or even below the limit of detection [57] and explained HMF and acrylamide content reduction in the treated husk by different chemical mechanisms [58].

In parallel, we have produced dark and milk chocolates with cocoa husk (Fig. 6). The content of the husk was 5, 10 and 15% for dark and 2.5 and 5% for milk chocolates. These percentages were selected based on EU legislation for quality of chocolate, where we aimed to obey the legitimate cocoa butter and non-fat cocoa solids content. The addition of both treated and untreated cocoa shells resulted in softening and darkening of samples, which could have a positive effect for consumers. However, the particle size distribution and rheology were negatively affected, which could pose the problem in the industry [58].

Conclusions

From the results listed above, it is evident that food industry by-products have a great potential to be used as a raw material in food industry and as a source of bioactive components for pharmaceutical industry. The results of the presented project research will contribute to solving the problem of large quantities of organic waste, having both environmental and financial effects. Research on industrial scale is needed in order to adjust processes where necessary and to maximize usage of plant raw materials. Special emphasis in the projects is on the possible commercial valorization of the research results and on the technology transfer of those scientific results to the industrial level.

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Fig. 6. Laboratory production of chocolate – refining in a ball mill, tempering and moulded chocolate.

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Ana Bucić-Kojić, Marina Tišma, Gordana Šelo, Josipa Grgić, Gabriela Perković, Mirela Planinić

Winery Production Residues as Feedstocks within the Biorefinery Concept

Josip Juraj Strossmayer University of Osijek, Faculty of Food Technology Osijek, Franje Kuhača 18, 31000 Osijek, Croatia (abucic@ptfos.hr)

Abstract

The concept of biorefinery was introduced as a response to the global energy crisis and climate change resulting from industrialization. Wineries produce large amounts of organic residues (grape pomace, skins, seeds, vinasse, wastewater), which are potential feedstocks for biorefineries for the sustainable production of biofuels and bio-based products (chemicals, materials, biopolymers, food, feed, pharmaceuticals, nutraceuticals), while reducing the environmental impact, which is the core of the circular bioeconomy.

Keywords: *winery production residues, biorefinery, bio-based products*

1. Introduction

The circular bioeconomy, or the sustainable production of renewable biological resources and the conversion of these resources and waste streams into value-added products (biofuels and energy; chemicals and materials; food and feed; and cosmetics and pharmaceuticals)-(Fig. 1.), is considered a key element for the successful functioning of the European Union economies in the future [1].

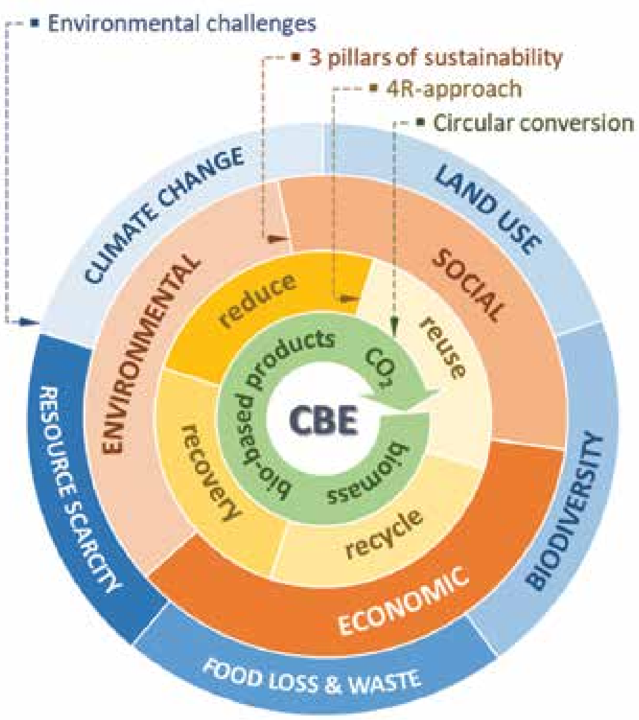

Fig. 1. Circular bioeconomy (CBE) as a solution for greatest environmental challenges (own illustration adapted from various sources: [2–4])

Indeed, current global challenges such as climate change, ecosystem degradation, and the increasing demand for food, feed, and energy require new methods of producing and disposing of production residues. In the transition from the current linear model of a bioeconomy based on an unsustainable “take-make-use-dispose” framework to a circular bioeconomy based on a 4Rs (“reduce-reuse-recycle-recover”) framework, the development of biorefineries plays an important role. A biorefinery can be defined as an infrastructure facility that incorporates various conversion technologies (mechanical, thermochemical, chemical/biochemical, and biological) that produce a bio-based product from various biomasses such as lignocellulosic materials, algae, or food waste in a sustainable way, following ESG (economic, social and environmental) criteria. Particular emphasis is placed on cascading biomass use (Fig. 2.), i.e., using the same feedstock biomass to produce several different products within the same or different biorefineries to reduce waste production [2, 5, 6].

Wine industry is an important economic sector both in the world and in Croatia, in terms of wine production, but also in the development of wine tourism. Grapevine (*Vitis vinifera*) is one of the most cultivated crops in the world with an average annual grape production of 75 million tons in the last 10 years, while in Croatia it amounted to 148 thousand tons in the same period [7]. It is estimated that 70-80 % of grapes are processed into wine, leaving behind production residues of organic origin (grape pomace, wine lees), inorganic origin (diatomaceous earth, bentonite and perlite), wastewater and greenhouse gasses [8]. More broadly, wine industry production residues include grape shoots, leaves, and stalks as well as vinasse, a byproduct of the distillation of wine lees to produce ethanol [9]. In recent years,

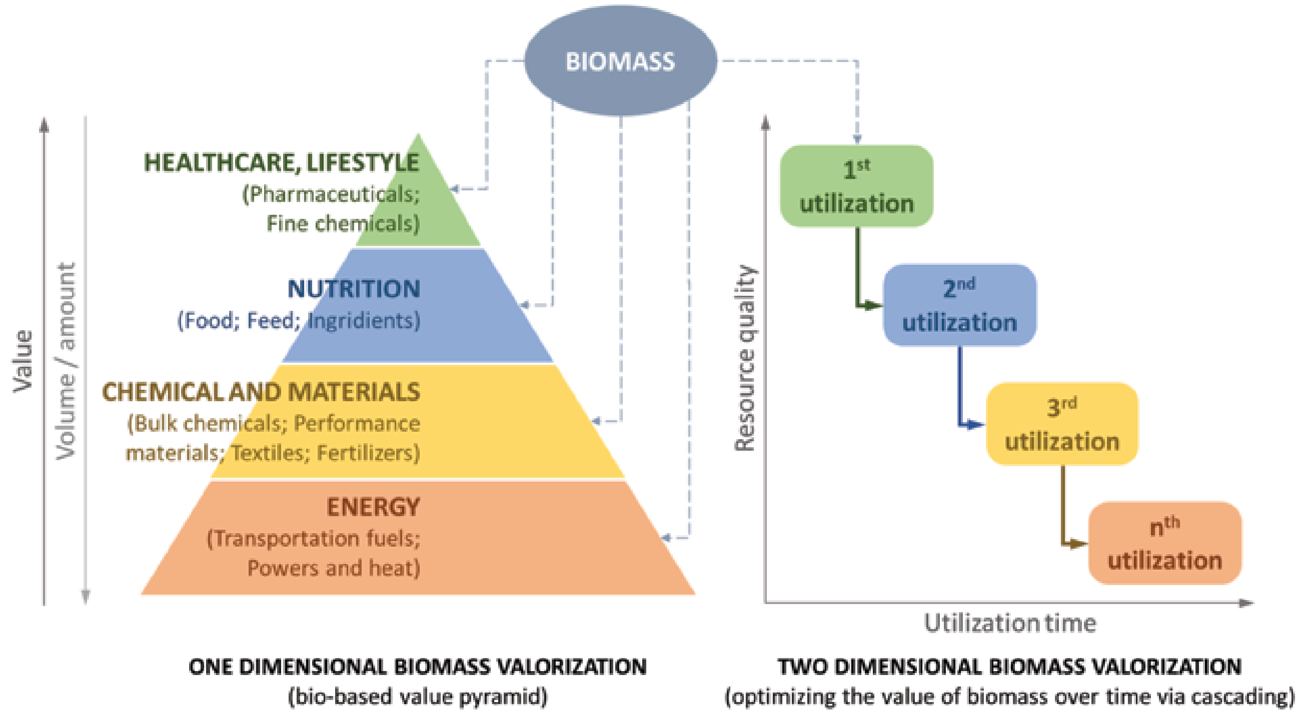

Fig. 2. Biomass valorization by circular bioeconomy (own illustration adapted from source [3])

grape pomace has been of particular interest because it is a raw material with low market value and a readily available natural source of polysaccharides, proteins, vitamins, minerals, fatty acids, fiber, oils, and biologically active compounds, including phenolic compounds [10]. Nevertheless, grape pomace is usually disposed of in landfills and vineyards or burned without a waste management strategy, which can cause numerous negative environmental impacts (hindered germination of plants in the soil, development of unpleasant odors, etc.) and economic losses.

