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A Comparative Study of Selected Properties of Biomass and Coal Fuels from Greece †

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Abstract: Three different types of biomass samples from Greece were studied (reed, olive kernel, and sawdust) and compared to lignite samples from coal deposits of Western Macedonia, Greece. All biomass samples exhibit higher calorific values (both as received and in dry basis) not only from the studied lignite but also from many other Greek coals. Despite its lower C content on a dry ash-free (daf) basis, the sawdust sample exhibits the highest calorific values, probably due to its lower total moisture content. It also exhibits the lower S and Cl content. As far as the ash content, reed and olive kernel presented higher values than the sawdust. Sylvite was identified in the biomass ashes. The presence of sylvite indicates the abundance of K in the organic matter that bonds with Cl and S during combustion. This explains the formation of arcanite that was identified in the reed and the olive kernel ashes. The presented differences in the composition of the ashes may lead to different environmental management or potential industrial applications.

Keywords: energy resources; Solid Fuels; proximate analysis; ultimate analysis; biomass; lignite

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1. Introduction

The continuous increase in the world's population and the growth of the economies of non-OECD countries such as China and India already have and will lead to a further substantial increase of energy demand in our very not so distant future [1]. The limited number of reserves of fossil fuels, on top of the Paris Agreement, are going to lead mankind to eventually search for new and non-emitting CO_2 sources of energy [2,3].

Renewable energy resources have become increasingly attractive due to their many advantages. They are getting cheaper [4], countries without fossil fuel deposits are becoming less dependent on wars and social anomalies while improving their energy security [5], and last but not least, renewables are environmentally friendly, emitting a minimum CO_2 and other potentially harmful gases (limited to the necessary for the production and the end-life management of the materials needed to produce the renewables) to the human health and the climate change. Moreover, biomass provides a renewable fuel source, which is carbon neutral and may decrease the carbon footprint by replacing lignite for electrical and thermal energy production. It has low sulfur, nitrogen, and potentially toxic elements' emissions [6]. It is one of those energy resources that are attractive due to their abundance in most countries of the planet, their low cost, and of course, their environmental efficiency, while it might lead as a major contributor to sustainable development. As a matter of fact, biomass has been used as an energy source since prehistoric ages by mankind for heating, cooking and industrial uses, way before the exploitation of fossil fuels. With the implementation of new regulations and directives for the restriction of fossil fuels consumption and the given motives for the exploitation of renewable energy resources, biomass seems a fitting solution for EU member states due to its abundance and its independence of specific climate conditions in contrast to wind or solar energy. In addition to that, the reusing of various wastes, which were mentioned above, contributes to the better environmental performance of each country [7]. During 2015, the percentage of energy production from renewables across the 28 state members of the European Union was 28.81% and biomass was the most significant source of renewables [8,9]. The highest energy production by sustainable resources was achieved in Sweden, while Finland and Latvia followed [10]. In the decade 2006 to 2016, there was a 66.6% increase in energy production from non-fossil fuel sources [9].

Today, the use of biomass is constantly increasing in the European Union and especially in Greece, where biomass is being increasingly exploited, holding 6th place among the 28 EU member states concerning the percentage of biomass used for energy production to the total renewable energy production [11]. The actual biomass quantity in Greece that derives from agricultural and light industry residues is a cheap energy solution that avoids the occupation of arable land while the residues are reused and exploited, contributing to the energy balance of the country. Specifically, olive trees' residue in Greece, could feed with their by-products 250,000 tn/a to biomass power plants [12] and contribute to the renewables' energy mix of the country.

Reed is a plant favored by Greek natural habitat [13] and may constitute a sustainable source of biomass for energy production. Another source of biomass in Greece could be considered sawdust, a solid waste derived from the small-sized carpentries around the country, as well as the medium and large-sized furniture industries. An essential criterion for the evaluation of the energy and environmental efficiency of all these different types of biomass is the determination of their calorific value, their elemental analysis (C, N, H, S), their moisture content, and their ash content and composition.

The main purpose of this study is to characterize the environmental and energy efficiency of different biomass, olive kernel residues, reed, and sawdust samples and compare their characteristics with Greece's main fuel for generating electricity, lignite [14]. It is noted that lignite's contribution to the energy mix of the country for electricity in 2015–2016 was about 32%.

