Exploitation of Olive Mill Solid Wastes as Alternative Biofuel, Valorization and Applications

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***Abstract*—Fossil fuel resource exploitation during the last century has increased greenhouse gas emissions in the atmosphere and raised concerns about future reliable energy sources. Consequently, the planet faces climate change's effects due to human footprint. The biofuels market is advancing globally due to growing concerns linked to the extensive consumption of fossil fuels and their continuous global reserve depletion. Due to their zero-carbon footprint emissions, they are a rational option for sustainable global economic and environmental development. The agriculture economy contributes to the planetary biomass and agricultural wastes and is identified as an abundant renewable worldwide resource that may serve for primary and secondary biofuel production.**

The olive cultivation in Mediterranean countries presents a promising opportunity for sustainable energy production. The biomass generated from olive tree trimming, olive mill extraction, olive husk (OH), and olive mill wastewater (OMWW) can be harnessed for biofuel production. The conversion of lignocellulosic biomass into secondary biofuels necessitates costly technologies, making it commercially viable in Albania. However, due to its calorific power, applying OH as a primary fuel in different production activities is an important alternative.

In 2022, Albania's olive tree area amounted to ~ 50 x103 ha, yielding 157.7 thousand tons of olive fruit and producing 15,000 tons of olive oil, further emphasizing the potential of this resource. The olive oil extraction process generates significant quantities of OH, totaling almost 50,000 tons during the past three harvesting seasons. The extensive utilization and olive processing facilities pose challenges in waste management. Utilizing solid mill wastes as biomass and processing them into primary or secondary biofuels offers a viable and eco-friendly option that is both sustainable and ecologically beneficial. However, additional research is needed to develop a practical, economically efficient, and successful method for converting biomass to liquid biofuels. This underscores the importance of our work in this field.

***Keywords*—Olive Husk; Biomass; Primary Biofuels; Mediterranean Region; Albania**

I. INTRODUCTION

The rapid industrialization and motorization of the global economy have significantly heightened the need for fossil fuels, giving the designation anthropogenic era. Fossil fuels constitute 80% of the primary energy utilized worldwide, with the transport sector alone responsible for over 60%. Due to extensive utilization, their supplies are depleting globally [1].

Extensive use has negatively impacted the ecosystem because of the rise of greenhouse gas (GHG) emissions. As a result, the planet faces different effects of climate change [2], [3]. Consequently, the exploration of alternative sources of energy that are renewable, sustainable, efficient, and of economic interest has driven the application of biofuels. The use of biofuels is advancing globally due to growing concern about climate change linked to the extensive consumption of fossil fuels and their continuous global reserve depletion. It is also important to highlight that these alternatives are considered with zero-carbon emission rates [4], [5].

Biofuels have the potential to become strategically vital fuel sources among the various energy choices. Their applications have been closely linked to the development of automotive engines and may be traced back to the early 19th century [6]. Examples of such substances include ethanol, methanol, biodiesel, hydrogen, and methane. Those sides proved to be more ecologically sustainable. Despite the economic development status, the potential for manufacturing this category of items by all countries impacts the economy by diminishing reliance on imported petroleum [7].

Renewable and carbon-neutral biofuels are crucial for ensuring a sustainable environment and economy. Worries over the ongoing dependence on petroleum-based motor fuels primarily drive the growth of today's biofuel business [8]. Bioethanol and biodiesel are the primary forms of biofuel utilized. The global output of biofuels has expanded to over 190 billion liters annually and is projected to maintain its consistent increase (Figure 1) [9].

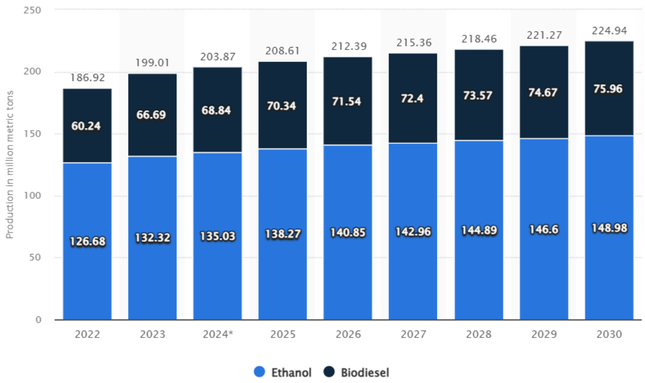

Figure 1. The 2023 biofuel production and 2030 worldwide forecast by product type (in million metric tons) [9].