This article presents some of the possibilities for the utilization of wine production residues within the concept of biorefinery.

2. Grape pomace

Grape pomace is the production residue of a winery, consisting mainly of skins, pulp and seeds, and may sometimes include stems. During wine production, 20 - 30 % of the mass of processed grapes remains as grape pomace [5]. The proportion of each component, as well as the chemical composition of the grape pomace, depends on the grape variety, geographical origin, agrotechnical growing conditions, and winemaking process. According to the literature, the proportion of grape seeds varies in the range of 15 - 52 $\%_{db}$ and the proportion of skin about 65 $\%_{db}$ of grape pomace [11]. According to the chemical composition, grape pomace is a lignocellulosic material containing 9.2 - 14.5 % cellulose, 4.0 - 10.3% hemi-cellulose and 11.6 - 42.2 % lignin. In addition, grape pomace contains 42.6 - 74.5% fiber, 7.0 - 23.5 % protein, 2.7 - 49.1 % sugar, and 4.8 - 6.7 % total phenols [12, 13]. Despite its rich chemical composition and multiple possibilities of its use (production of enzymes, biofuels, biopolymers, etc.), grape pomace is most often brought into context with phenolic compounds that can be used in various foods, cosmetics, and pharmaceutical products.

3. Bioactive phenolic compounds of grape pomace

Bioactive phenolic compounds of grape pomace include phenolic acids, flavanols, proanthocyanidins, flavonols, anthocyanins, and stilbenes [14]. The potential uses of phenolic compounds are diverse, but recently they have been mostly associated with beneficial effects on human health due to their potential multiple biological activities such as antiallergic, antitumor, anti-inflammatory, antimicrobial, antioxidant, anti-aging, antihyperpigmentation, antithrombotic, cardioprotective, and cardioprotective etc. [14–16]. Since phenolic compounds are unstable bioactive components, especially when exposed to light, oxygen, and elevated temperatures, they need to be protected from degradation in order to be used for further purposes. Therefore, all processes for the preparation of extracts rich in phenolic compounds from natural raw materials, including grape pomace (storage, sample preparation, extraction, stabilization of extracts), must be carried out under controlled and specified optimal conditions. Extraction is the first step in the isolation of phenolic compounds from raw materials and it depends on numerous process conditions (time, solvent concentration, temperature, etc.) that must be optimized to obtain maximum yields of biologically active phenolic compounds. Due to the structural diversity of phenolic compounds, there is no standardized method for their isolation [17], and application of eco-friendly methods are preferred. The most commonly used method for the preparation of grape pomace extracts is the conventional solid-liquid extraction along with the application of an appropriate solvent (ethanol, methanol, acetone, their mixture with water, etc.) which extracts the targeted easily soluble polyphenols. Recently, alternative solvent extraction methods have been developed, such as ultrasound assisted extraction, microwave assisted extraction, application of pulsed electric fields and pulsed ohmic heating, application of high voltage electrical discharge (cold plasma), pressurized liquid extraction and supercritical fluid extraction which have certain advantages such as better extraction yield, higher extraction rate, economic and energy efficiency, and lower negative environmental impact [17, 18].

During winemaking, only 30 % of phenolic compounds are extracted into the wine, while 70 % of bioactive phenolic compounds remain in the grape pomace. However, a significant proportion (98 %) of the phenolic compounds in grape pomace are incorporated or trapped in a complex lignin structure, making them difficult to extract using conventional extraction methods [19]. The extraction of trapped polyphenolic compounds requires additional processes of degradation, most common being acid hydrolysis or application of commercial enzymes, which increases the costs of production and/or is not environmentally friendly [20, 21]. Recently, there has been a growing interest in the development of bioprocesses for the production/extraction of bioactive substances which enable the production of high quality extracts in an environmentally friendly way. One of the mentioned bioprocesses is solid-state fermentation (SSF) which has a great potential for converting agricultural and food waste into numerous high-value products, including polyphenolic compounds, under the concept of lignocellulosic biorefinery. In order to maintain the biological activity of the extracts due to the presence of phenolic compounds, different stabilization methods could be used, one of which is encapsulation.

3.1. Solid-state fermentation

SSF is a process in which microorganisms grow on a moist, solid material under controlled conditions, without the presence of free water or with a minimal amount of free water [22]. Grape pomace is a suitable substrate for the cultivation of various microorganisms (yeasts, bacetria and fungi) under SSF conditions because it contains the nutrients necessary for their growth [23]. The filamentous fungi are the most commonly used microorganisam in SSF process especially those from fungal kingdom sub-division Basidiomycota and Ascomycota [24]. During growth on the substrate, microorganisms synthesize a number of enzymes (pectinase, cellulase, tanase, β-glucosidase, xylanase, etc.) [21, 25] that can degrade polymer structures and release phenolic compounds from the complex lignocellulose structure making them more readily available for extraction [20]. The efficiency of the SSF process is influenced by a number of factors such as type of bioreactor, substrate (chemical composition, moisture content, water activity, particle size, layer height), inoculum (concentration, culture age, morphology), carbon and nitrogen source, micro- and macro-elements, addition of inducers of synthesis of certain enzymes, mixing, temperature, pH and oxygen concentration. SSF can be implemented in different types of bioreactors (tray, packed beds, rotating disc or drums, fluidized bed, air-lift, immersion, etc.) [26, 27]. Tray bioreactors are a traditional type of bioreactor most commonly used in laboratory scale for enzyme production [28], for lignin degradation [28, 29], for the application of biologically treated material in the process of biogas production [30], and they are also used in commercial processes in various industries (production of fermented foods such as tempeh, or production of enzymes) [25, 26, 31]. The reason for this is simple design and operation, and easier scale-up compared to other bioreactors.

3.2. Encapsulation

Encapsulation is the process where the active matter or their mixture is coated with a polymer that protects it from negative external influences (physical, chemical, and biological degradation). Coatings may vary in their origin and properties, but preference is given to those that are generally recognized as safe (GRAS). The choice of coatings depends on the encapsulation method used and the encapsulation objective, e.g., masking of undesirable odors, maintenance of storage stability, targeted delivery, and controlled release of the active ingredient during digestion [32]. Polysaccharides (cellulose and its derivatives; plant exudates such as gums, starches, and starch derivatives; seaweed extracts such as alginates), proteins of plant or animal origin, and lipids are most commonly used for encapsulation of biologically active extracts. Of particular interest is the use of coatings isolated from natural sources, including food industry residues, such as soy and whey protein isolates, Arabica gum, gelatin, and starch, which is important for developing sustainable encapsulation processes.

Various encapsulation techniques are known, such as spray drying, freeze drying, extrusion, ionic gelation, emulsification, molecular inclusion, and coacervation [33]. The most commonly used encapsulation techniques are spray drying with maltodextrin as coating and ionic gelation with sodium alginate and calcium chloride.