2. Materials and Methods

Three different types of biomass samples from Greece were studied (reed, olive kernel residues, and sawdust) and compared to lignite samples from coal deposits of Western Macedonia, Greece. Reed was sampled from "Antonis Tritsis" environmental park, Ilion, Athens, Greece. Olive kernel and sawdust samples were provided from BIOGYPS KARVELIS S.A. and Polyeco S.A., respectively. Furthermore, one lignite sample from Achlada and one from Mavropigi lignite deposits (Western Macedonia, Greece) were provided and pulverized by the Center for Research and Technology of Hellas (CERTH). These samples were characterized by standard methods for proximate and ultimate analysis, ash composition, chlorine and sulfur content, and heating values (Table 1). Moreover, PXRD data and SEM images of their ashes were used for the mineralogical, morphological, and microstructure characterization of their ash yields. EDX micro-analysis was applied for the determination of the ashes' mineral chemistry.

Table 1. Standard analytical methods used for the analysis of the biomass and lignite samples, respectively.

Test	Biomass Samples	Lignite Samples
Total moisture	EN 14774 [15]	ASTM D7582 [16]
Ash content	EN 14775 [17]	ASTM D7582 [16]
Calorific value	EN 14918 [18]	ASTM D5865 [19]
Carbon	EN 15104 [20]	ASTM D5373 [21]
Hydrogen	EN 15104 [20]	ASTM D5373 [21]
Nitrous	EN 15104 [20]	ASTM D5373 [21]
Sulphur	EN 15289 [22]	ASTM D3177 [23]
Chlorine	EN 15289 [22]	ASTM D4208 [24]

Shredding of the reed and lignite samples took place in Polyeco's laboratories, (Aspropyrgos, Greece). Determination of the calorific value was implemented at the Solid Fuels Laboratory of the CERTH, using a bomb calorimeter PARR 6400 (Table 1). Sulfur and chlorine content were determined in the same laboratory using a closed pressurized bomb and ion chromatography system (Table 1). A Perkin Elmer 2400 CNHS/O series II elemental analyzer was used for elemental analysis of C, H, and N (Table 1) in CERTH.

The combustion of the biomass and the lignite samples took place in the laboratories of Economic Geology and Geochemistry of the Department of Geology and Geo-environment of the National and Kapodistrian University of Athens (NKUA), following the EN 14774 [15] and ASTM D7582 [16] standard procedures.

X-ray powder diffraction (XRD) data were obtained at the Department of Geology and Geoenvironment, NKUA, using a Siemens Model 5005 X-ray diffractometer (Bruker AXS GmbH., Karlsruhe, Germany), CuKα radiation at 40 kV, 40 nA, 0.020 degrees/s step size and 1 s step time. The XRD patterns were evaluated using the EVA 10.0 program of the Siemens DIFFRAC (Bruker AXS GmbH., Karlsruhe, Germany) and the D5005 software package (NKUA).

The morphology, microstructure, and composition of the biomass and ash samples were studied by scanning electron microscope (SEM JEOL JSM 5600, operated at 20 KV). The samples were carbon plated and positioned on a conductive sticker. Micro-photographs of the samples were collected during SEM observations.