II. PRIMARY AND SECONDARY BIOFUELS

Biofuels may be classified into two main categories: primary and secondary biofuels. Primary biofuels, such as firewood and pellets, are primarily used for heating and cooking. Secondary biofuels refer to primary fuels that have undergone processing and are now in the form of solid material (such as coal), liquid (such as ethanol and biodiesel), or gaseous (such as biogas and hydrogen) [5]. Secondary biofuels are classified into three categories, first, second, and third-generation, based on the raw materials utilized and the manufacturing process employed (Figure 2). Secondary fuels are utilized in many applications, such as transportation and high-temperature industrial activities [1].

Alcoholic fuels have the potential to substitute for gasoline, while biodiesel, green kerosene, and dimethyl ether (DME) are appropriate for use in diesel engines [10]. Various technologies exist to produce biofuels, including fermentation of sugar substrates, catalytic conversion of ethanol to mixed hydrocarbons, hydrolysis of cellulose, fermentation for the production of butanol, transesterification of natural oils and fats into biodiesel, hydrocracking of natural oils and fats, and pyrolysis and gasification of various biological materials [11].

III. RESULTS AND DISCUSSION

A. Secondary biofuels of 1st generation

First-generation secondary biofuels, produced from sugars, grains, or plant seeds, follow a relatively uncomplicated process. The most widely recognized biofuel of the first generation, ethanol, is derived from the fermentation of sugar found in agricultural plants such as corn kernels or other high-content crops [11]. This process, often involving organic material with a significant amount of sugar and yeast enzymes, converts six-carbon monosaccharide molecules, glucose, into ethanol. The technique involves the extraction of sugar from raw sources, with distillation and dehydration as the concluding stages to achieve the targeted level of bioethanol concentration. Bioethanol can then be mixed with fossil fuels or used as pure fuel. Hydrolysis transforms starch into glucose in the presence of cereals as raw ingredients further simplifies the process. When comparing the two carbohydrate reserves, starch and cellulose, it is evident that starch undergoes conversion into glucose at a significantly faster pace than cellulose [1].

Biodiesel is a prominent example of a first-generation biofuel. It is derived from vegetable oils by transesterification or cracking [12], [13]. Transesterification can be achieved by combining vegetable oil (triglycerides), alkaline, acidic, or enzymatic catalysts, and alcohol (methanol or ethanol) in the combination [6]. The resulting products from this procedure consist of methyl esters of fatty acids, often known as biodiesel and glycerin. It is important to note that the biodiesel manufacturing technique utilizes just a fraction of plant biomass as its raw material, leaving behind a significant portion as organic waste [13].

First-generation fuels are commercially produced in enormous quantities globally. Technological issues, such as chemical stability, accompany the development of first-generation biofuels. Another issue in employing biomass as a biofuel processing option is related to the same raw agricultural seed used as a food supply, a cause for controversy [5].

B. Secondary biofuels of 2nd generation

Two fundamentally different approaches generally produce second-generation biofuels. The first source is lignocellulosic biomass originating from agricultural activity, as non-edible waste produced by plants, and the second source is whole non-edible plant biomass (e.g., grass or trees grown specifically for energy production) [14], []. The main advantage of the production of second-generation biofuels from non-edible raw materials is that it limits the direct competition of the use of arable land for planting crops for the production of biofuels, affecting the 'food versus fuel' duality. This promising approach opens up new possibilities for sustainable biofuel production, providing a hopeful outlook [15].

Second-generation biofuel production requires more advanced technology, investment per production unit, and extensive-scale facilities for capital costs. Future ethanol production will include traditional cereal/sugar crops and lignocellulosic biomass [16].