4. Bioproducts based on winery residues

Products that can be made from grape pomace or individual parts of it (seeds, skin, stems) can be divided into primary and secondary products. Primary products include e.g. grape pomace extract rich in phenolic compounds, pellets, grape seed oil and enzymes. Secondary products can be produced by using primary products or by processing and converting primary products under the biorefinery concept (bioethanol, biogas, electricity and heat, bio-based materials) (Fig. 3.). Grape pomace extracts rich in phenolic compounds can be further used for a wide range of functional products such as food (e.g. functional cookies), pharmaceutical products (dietary supplements) or cosmetic products (creams), positively affecting the stability, organoleptic and technological properties of the functional product [15, 34]. Functional products are becoming increasingly important due to the increased awareness of the importance of consuming functional products to prevent modern disorders and diseases (diabetes, cancer, cardiovascular and neurodegenerative diseases). Functional products are products that naturally contain or are enriched with biologically active ingredients. Biologically active substances from natural sources are preferred over synthetic compounds, which can often be carcinogenic [8].

Grape seed oil is the best known commercial product from the production residues of the winery, which can be obtained by pressing or extraction. Being rich in antioxidants (vitamin E) and unsaturated fatty acids, it is very stable and is used in cooking and as a base oil in cosmetics, food and pharmaceutical products [10].

Grape pomace can be used in various feed formulations, but due to its high content of insoluble lignin and indigestible polysaccharides (cellulose, hemicellulose, pectin), this requires pretreatment by biological, chemical, or physical methods, or a combination thereof [11]. Even when grape pomace is used as a biofertilizer, pretreatment (e.g. composting) is required to obtain the appropriate properties (high content of nitrogen, potassium, and phosphorus, organic matter for soil nutrition without negative environmental impact).

In addition, it can serve as a substrate for the production of enzymes (cellulase, pectinase, xylanase,

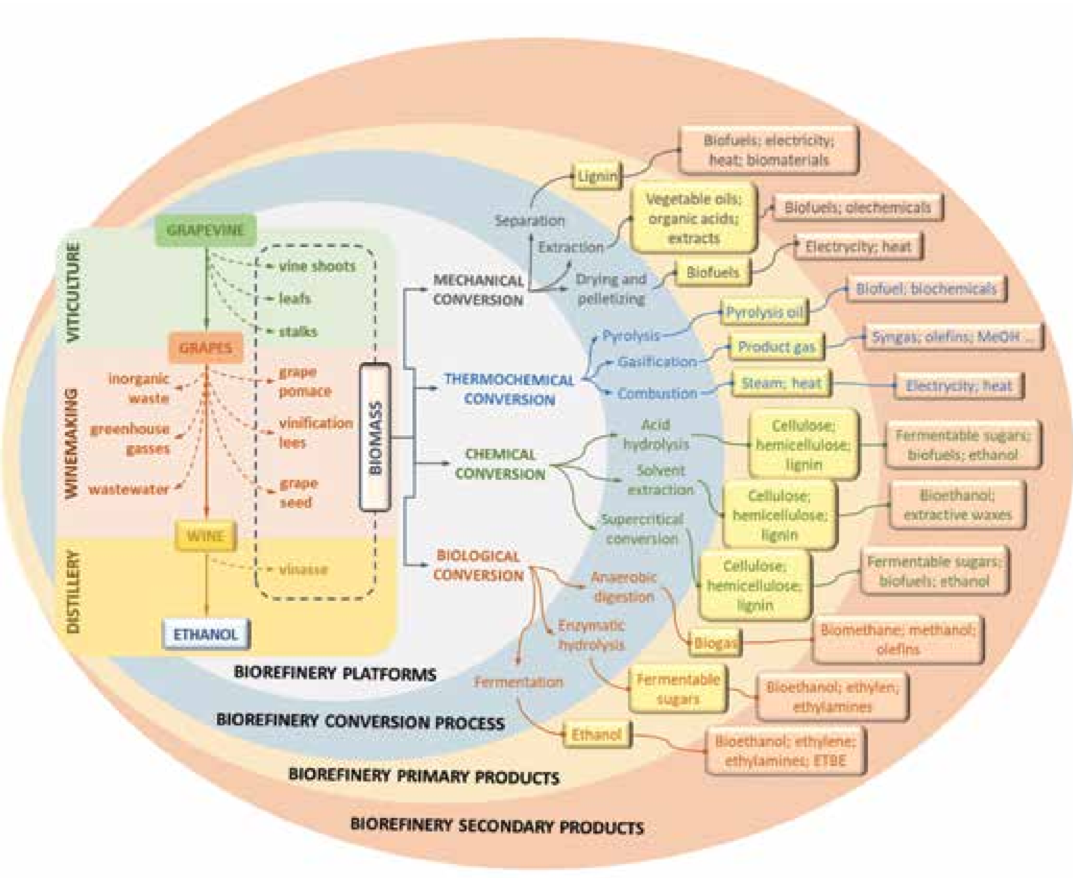

Fig. 3. Biorefinery concept applied od winery (own illustration adapted from various sources: [2, 5, 9, 35])

β-glucosidase, exo-polygalacturonase, laccase, lignin peroxidase, manganese peroxidase) from grape pomace in SSF process with various microorganisms or can be used as an inducer for enzyme synthesis in submerged fermentation [9, 21].

Due to its chemical composition and high energy value, grape pomace can be used for the production of biofuels (pellets, biochar, bioethanol, biogas) as an alternative to fossil fuels. Grape pomace pelletization, in which involves drying the pomace to a moisture content of less than 12 %, yields biofuel that can be used to generate heat and electricity in wineries or other facilities. Biochar from grape pomace is usually produced by thermochemical processes or pyrolysis. Hydrothermal carbonization can be used as an alternative to pyrolysis, especially for biomass with a higher moisture content. Apart from being a source of energy, it can be used to improve the chemical, biological, and physical properties of soil and to remove pollutants from wastewater [5, 36]. In the production of bioethanol from grape pomace, unlike sugar feedstocks, it is necessary to pretreat them with chemical, thermochemical, or biological processes to break down a complex lignocellulose structure into simple sugars, from which bioethanol is formed by fermentation. The production of biogas from pomace is possible through the process of anaerobic co-digestion with manure with prior degradation of lignin by some of the mentioned conversion processes [5]. Grape pomace can also be used as filler for the production of polymeric multicomponent composites [37].

Vinasse is potential feedstock for the production of tartaric acid, which is used as a preservative in the food industry, as well as lactic acid and xylitol [9]. Grape pomace and vinasse can be used as substrate for the production of protein-rich fungal biomass.

The *skin* of grapes can be used for the production of natural dyes which can be later used in textile industry [38].

Wastewater from wineries can be used as a substrate for the cellulose production as well as for single-cell proteins production that can serve as food or feed additives [9].

In the cascading approach (Fig. 4.), the product with the highest value is produced first, then the second-highest, and so on. In this context, the solid residue of grape pomace or seeds left after the extraction of phenolic compounds or oil production is still rich in nutrients such as proteins and fiber and can be further processed