3. Results and Discussion

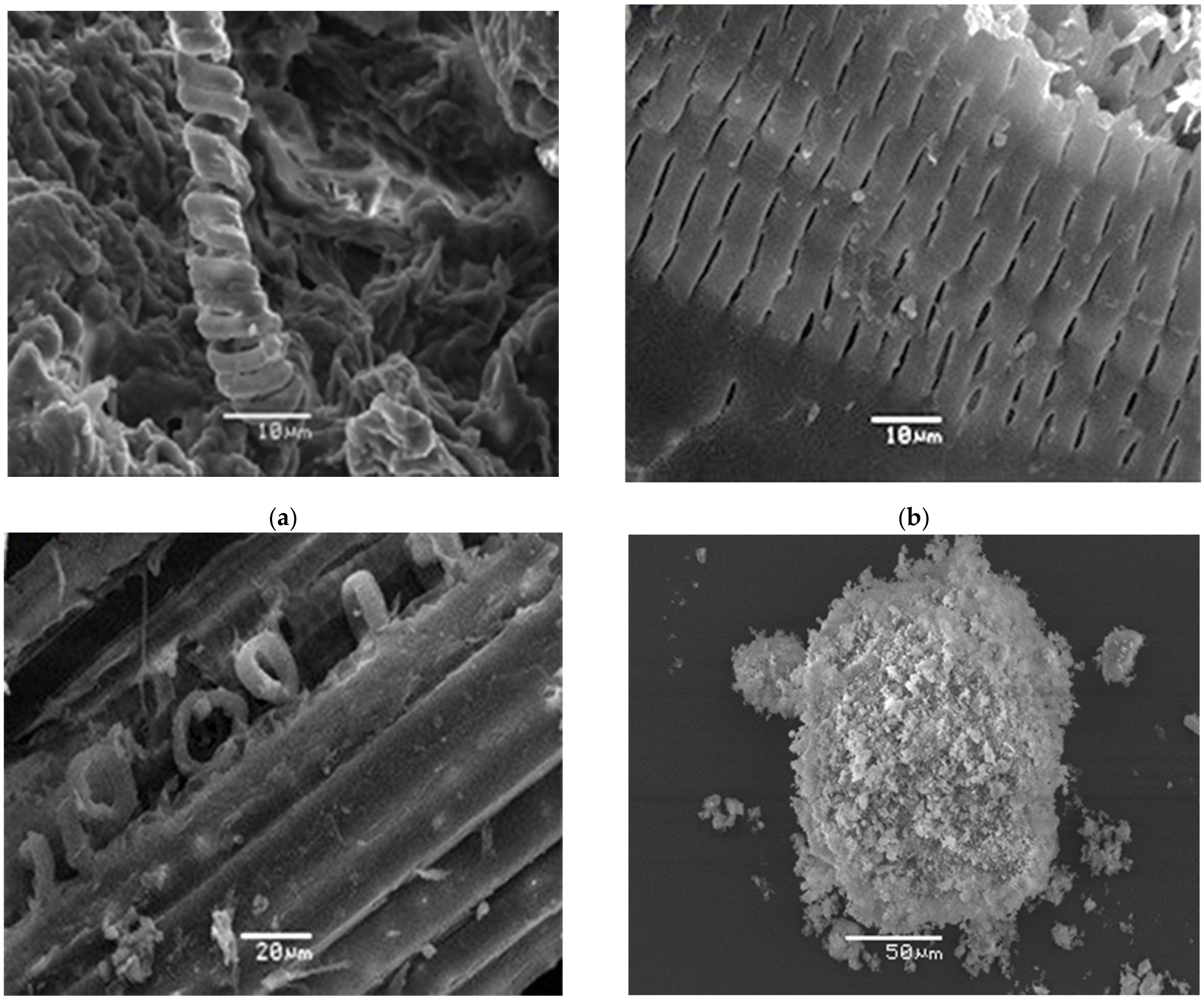
Moisture, ash yields, volatile matter, fixed carbon, and gross calorific values for the biomass and lignite samples studied are presented in Table 2. All biomass samples exhibit higher calorific values (both as received and in dry basis) not only from the studied lignites but also from most of the Greek coals. The lignite deposits in Greece are mainly located in Megalopolis, Elassona, Florina, Ptolemaida, Amynteo, and Drama. Lignites from these locations were classified into three categories based on their calorific value. Megalopoli, Amynteo, and Drama deposits had a lower calorific value, ranging from 900 to 1100 kcal/kg (db), Florina and Elassona deposits had a calorific value between 1800 to 2300 kcal/kg (db), while Ptolemaida deposits—which are the most extensive and commonly exploited today—exhibited a calorific value of 1250–1350 kcal/kg (db) [14]. Despite its lower carbon content, the sawdust sample exhibited the highest calorific values among the biomass samples, probably due to its lower total moisture content (Table 2). It also exhibited lower sulfur and chlorine content, which is expected to result in a decreased level of boiler corrosion and atmospheric emissions during combustion [25].

Table 2. The results of proximate and ultimate analyses and the calorific values of the biomass and lignite samples studied.