The processes used to convert biomass into second-generation biofuels are classified as biochemical or thermochemical. Some second-generation biofuels, such as ethanol and butanol, are produced through a biochemical process, while other second-generation fuels are produced thermochemically. These thermochemical fuels include methanol, refined Fischer-Tropsch (LFT) liquids, and dimethyl ether (DME) [1], [17].

Several essential characteristics distinguish the thermochemical process from the biochemical process, including the flexibility of the feedstock and the diversity of the fuels produced [18].

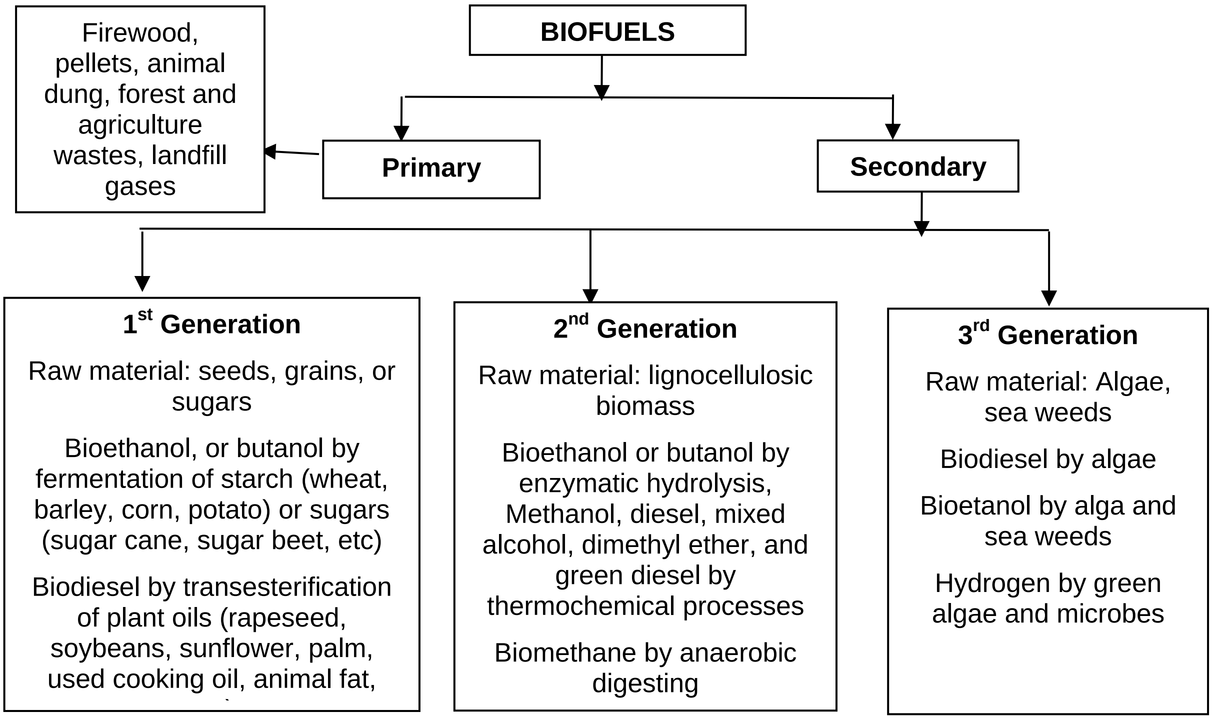

Fig. 2. *Biofuels classification according to Nigam and Sing (2011) [1].*

The use of 'agricultural' biomass, which refers to biomass derived from agricultural activities, in producing second-generation biofuels provides better land use efficiency than first-generation biofuels. Furthermore, the lower cost of processing materials and the use of 'non-edible' biomass, which refers to biomass not used for human consumption, favor the promotion of second-generation biofuels. Biofuels of the first generation have high production costs due to competition with land funds used to produce crops for food production. The rapid expansion of global production of biofuels from grains, sugar, and oilseeds has increased the cost of some food products. These limitations favor finding non-food biomass for biofuel production [19].