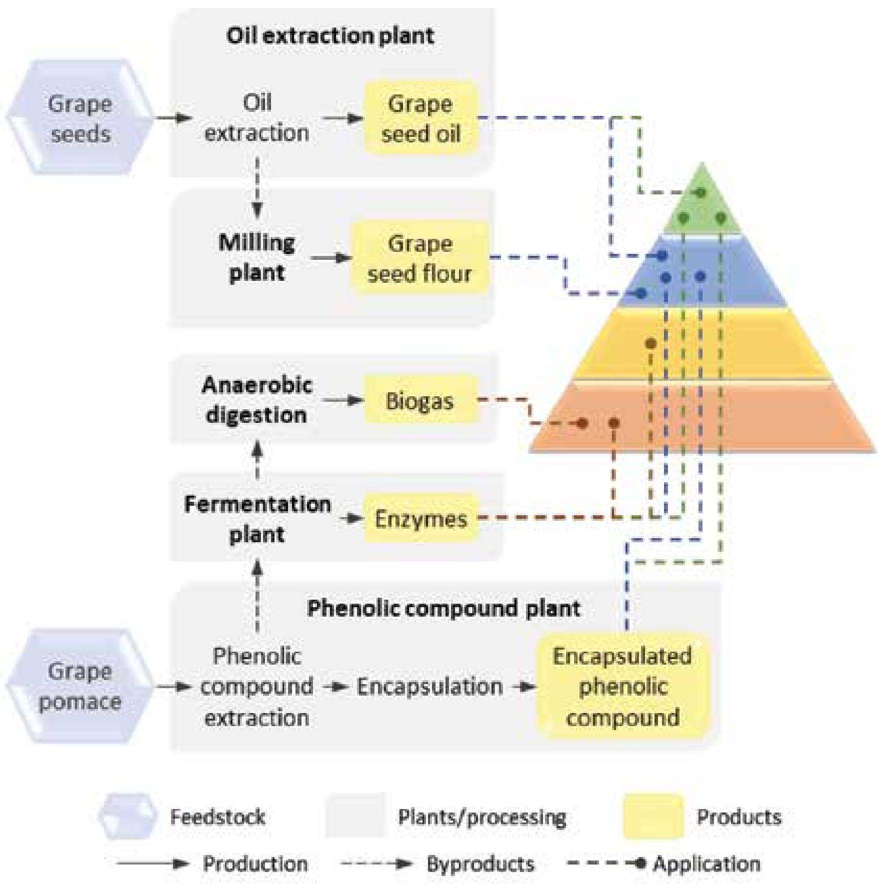

Fig. 4. Scheme of a cascading biorefinery proposal based on main winery residues

into flour, which can be used as an additive in bakery products or as a feed ingredient. In addition, this residue can be used for the production of pellets with a slightly lower heating power than the original grape pomace and for the production of biochar. In addition, the grape pomace remaining after SSF process can be used for bioethanol and biogas production.

5. Conclusion

The circular bioeconomy through the concept of biorefinery offers the possibility of efficient use of biomass, including organic residues from wineries, for the sustainable production of high value-added products. The advantages of the circular bioeconomy over the linear one are better resource and environmental efficiency, lower greenhouse gas emissions, reduced dependence on fossil resources, and valorization of waste biomass. This approach focuses on recycling, reusing, reducing, and recovering (4R-approach), and maintaining a sustainable production process to produce useful organic products. Since biorefinery requires capital investment to economically justify the conversion of industrial residual biomass into high value products, integrated technologies must be introduced to produce multiple products within a biorefinery concept (co-production of bioenergy such as bioethanol, biogas, electricity, heat, and bioproducts such as biochemicals, biomaterials, bioplastics, food, feed). The use of winery residues through cross-sector integration and cascade use of feedstocks has the potential for full valorization into high value products within the biorefinery concept.

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Sandra Budžaki, Ivica Strelec, Natalija Velić

Transformation of Eggshells, Spent Coffee Grounds, and Brown Onion Skins into Value-Added Products

Josip Juraj Strossmayer University of Osijek, Faculty of Food Technology Osijek, Franje Kuhača 18, 31000 Osijek, Croatia (sandra.budzaki@ptfos.hr)

Abstract

Unlike other wastes from the agri-food industry (straw, corn, cob, stalks, seeds, husks) widely used in biorefineries as feedstock for the production of high-value products (chemicals, biofuels and bioenergy), eggshells, spent coffee grounds, and brown onion skins have not yet found a suitable place in sustainable production, but are mainly landfilled burdening the environment. This paper aims to point out the great potential of eggshells, spent coffee grounds, and brown onion skins as secondary raw materials in sustainable development, with minimal production of waste streams.

Keywords: *eggshells, spent coffee grounds, brown onion skins, value-added products, "zero-waste" model*

1. Introduction

Sustainable production from its conceptual introduction by Agenda 21 [1] to the present time represents an industrial development that meets the needs of current generations without compromising the ability of future generations to meet their needs [2]. It should be noted that this definition does not include only activities related to the environmental protection, but also a whole range of activities related to the protection of natural, cultural and social values that are closely connected to material goods. Therefore, sustainable development includes a series of technical, technological, economic and social changes carried out by the needs of present and future generations. One of the pillars of sustainable development is the circular economy model, which aims to ensure sustainable development at every stage of product creation, processing and transformation by creating a "closed-loop" economy [3]. The new value is created through a "closed loop", approaching a model that avoids the creation of waste, i.e., the "zero waste" model. One of the five major challenges of sustainable waste management in the agri-food industry using the "zero-waste" model [4] is the development of innovative waste utilization techniques for the production of chemicals, fine chemicals, bioactive compounds, enzymes, and functional materials (Fig. 1.). These products have at least twice the added value of the products derived from currently prevailing outdated waste management strategies, which are not consistent with sustainable development. Outdated waste management strategies result in low value-added products such as animal feed, treated waste from the processes of composting, anaerobic digestion, and incineration, which can also have a negative impact on the environment and ultimately and most undesirably result in landfilling [5].

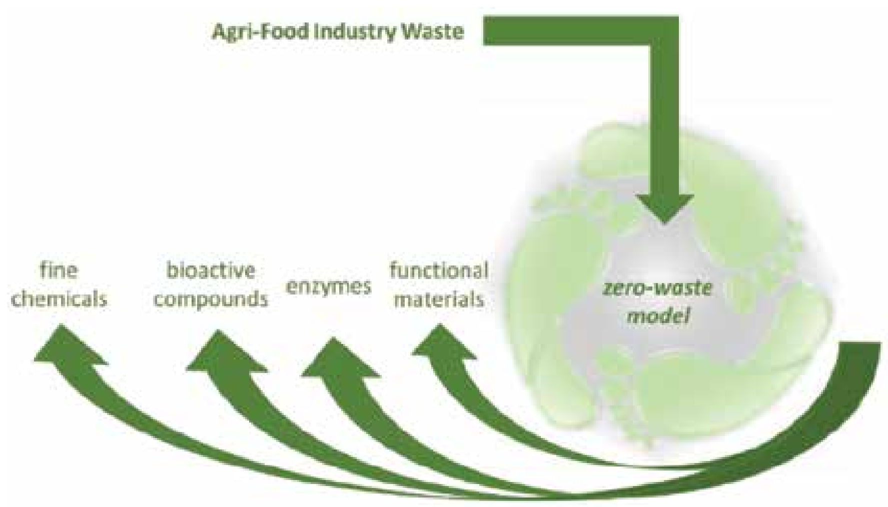

Fig. 1. Schematic presentation of possible products within "closed loop" economy from agri-food industry waste

Since food industry produces large amounts of waste, which mostly present an environmental problem, there is a need for its characterization and valorization in order to develop a model of its cost-effective recovery. Waste from the agri-food industry has the potential for the production of very valuable semi-finished products and products intended for further use in the food, pharmaceutical and biotechnology industries, and the concept of valorization is being intensively introduced into the management of agricultural and food waste [6].

Among the numerous wastes generated in the agri-food industry, eggshells, spent coffee grounds and brown onion skins are particularly "suitable" candidates (raw materials) for the preparation of enzymes (lysozyme), fine chemicals and/or functional materials as carriers for the immobilization of enzymes. The latter is supported by the available information on their structure, chemical composition and current knowledge on their potential use. The usage of innovative transformation techniques of this waste could result in protein-based carriers (eggshell membrane) and cellulose and/or hemicellulose based carriers (spent coffee grounds and brown onion skins). This has also been proved by research within the framework of a project funded by the Croatian Science Foundation "Immobilization of Lipases on Functionalised Carriers Produced from Selected Agro-Food Industrial Waste – ImoLipWaste" conducted by a research group at the Faculty of Food Technology of Osijek.

2. Are the eggshells a waste?

The eggshells are waste from the agri-food, pharmaceutical and biotechnology industries, family farms and crafts, but also restaurants and households, whose total annual production can be estimated based on data on the total mass of hen eggs produced, and the fact that the eggshell accounts for between 10–11% of the total egg mass [7]. According to the Food and Agriculture Organization (FAO), a total of 78949,623 t of chicken eggs was produced in the world in 2018, of which 7,770,000 t in the European Union, including the United Kingdom, and 47,150 t in the Republic of Croatia [8]. Consequently, it can be estimated that in 2018 the world generated at least 7,894,962 t of eggshell as waste, of which 777,000 t in the European Union and 4,715 t in the Republic of Croatia. It is crucial to emphasize that about 15 to 20% of the total eggs produced are further used in chicken hatcheries, and after hatchery remained eggshells becomes a waste.