	Treatment	Unit	Reed	Sawdust	Olive Kernel	Mavropigi Lignite	Achlada Xylite
Total moisture	Per se [1]	%	19.65	6.51	7.55	56.70	31.72
Ash content	Dry basis	%	3.69	2.34	5.60	9.10	30.40
Ash content	Per se [1]	%	2.96	2.19	5.18	23.92	41.20
Gross Cal. Value	Dry basis	Cal/gr	4340	4525	4440	1650	1621
Low. Cal. Value	Dry basis	Cal/gr	4037	4200	4137	1230	1198
Gross Cal. Value	Per se [1]	Cal/gr	3412	4231	3567	1320	1360
Low. Cal. Value	Per se [1]	Cal/gr	3096	3889	3209	1092	979
Carbon	Dry basis	%	50.07	49.44	53.39	48.10	36.72
Carbon	daf [2] basis	%	51.60	50.55	56.31	63.22	62.45
Hydrogen	Dry basis	%	5.82	6.26	6.08	3.20	2.80
Hydrogen	daf [2] basis	%	6.00	6.40	6.41	4.21	4.76
Nitrous	Dry basis	%	0.31	4.69	1.59	1.72	0.92
Nitrous	daf [2] basis	%	0.32	4.80	1.68	2.26	1.56
Sulphur	Dry basis	%	0.30	0.12	0.17	2.08	1.10
Sulphur	daf [2] basis	%	0.31	0.12	0.18	2.73	1.87
Chlorine	Dry basis	%	0.25	0.10	0.15	nm [3]	nm [3]

[1] As sampled, without any treatment; [2] on a dry, ash-free basis; [3] not measured.

Scanning electron microscopy of biomass samples revealed some characteristic organic structures of original lignocellulosic biomass (Figure 1). Lignocellulose is the non-starchy, fibrous part of plants. Reed samples exhibit porous structures in a specific arrangement and herbal tissue creating windings along some corrugated surfaces. Porous geometry appears elongated and arranged in parallel and dense forms. The variance of size shows that the material is heterogeneous. Sawdust shows porous structures resembling those of reed but with a difference in geometry, as well as in shape. These pores seem to have a linear arrangement, and the pores appear in concentric circular shapes, with the inner circle being void and with defined limits. These areas are very good at absorbing moisture [26] and this can be compared with other high values of moisture up to 45% that have been measured in other samples of sawdust [27]. In the olive kernel sample, lignocellulosic fibers can be observed. These fibers seem to resemble those of reed but much thicker, denser, and longer.



(c) (d)

Figure 1. Scanning electron microscope micro-photographs of the original biomass samples: (**a**) lignocellulosic fibers in olive kernel residues biomass, (**b**) vegetal porous structure in reed biomass, (**c**) fiber structures in reed biomass, (**d**) arcanite spherical agglomerate in reed biomass ash.

In all biomass ashes studied, the initial plant structures are preserved partly after the combustion (Figure 2). Despite the fact that this could be attributed to the relative low temperature of combustion, authors have witnessed similar structures in olive kernel residues' ashes coming from a 5 MW biomass electrical energy plant, combusted in about 950 °C. The results of the XRD (Figure 3) analyses showed that all samples contained an amorphous phase while all biomass ash samples contained calcite and sylvite. According to the SEM study, the amorphous phase in the biomass ashes was mainly constituted of unburned organic residuals, while in lignite ashes, the amorphous phase was principally glass, formed during the co-combustion of the initial mineral compounds and the organic matter of lignites. The existence of calcite in biomass ashes indicated that the biomass was enriched in calcium. Previous authors [26] that have studied forest biomass ashes of different particle sizes and combustion temperatures have also detected calcite in almost every forest biomass ash sample in low to medium density. Calcite was also detected in lignite ashes. Fairchildite ($K_2Ca(CO_3)_2$) has been detected in olive kernel ash while cristobalite, diopside, and gismondite ($CaAl_2Si_2O_8 \cdot 4H_2O$) were found in sawdust ash. Clay minerals, probably of clastic origin, co-precipitated with the organic matter in the basin, were determined only in the coal-ash samples.