C. Bioethanol as biofuel

Bioethanol production encompasses biomass fermentation. Its use as a biofuel is crucial because it is considered 'renewable' and 'friendly' to the environment. The global first-generation bioethanol production in 2023 was about 132.32 million tons [9].
Crops such as wheat, sugarcane, and corn are the most essential natural biological resources used to produce bioethanol. Therefore, suitable raw materials for bioethanol production can contain fermentable sugars. Biomass sugars, which can directly ferment into ethanol, are the least complex method used in ethanol production [11]. Bioethanol production includes the pretreatment of substrates, the process of saccharification to release fermentable sugars from polysaccharides, the fermentation of sugars (monosaccharides), and finally, the process of ethanol distillation (Figure 3). The pretreatment procedure facilitates the separation of cellulose, hemicellulose, and lignin [20].

In this way, the complex carbohydrate structure of cellulose and hemicellulose face fragmentation by enzymatic hydrolysis into monosaccharides. Cellulose is a polysaccharide of glucose molecules. The complex structure requires treatment for depolymerization into glucose molecules, which proceed with fermentation into bioethanol by fermenting yeasts.
Lignocellulosic biomass, the world's most abundant organic raw material, will play a central role in the future when it comes to producing fuels, chemicals, and materials [20]. Hemicellulose contains, besides glucose, sugars with 5C atoms, such as xylose and pentoses. The fermentation process of hemicelluloses in ethanol is carried out on a more complex scale because it requires efficient microorganisms that can ferment sugars of this type. The structure of lignin consists of several aromatic alcohols and phenols [21]. It is not fermentable, but it can be recovered and used as fuel, providing the heat of the process, and through the depolymerization process, it converts into bio-oil. Cellulase enzymes speed up the process of hydrolysis. Substrates, cellulase activity, and reaction conditions affect how enzymes break down cellulose. Some problems that need to be solved are keeping microorganisms working well over time, developing better pretreatment technologies for lignocellulosic biomass, and combining the best parts of cost-effective ethanol production systems [16].

D. Butanol as biofuel

Butanol (C4H10O) has a longer carbon chain than ethanol and is better mixed with gasoline and other hydrocarbons. It is less volatile than gasoline or ethanol and produces higher amounts of thermal energy than ethanol. Butanol contains 110,000 BTUs per hydrocarbon unit, about the same as gasoline (115,000 BTU). Chemically, it is less corrosive than

ethanol and easily transported and distributed through existing pipelines and filling stations. An 85% butanol/gasoline mixture can be used in unmodified gasoline engines [14].
It was a very early industrial product in use, dating back to before the development of the petroleum industry. It was a product of starch fermentation, a complicated anaerobic process through Clostridium acetobutylicum strains in the mixture of acetone, butanol, and ethanol, where different enzymes facilitate the fragmentation of polymeric carbohydrates (Figure 4). This biochemical process is known as "acetone-butanol-ethanol (ABE) fermentation," resulting in a 3:6:1 mixture ratio [22].

Table 1.Cellulose, Hemicellulose, and Lignin Content in Agricultural Residues and Wastes [16].

Agricultural residue	Cellulose	Hemicellulose	Lignin
Wheat straw	33–40	20–25	15–20
Rice straw	40	18	55
Corn Cobs	45	35	15
Nutshells	25–30	25–30	30–40
Cottonseed hairs	80–90	5–20	0
Leaves	15–20	80–85	0
Solid cattle manure	1.6–4.7	1.4–3.3	2.7–5.7
Swine waste	6.0	28	
Primary wastewater solids	8–15	–	24–29
Paper	85–99	0	0–15
Newspaper	40–55	25–40	18–30
Sorted refuse	60	20	20
Waste papers (chemical pulp)	60–70	10–20	5–10

E. Olive oil production in Albania

Olive oil is a globally distinguished food product produced mainly in the Mediterranean Basin. It is a product with a closed production cycle, directly impacting the rural economies [24], [25]. Beyond the economic value, the olive oil production sector has created environmental problems [26]. The extraction process produces substantial amounts of solid waste, a composition of olive husk, crude olive cake (OH), olive mill wastewaters (OMWW), a mixture of vegetables, and added technological water. The OMWW is a smelling acidic red-to-black liquid with high conductivity. Its composition varies qualitatively and quantitatively according to olive variety, climate conditions, cultivation practices, storage time, and extraction process. It comprises 83-92% water, 4-16 % organic matter, and 1- 2% minerals [27].
The country's olive oil (OO) extraction industry is organized in small Olive Mills covering olive cultivation regions. The highest contribution in olive oil production is reached mainly in the Southern and Western areas and the river valleys [25]. Three-phase extraction technology dominates the OO industry (Figure 5) [26].