The research by [9,10,11] indicates the potential of the eggshells as a carrier for enzyme immobilization. In addition, the eggshell is a very "cheap" source of natural calcium in the form of calcium carbonate, from which inorganic and organic calcium salts can be produced by relatively simple processes. Organic and inorganic calcium salts are used in the food, pharmaceutical and chemical industries (additives, food supplements), as well as for the production of calcium-based fertilizers for agricultural purposes [7]. Among the numerous methods for obtaining calcium salts from eggshells, the best known are those based on dissolving calcium carbonate from calcified matrix in dilute solutions of acids such as hydrochloric, acetic and o-phosphoric acid. In addition to calcium salts, eggshell membranes are also formed as a by-product during dissolution with acids (Fig. 2.). In addition to being an inexpensive source of collagen and hyaluronic acid for the pharmaceutical and cosmetic industries [7], eggshell membranes also have great potential to be used as a carrier for enzyme immobilization [12]. This is supported by numerous studies focused on the development of biosensors based on the enzyme immobilization on eggshells membranes as solid support [13]. When the production of calcium salts by eggshell treatment with dilute acids is compared to the the eggshell membranes production for biosensor development, it can be observed that ground eggshells are used for the production of salts, and intact eggshells or eggshell halves for biosensor related eggshells membranes production. Moreover, in biosensor development, partial decalcification of the eggshells by

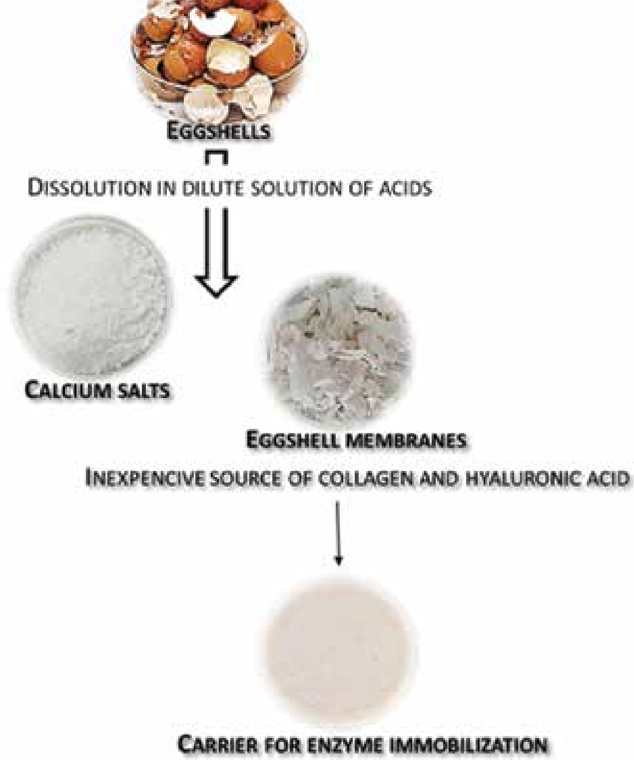

Fig. 2. Possible "zero-waste" model of eggshell waste transformation

acids is performed in order to facilitate the separation of large pieces of the eggshell membranes from the remaining eggshells. Accordingly, it seems quite likely that these two types of production processes could be combined for simultaneous production of calcium salts and larger pieces of eggshell membranes, using halves and/or larger pieces of eggshells instead ground ones. This would greatly facilitate the process of eggshell membrane separation and handling. Furthermore, produced eggshell membranes could be uniformly milled to particle size necessary for enzyme immobilization.

The exploitation of eggshells for the production of value-added products has great potential, related to its rich composition of various high-value compounds, Eggshells have already found its application in the production of feed for livestock and pets, organic fertilizers and soil improvers, compost and biogas, as well as, raw materials for cosmetic industry, active implantable medical products, in vitro diagnostic medical devices, veterinary medical products and for the pharmaceutical industry. By use of innovative transformation techniques and advanced technical and technological solutions, it is possible to prevent the occurrence of risks for humans and animals and to use eggshells for the production of products for human consumption.

3. Hidden treasure behind a cup of coffee

Coffee, a beverage made from roasted and ground coffee beans, is one of the three most popular beverages besides tea and water. Coffee is a tree-like plant shrub from the *Rubiaceae* family, and originates from the Ethiopian province of Kafa. The average annual coffee production in the world exceeds 10 million tons, leaving behind about 6 million tons of spent coffee grounds after the production and consumption of about 2.3 trillion cups of coffee [8]. The aforementioned seems to be quite a good reason for the serious exploration of the possibility of using spent coffee grounds for the production of value-added products and/or secondary raw materials, rather than its landfilling. Namely, spent coffee grounds possess a significant nutritional value, as well as, large proportion of various valuable biocomponents, which can be further used in pharmaceutical, biochemical/chemical, food industry and civil engineering. According to the literature [14] spent coffee grounds contains about 60% of water-insoluble lignocellulosic material or about 50% of insoluble fibers in dry matter with domination of cellulose and hemicellulose, about 13.6-17.5% proteins, 10-15% lipids, 4% of total polyphenols and 2.5% of condensed tannins in dry matter. Since spent coffee ground components differ in polarity, they can be isolated by the use of standard

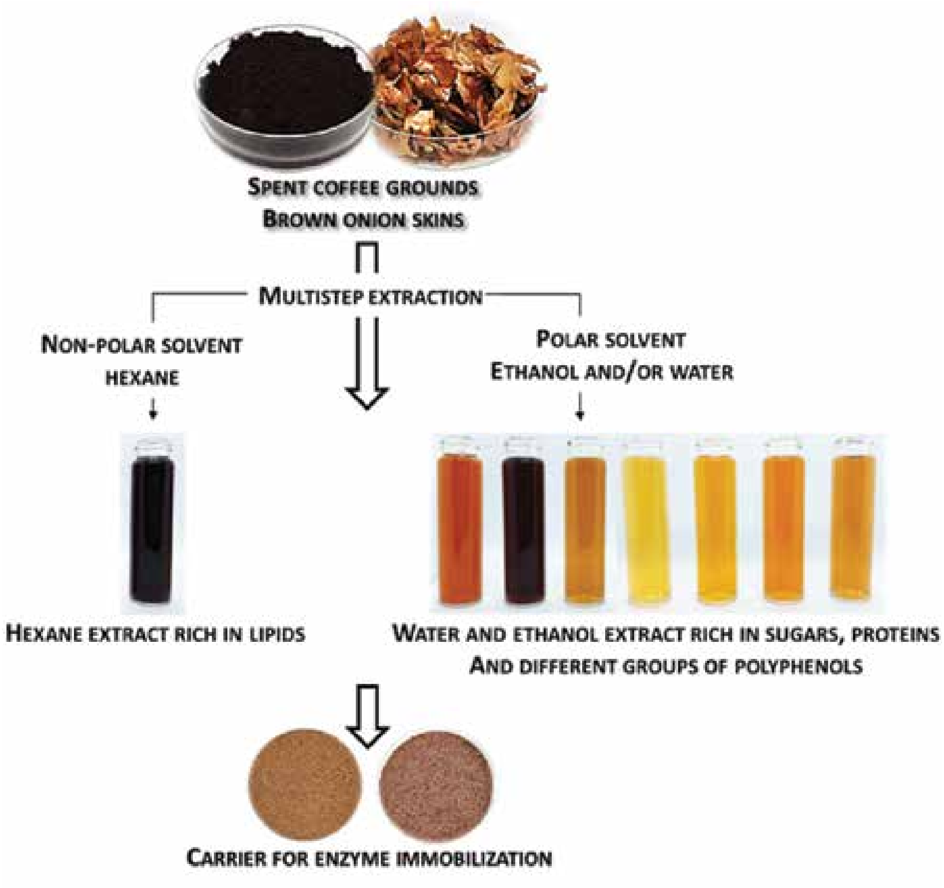

Fig. 3. Possible "zero-waste" model of spent coffee grounds and brown onion skins transformation

extraction methods with different solvents ranging from non-polar to polar, as well as by innovative extraction techniques (Fig. 3.) including supercritical CO_2 extraction and subcritical ethanol and water extraction [15].