F. Olive Mill Solid wastes as biomass

In 2022, the olive tree area in Albania amounted to 49,476 hectares, with a total olive fruit yield of 157,710 tons and an olive oil production volume of 15,000 tons. Beyond the concern of their negative environmental impact, these wastes contain valuable resources of organic matter and nutrients. According to the equation proposed by De Ursinos et al. [28], the olive waste produced in Albania during the harvesting year 2022 is approx.—50 thousand tons OH.
Recently, an extraction plant collected olive cake and has implemented solvent extraction. Olive Mill Wastewaters may find the option of water irrigation and application. In addition, there is a lack of awareness from producers and legal institutions regarding the environmental pollution caused by the olive oil extraction industry. Vegetable waters are discharged directly to the surface streams. At the same time, olive husk is used as feedstuff for animals or, in some cases, is dried up and used as a calorific source in different situations [29].

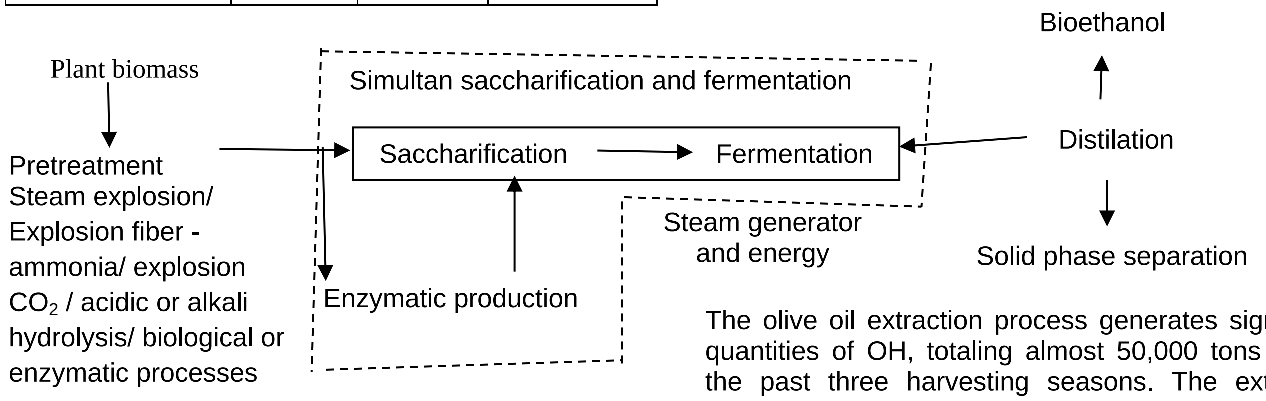

Fig. 3. *Bioethanol-producing processes from biomass.*

The olive oil extraction process generates significant quantities of OH, totaling almost 50,000 tons during the past three harvesting seasons. The extensive utilization and olive processing facilities pose challenges in waste management. Utilizing solid mill wastes as biomass and processing them into primary

or secondary biofuels offers a viable and eco-friendly option that is both sustainable and ecologically beneficial. OH is mostly utilized as a biofuel, serving as an energy source through combustion.
The conversion of lignocellulosic biomass into secondary biofuels necessitates costly technologies, making it commercially unviable in Albania. Additional research is required to develop a practical, economically efficient, and successful method for converting biomass to liquid biofuel.

Table 2. Calorific power of the Olive Mill solid waste according to the extraction system.

Solid waste type	Value (kcal/Kg)
Pressed spent olives	2800-3000
3-phase spent olives	2500-2800
Degreased spent olives	3500
Stone	4000

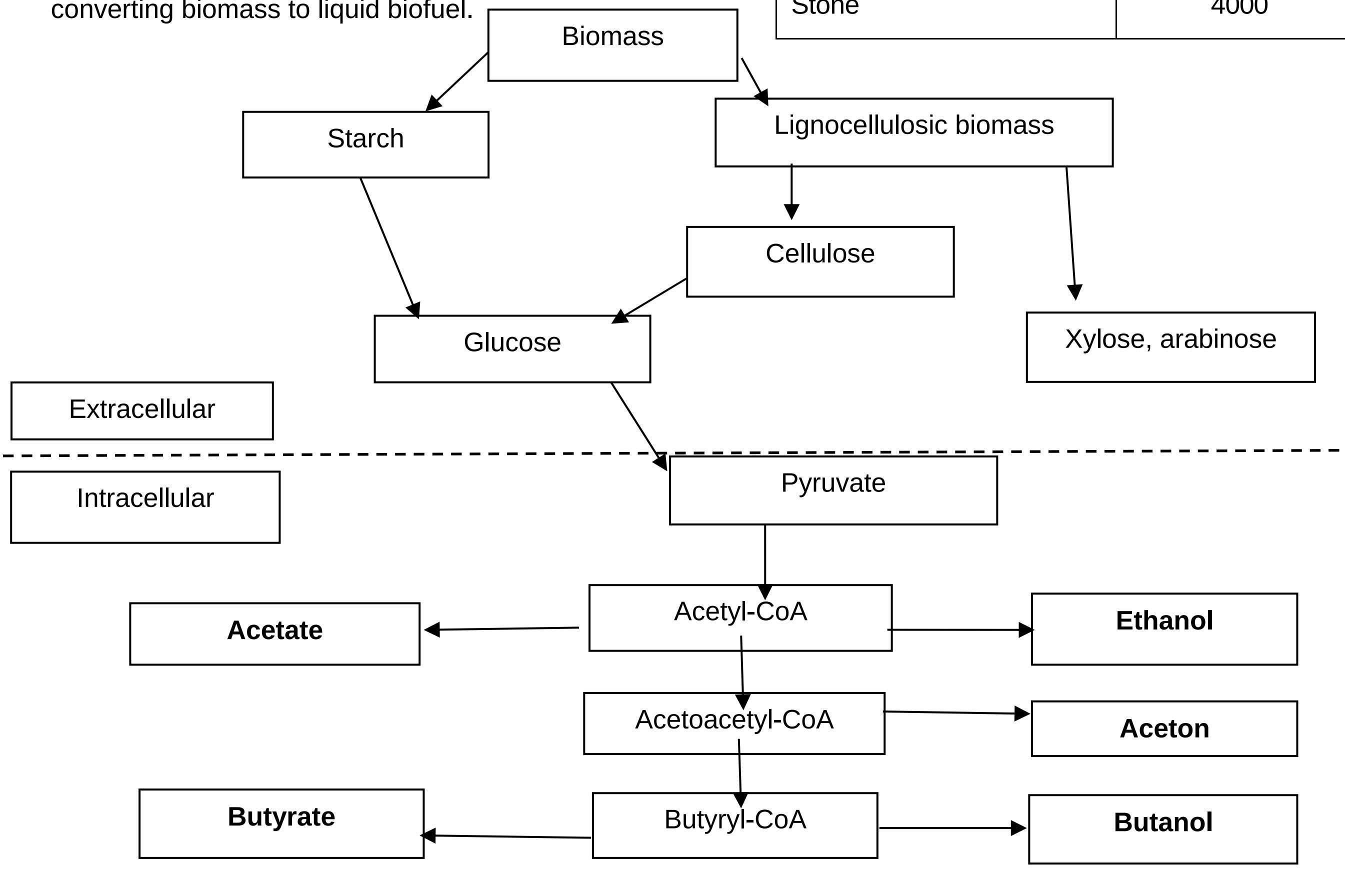

Figure 4. The production scheme of ABE mixture (Source: [23]).