4. All sides of the brown onion skins

For the most of the world's population, onion (Allium cepa L.) is a daily part of the diet and one of the oldest cultivated plants in the world. According to FAO 104 million tons of onions were produced worldwide in 2020, with 16,350 tons in Croatia [8]. With the increased need for processed onions, the production of waste biomass from onions also increases. About 37% of the total mass of fresh onions is classified as waste during processing. Depending on the type of onion processing, two types of waste are distinguished. Waste biomass of onions after calibration and packaging is the outer shell of the so-called onion skin, while waste biomass of onions lagging behind industrial onion production includes onion skins, two outer fleshy layers, roots, upper and lower bulbs, and waste includes small, deformed, diseased or damaged bulbs [16]. This type of onion waste is a major problem for the industry because it cannot be used as animal feed due to its characteristic strong and sharp smell, and it cannot be used as an organic fertilizer due to the development of phytopathogens such as *Sclerotium cepivorum*. The use of onion waste as a potential source of functional ingredients is one of the ways to valorize it (Fig. 3.), especially with food industry modern trends connected to the growing need for the production of functional foods. Nevertheless, the chemical composition of onion waste can vary significantly and therefore it is necessary to determine the content of target compounds separately. However, onion waste is generally identified as a source of flavor compounds, fibers, non-structural carbohydrates and polyphenols, which opens opportunities for its further use as a source of functional food ingredients to improve antioxidant and prebiotic quality of new products [17,18].

5. ImoLipWaste Project

Waste from the agri-food industry, which is widely available, could be used for the production of "cheap" enzyme immobilization carriers, subsequently leading to the reduction in waste disposal costs, due to the usage of produced waste for carrier production and selling, instead of cost-effective waste disposal. In the long run, such an approach could make an additional profit, instead of the costs in the industry, especially if bioactive components present in the extracts would be further used as additional products. Such an approach has been used within the ImoLipWaste project research where appropriate techniques for the transformation of eggshell, spent coffee ground and brown onion skin wastes into functional materials based on protein fibers (Fig. 2.) and/or lignocellulosic material (Fig. 3.) for enzyme immobilization was used, and obtained by-product extracts for further use/production was evaluated. Based on the project preliminary research, it seems that economy-based production of "cheap" carriers for enzyme immobilization from eggshells, spent coffee grounds and brown onion skins, is inevitably connected to the production of bioactive components present in the extracts during carrier preparation, where achieving of the greatest profit using "zero-waste" model approach can be obtained. Among the variety of bioactive compounds present in the aforementioned waste, the most prominent was found lysozyme (eggshells), caffeine (spent coffee grounds) and quercetin (brown onion skins).

The material remaining after the extraction of bioactive compounds from eggshells, spent coffee grounds and brown onion skins, presents the carriers for lipase immobilization, which can be used in food, biochemical, chemical and/or pharmaceutical industry. The usage of the immobilized lipases reduces production and operational costs due to the fact that immobilized lipases can be re-used in the production process. Moreover, such an approach enables sustainable industrial production and creates added value for waste/by-products of the agri-food industry, which is the basis of circular management. Furthermore, immobilized lipases open the possibility for developing new production technologies, such as the transition from batch to continuous processes. The introduction of continuous processes instead of batch processes reduces the size of the reactor and therefore the investment costs, facilitates process control in the event of minor deviations in product quality.

6. Conclusions

It is to be expected that in the next few decades new and/or improved existing technologies will follow the trend of sustainable production, enabling the better utilization of agri-food waste for everything for which fossil reserves are still used today. Besides its potential use in the energy production (bioenergy/biofuels), agri-food industry waste clearly represents the material for the production of multiplicity of various high-value added biomaterials where approach to the "zero-waste" model can be achieved. However, such an approach clearly requires the use of innovative techniques.

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Jurislav Babić[1], Natalija Velić[1], Mario Panjičko[2], Goran Vujić[3], Dušan Milovanović[3], Milan Mirić[4]*

A Path to Sustainable Municipal Waste Management: From Engineering Practices to Education and Training

[1] Josip Juraj Strossmayer University of Osijek, Faculty of Food Technology Osijek, Franje Kuhača 18, 31000 Osijek, Croatia (natalija.velic@ptfos.hr)
[2] Centre for the development of Sustainable Technologies Ltd. –CROTEH, Zagreb, Croatia
[3] Faculty of Technical Sciences, University of Novi Sad, Novi Sad, Serbia
[4] Regional development agency Srem, Ruma, Serbia

Abstract

With population growth and increasing municipal solid waste generation, efficient and sustainable waste management strategies become important to protect both human health and the environment. Although the transition from the prevailing waste management systems, such as landfilling, to modern waste management systems based on the 3R principle (reduce, reuse and recycle) is imperative, it is still very difficult to achieve in many countries. The waste management systems applied are in line with the country's income level, and the sustainable cutting-edge technologies are almost exclusively applied in high- and upper-middle-income countries. Thus, the modernization of the waste management system is achieved mostly through the growth of the gross domestic product. Moreover, the efficiency of modern sustainable waste management systems depends not only on the implementation of appropriate technical and economic strategies, but also needs to be supported by the education of citizens and the training of waste management professionals. The paper gives an overview of the educational-professional project "EDUcation for CLImate Change mitigation in the municipal solid waste sector", which is part of the European Climate Change Initiative (EUKI).

Keywords: *municipal waste management, sustainability, landfills, training in waste management, greenhouse gas mitigation, EUKI*

1. Introduction

The protection of human health and the environment and the conservation of resources have been recognized as the main objectives of waste management. Nowadays, it is not enough to achieve these goals through proper waste management, but it is also necessary for waste management to be sustainable. Sustainable waste management not only takes into account the present generations but also extends these goals to future generations [1]. The increasing transformation of materials has a major impact on waste management. The transformation of materials was up to 5-6 tons per year in prehistoric population, while in modern population it is up to 80-90 tons per year today [2]. This level of material use has resulted in the global generation of approximately 2.01 billion tonnes of municipal solid waste annually, of which probably more than 33% is not managed in an environmentally sound manner (in a conservative sense) [3]. The waste hierarchy (Figure 1) is the most popular graphic in any presentation on waste management.

However, the question arises whether the hierarchy can be implemented equally in all countries, regardless of their level of development.

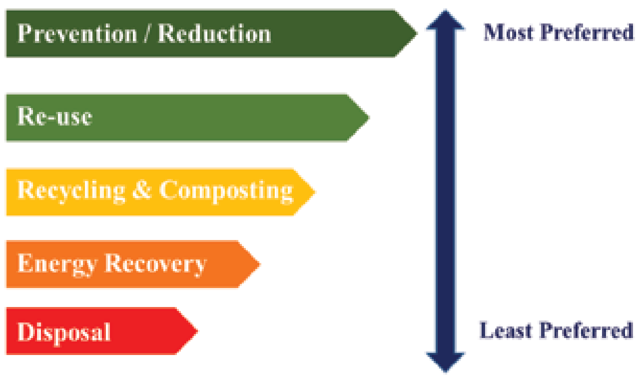

Fig. 1. Schematic presentation of the waste hierarchy

For a long time, the prevailing view was that landfilling and incineration of waste were the accepted options for municipal waste management. However, the development of modern waste management technologies has been driven by the fact that natural material and energy resources are not unlimited, and by the decline in landfill capacity in the 1980s [4]. In addition to prevention/reduction of waste generation, modern waste management systems include recycling and composting, as well as reuse of transformed materials wherever possible (Fig. 1). Many incentives encourage the transition to the aforementioned modern waste management systems (implementing the "3R" principle: reduce, reuse and recycle), as the far-reaching effects of these incentives are energy conservation and greenhouse gas emission reductions. The landfilling, either in sanitary landfills with gas collection systems or in unsanitary landfills or dumps, is still the most widespread municipal waste disposal system, responsible for 11% of all global methane emissions (making it the third-largest anthropogenic source of methane) [5]. The landfilled organic fraction of municipal solid waste initially degrades aerobically, producing only very small amounts of methane. However, once anaerobic conditions are established and methanogenic bacteria become active, the landfill begins to emit significant amounts of methane, which can continue for years (even for landfills that have completed their life cycle) [5]. Thus, the transition to more sustainable waste management systems is imperative. Even more in the EU, where a bold package of measures called the European Green Deal has been adopted in 2020, aimed at overcoming climate change and environmental degradation as the greatest existential threats to both Europe and the world. The plan to reduce greenhouse gases by at least -55% by 2030, compared to the levels in 1990 (set by the legally binding European Climate Law) aims to make EU climate-neutral by 2050. In addition to climate action, the European Green Deal also seeks to transform the EU into a "modern, resource-efficient and competitive economy by ensuring that economic growth is decoupled from resources and that no person or place is left behind" [6].