G. Olive husk applications as primary biofuel

The olive husk is the primary residue generated in olive oil production, which contains a certain oil quantity that is not extracted physically. It is usually extracted in the plants to produce pomace olive oil. The composition of the olive husk solid waste depends on the extraction system applied. A pressed extraction system usually produces OH of 28.2% humidity, while a 3-phase extraction system produces OH (48.3% humidity).
For hundreds of years, OH has traditionally been used as fuel domestically or in oil mills to produce the heat necessary for extraction—the calorific power of the different by-products related to OH.
Waste has recently been used successfully as the first combustion material in bakery ovens. The first material is kept in space to remove moisture naturally, then deposited in storage areas without exposure to atmospheric agents. Therefore, it is interesting to study the energy efficiency of these industrial processes for both energetic and economic reasons.

Traditional baking combines hot air (natural convection), infrared radiation, and direct conduction (Ploteau, Nicolas, Glouannec, 2012). Both techniques are applied on an industrial scale, continuous and batch baking (Mansour et al., 2024). Applying renewable energy sources is a promising solution for technological processes like baking. The Mediterranean basin, considered the leading world producer of olives, possesses a vast potential to exploit the olive mill solid wastes as a renewable energy resource. Examples of batch-baking units that use OH are under activity in rural areas of the western region. A quantity of 150 kg of dry OH per cycle. Suppose a batch baking unit capacity of Rotary Oven Electric and Diesel- 240 Loaf has a total power of 75 kW. In that case, the replacement with OH disposed of with no cost is a promising solution for the future in the region. Finally, the exploitation of different options for applying OH biomass as biofuel will increase the sustainability of the rural areas in the Mediterranean region.

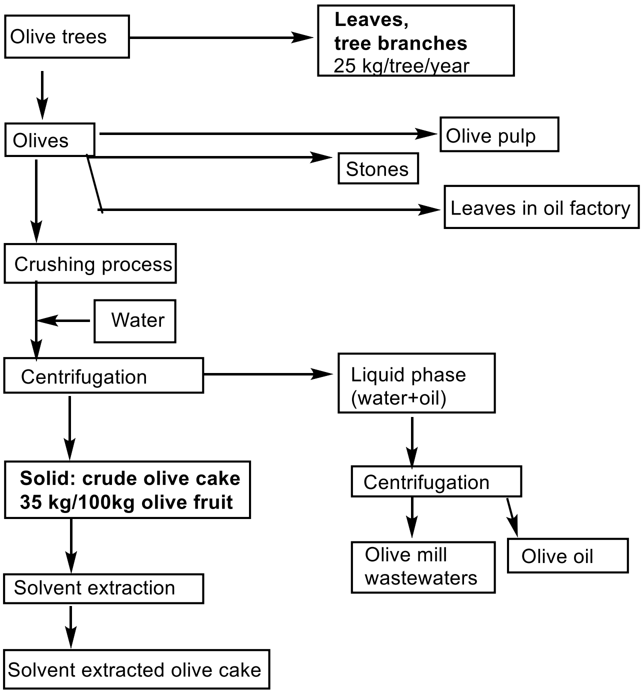

Figure 5. Olive tree and olive oil extraction by-product scheme (Source: [26], [28]).

IV. CONCLUSION

A substantial amount of olive mill solid waste is generated during olive cultivation and oil extraction. This amount is calculated to be almost 50,000 tons every harvesting season. The widespread use and processing facilities for olives make waste management difficult. Using solid mill wastes as biomass and converting them into primary biofuels, OH is mainly utilized as a biofuel, serving as an energy source through combustion. Meanwhile, to avoid the pressure of using arable lands for oilseed production as biomass for biofuels, we propose this alternative by using solid mill wastes as biomass and converting them into secondary biofuels, which provides a practical, environmentally favorable, sustainable, and ecologically advantageous choice. Conversion of lignocellulosic biomass into secondary biofuels requires expensive technology, which makes it economically impractical in Albania. Further investigation is necessary to design a viable, cost-effective, and prosperous technique for transforming biomass into liquid biofuels.

V. ACKNOWLEDGMENT

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