2. Waste management systems and country's income level

To what extent do the applied waste management systems correspond to the country's income level? In general, high-income countries generate approximately 34% of the global waste, even though they make up only 16% of the global population. However, the projected increase in waste generation for high-income countries by 2050 is much smaller (19%) than the increase projected for low- and middle-income countries (40%). This is likely due to the fact that an incremental increase in income leads to an increase in demand and consumption, which in turn leads to an increase in waste generation [3].

A crucial step in waste management is the collection. In high- and upper-middle-income countries, the collection rate is almost complete, while in low-income countries in urban and rural areas, only about 48% and 26% of the total amount of waste is collected, respectively [3].

As mentioned earlier, landfilling and open dumping are still the predominant methods of municipal solid waste disposal, accounting for 37% and 31% of the total amount of waste, respectively. Of this, 8% are landfills with gas collection systems. Recycling and composting account for 19%, and incineration for final disposal 11% of the total waste volume [3]. Modern sustainable waste management technologies that include different waste transformation processes, such as recycling, composting, energy recovery, biogas and other alternative fuels production are used almost exclusively in high- and upper-middle-income countries [3]. Even though low-income countries mostly rely on open dumping (93%), the chances of a successful transition from open dumping (or other rudimentary waste management technologies) to more advanced and sustainable waste management methods are high if locally appropriate solutions are implemented [3].

The Environmental Kuznets curve (EKC) attempts to explain the economic correlation between a society's standard of living and the degradation of the environment [7].

The hypothesis underlying the EKC was first formulated in the early 1990s by researchers Grossman and Krueger, who asserted a negative scale effect of economic growth on the environment-pollution increases as economic activity intensifies. However, technological progress and changes in the structure of production leading to a transition from capital-intensive industrial activities to a service economy can reverse this negative effect and reduce pollution [7].

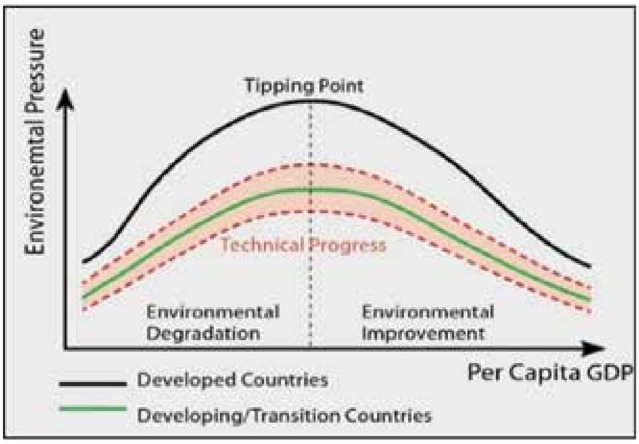

Fig. 2. The Environmental Kuznets curve

The same hypothesis can be applied to waste management. The EKC confirms that it is impossible to skip certain steps in the development of the waste management system in underdeveloped countries, which means that transition countries must find a less costly solution to waste management problems [8].

In order to achieve some technical progress in waste management and a reduction in the negative impact on the environment, a simultaneous growth in the gross domestic product (GDP) is required. Above a certain threshold, as income increases, so does environmental awareness and demand for improving environmental quality, as well as the willingness to fund the steps that lead to that quality improvement [7]. In other words: only through the growth of GDP, and thus the living standards of the population can the modernization of the waste management system with advanced technologies be achieved.

3. The importance of education and training in the field of municipal solid waste management

The efficiency of modern sustainable waste management systems does not depend solely on the implementation of appropriate technical and economic strategies. Environmental education, and particularly waste management education, is one of the pillars of successful waste management because it changes attitudes and provides learners with knowledge, values, and skills that enable a change in the way communities manage waste. i.e. it raises public awareness and enables "effective participation in the implementation of the waste management system" [9].

The case study of the city of Toyama in Japan shows that education is the key to sustainable waste management, because the most advanced waste management technologies and practices will not be sustainable if they lack citizen participation. Moreover, without citizen education, they will not be sustainable in the long run. Waste management in Toyama is based on the "3R" principle and aims to become a recycling-based society by integrating comprehensive education and waste management. This is also implemented in key municipal documents (Toyama City Basic Environmental Plan and City Basic Plan for General Waste Disposal). One of the most important examples within the Toyama city model is Eco-Town, an educational centre for waste recycling, which aims to raise citizens' awareness of the methods and importance of recycling and to promote cooperation among all stakeholders in waste management: citizens, businesses and the government [10].

In addition to waste management that targets citizens of all ages and backgrounds, training waste management professionals is a vital aspect of a sustainable waste management system. The common duties of waste management professionals among others include the development of contaminated or/and hazardous waste disposal procedures, development of storage protocols and recycling programs, management of waste facilities, providing outreach and marketing, working with accounting and budgetary milestones, selling waste materials to third parties, assist with the development of information and promotional materials, aim to meet waste reduction and recycling targets [11]. The list of duties and responsibilities of a higher-level waste management officer is even more complex, as they must be in line with all technological, organizational and legal developments and practices in the field of waste management. This also requires appropriate qualifications, e.g. a degree in waste management or similar (biological or biochemical sciences, chemical and physical sciences, civil engineering, structural or mechanical engineering, earth sciences, environmental sciences, etc.).

Considering all this, and the aforementioned fact that the best waste management technologies cannot work and be considered sustainable without human contribution (in this case a skilled waste management professional), the establishment of training centres for waste management professionals is extremely important.

4. EDU-CLIC: EDUcation for CLImate Change mitigation in the municipal solid waste sector

The educational-professional project "EDUcation for CLImate Change mitigation in the municipal solid waste sector" is a part of the European Climate Initiative (EUKI) [12]. EUKI is a project financing instrument by the German Federal Ministry for Economic Affairs and Climate Action (BMWK). The EUKI call for project ideas is implemented by the Deutsche Gesellschaft für Internationale Zusammenarbeit (GIZ) GmbH. It is the overarching goal of the EUKI to foster climate cooperation within the European Union (EU) in order to mitigate greenhouse gas emissions.

The project is carried out by 5 members of the project team from the Faculty of Food Technology Osijek (PTFOS), reinforced with an external expert in municipal waste treatment from the company CROTEH Ltd.. In addition to the PTFOS project team, there are also 5 team members from the Faculty of Technical Sciences, University of Novi Sad, Serbia and 4 members from the Regional Development Agency Srem, Serbia.

The activities planned under this project aim to establish the infrastructure for a training center for landfill operators and to develop a plan to upgrade the

Sremska Mitrovica regional landfill as a pilot landfill to achieve a long-term goal - to reduce greenhouse gas emissions in municipal waste management in Serbia.

Waste management in Serbia is mainly based on landfilling, which contributes significantly to the national carbon footprint. In fact, there are over 3,600 landfills in Serbia, most of which are unsanitary and illegal dumps. Annual methane emissions from these landfills amount to 60,000 tons [13]. The Serbian National Waste Management Strategy from 2010 to 2019 (Official Gazette of RS - 29/2010) [14] provides for the closure and remediation of all current unsanitary municipal solid waste landfills and the construction of 29 regional sanitary landfills, as well as recycling yards and transfer stations. According to the Strategy and the Law on Waste Management, the optimal solution for waste management in Serbia is proposed to be the establishment of regional centers based on the construction of sanitary landfills with additional treatment technologies and covering at least 250,000 inhabitants. So far, only 11 sanitary landfills have been built since 2002, and most of them do not comply with the operating procedures for modern landfills. In Serbia, there are no composting plants or other facilities for organic waste treatment, except for some pilot projects for green waste composting. However, the strategic documents envisage the construction of several mechanical biological treatment, anaerobic digestion and composting facilities.

A major problem in Serbia is the implementation of the system of source separation [15]. The lack of a systematic solution, regulations and public awareness of the need for source separation of waste as the first and crucial step towards the implementation of a functioning waste management system is a problem that needs to be solved as soon as possible at the national level. The implementation of a proper waste management system (separate collection, waste treatment, disposal on sanitary landfills) involves costs, and this additional financial burden should be borne by all citizens, which will be another major problem in the future.

This shows that there is a need to improve waste management infrastructure and practical education (training) of personnel involved in municipal waste management, especially landfill operators. As there are currently no formal or informal forms of such training in Serbia, the EDU-CLIC project seeks to meet this need while mitigating the negative environmental impacts of landfills and reducing methane emissions from landfills in the long term. A plan for upgrading the Sremska Mitrovica Regional Landfill will be developed within the project in order to obtain a modern waste management center. The developed plan for upgrading the landfill will serve as a blueprint for other regional landfills in Serbia. In addition, the project envisions the establishment of infrastructure

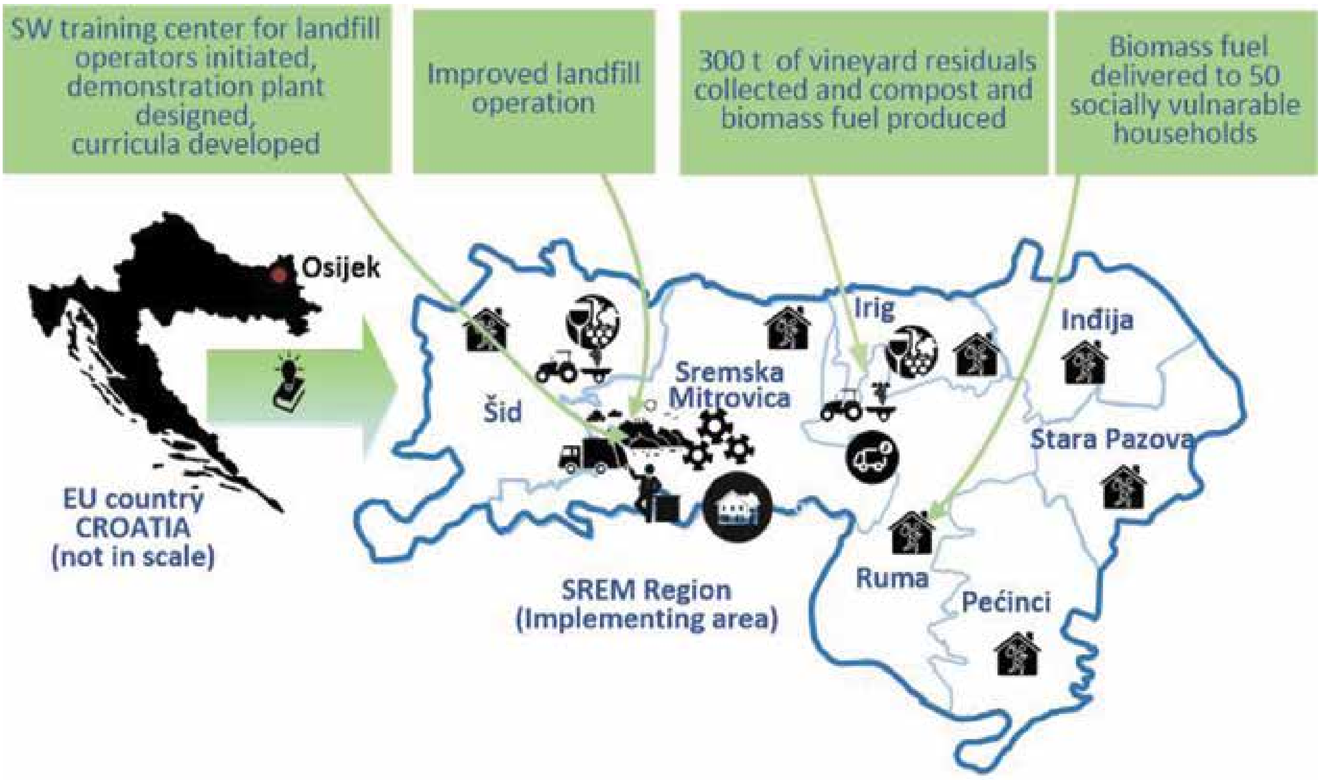

Fig. 3. Schematic representation of the project results

for a training center at the Sremska Mitrovica Regional Landfill and the development of a curriculum for training employees and operators in the field of municipal waste management. In order to make the management of municipal waste at the Sremska Mitrovica Regional Landfill more sustainable, activities/strategies are also planned to reduce methane emissions by 16.65 tons per year. The agricultural waste (biomass) generated after vine pruning will be collected from vineyards in the region and used for the production of compost and firewood briquettes. Thus, two practical strategies for methane reduction are envisaged: (1) the use of compost as a daily landfill cover and (2) the conversion of green waste and agricultural residues into biomass fuel.

The unique feature of the project is that it aims to offer the renewable energy source as fuel for vulnerable groups, thus realizing the principles of the circular economy and the three pillars of sustainability: economic viability, social justice and environmental protection.

The entire municipal waste management sector in the region is expected to benefit from the EDU-CLIC project, as all stakeholders involved in municipal waste collection and disposal will gain insight into ways to reduce greenhouse gas emissions and improve overall quality of environment through effective landfill modernization and a developed training program.

5. Conclusions

The importance of adopting sustainable waste management systems and the transition from the less sustainable but still prevalent waste management strategies in low-income countries was outlined. In addition, the importance of education and training in waste management was also discussed. The EUKI project EDU-CLIC implemented in Serbia was presented, which aims to reduce greenhouse gas emissions and improve the overall quality of environment through the effective modernization of the selected landfill and a developed training program for waste management professionals.

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Activities of the Croatian Academy of Engineering (HATZ) in 2022

Auspices, Organization/Coorganization of Conferences

Auspices

- University of Zagreb Faculty of Textile Technology, 14th International Scientific and Professional Conference "Textile Science and Economy", University of Zagreb, Faculty of Textile Technology, Zagreb, January 26, 2022, Zagreb;
- Professional meeting "Buildings 3+, Security, comfort, quality", University of Zagreb, Faculty of Architecture, February 17 to 18, 2022, Zagreb, live lectures with live video streaming;
- 14th meeting of young chemical engineers, Faculty of Chemical Engineering and Croatian Society of Chemical Engineers and Technologists, February 24 to 25, 2022, Zagreb;
- International professional meeting „7th International Conference on Road and Rail Infrastructure – CETRA 2022", University of Zagreb, Faculty of Civil Engineering, May 11 to 13, 2022, Pula;
- 9th International conference "WATER FOR ALL", J.J. Strossmayer University of Osijek, Faculty of Food Technology, May 19 to 20, 2002, Osijek;
- International conference 19th Ružička Days "Today Science – Tomorrow Industry" , J.J. Strossmayer University of Osijek, Faculty of Food Technology, September 21 to 23, 2022, Vukovar;

Organization/Coorganization of Conferences

- International scientific conference „Printing & Design 2022", University of Zagreb Faculty of Graphic Arts), Zagreb University of Applied Sciences, (North University, Varaždin – Koprivnica, Croatian Academy of Engineering, May 12, 2022, Zagreb;
- 7th Croatian Congress of Microbiology with International Participation, Croatian Microbiology Society and Croatian Academy of Engineering, May 24 to 27, 2022, Sv. Martin na Muri;
- International scientific conferences: „ Conference on Sustainable Development of Energy, Water and Environment Systems – SDEWES", May 22-26, 2022, July 24-28, 2022, November 6-10, 2022;
- Conference „10th International Congress od Food Technologists, Biotechnologists and Nutritionists", Faculty of Food Technology and Biogtechnology, Croatian Academy of Engineering, November 30 to December 2, 2022, Zagreb;

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