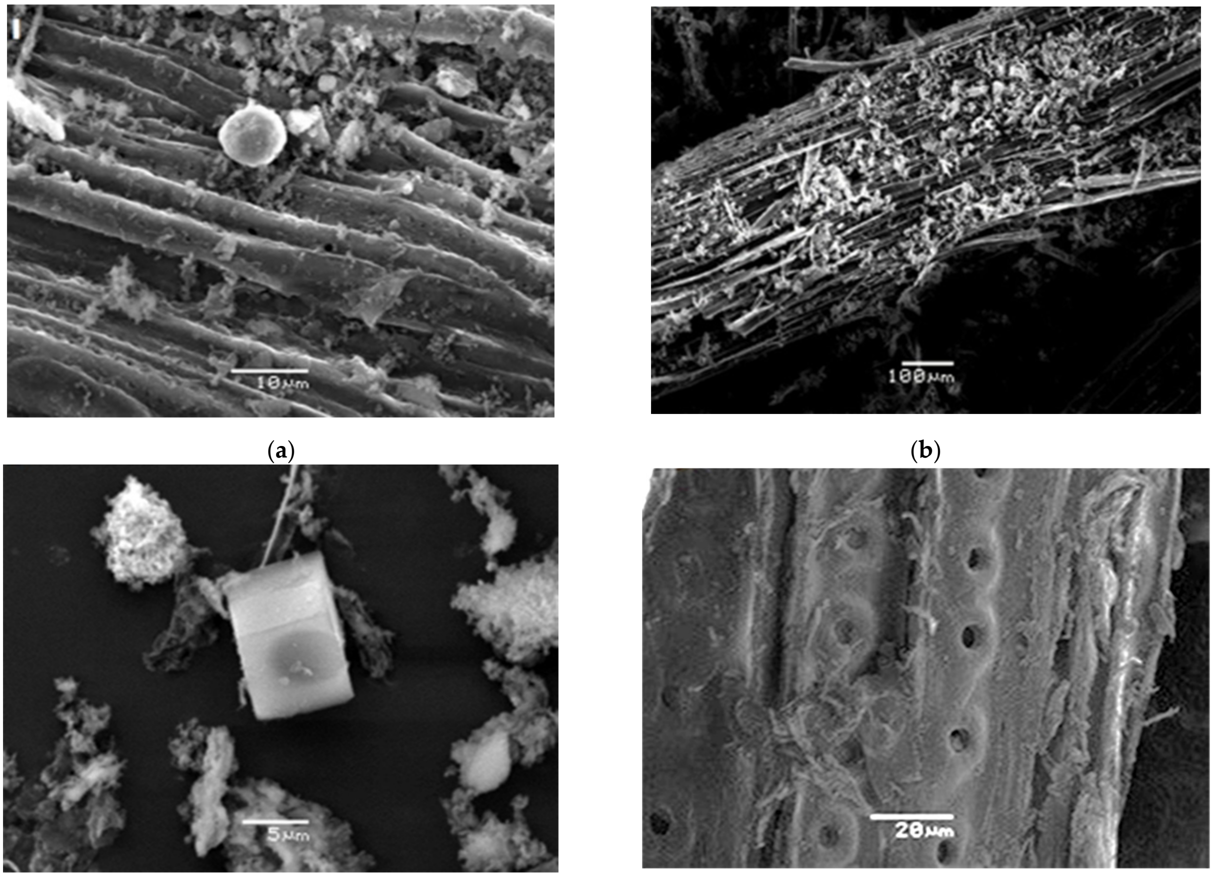


Figure 2. SEM micro-photographs of the biomass samples: (**a**) remaining structure along with a cenosphere in the olive kernel residual biomass ash, (**b**) unburnt porous carbon in the sawdust biomass ash, (**c**) newly formed euhedral calcite crystal in sawdust biomass ash, (**d**) porous structures of sawdust biomass.

The presence of sylvite in biomass samples indicates an abundance of potassium which bonds with chlorine during the combustion of the organic matter. Sylvite was identified by [28] in *Miscanthus* ash, a plant similar to reed. Furthermore, [29] studied various woodchip ash samples through XRD methods, and identified sylvite. Sylvite was also identified in biomass ash samples by [26]. Anhydrite was present in the reed ash sample, indicating an abundance of calcium and sulfur that they bond together due to the combustion. According to [29], anhydrite was found in woodchips ash samples, too. Arcanite (K_2SO_4) was detected in the reed ash and the olive kernel ash sample. According to [29], who examined with XRD methods various ash samples of arboreal biomass (*Pinus halepensis, Pinus brutia, Olea europaea, Cupressus sempervirens, Pistacia lentiscus, Quercus coccifera*), arcanite was identified in the *Olea europaea* ash sample. An amorphous phase is commonly found in coal fly ash in high proportions and is combined with variable amounts of crystalline phases, consisting of the main components of fly ash (e.g., [29–32]). It has also been observed in all types of biomass ash studied. The simultaneous presence of arcanite and sylvite in the reed biomass ash sample and the olive kernel ash sample, along with fairchildite, indicates that in the specific types of biomass, there is an abundance of potassium. It is noted that K is a nutrient element for plants. These differences in the composition of the ashes may lead to different environmental management or potential industrial applications.

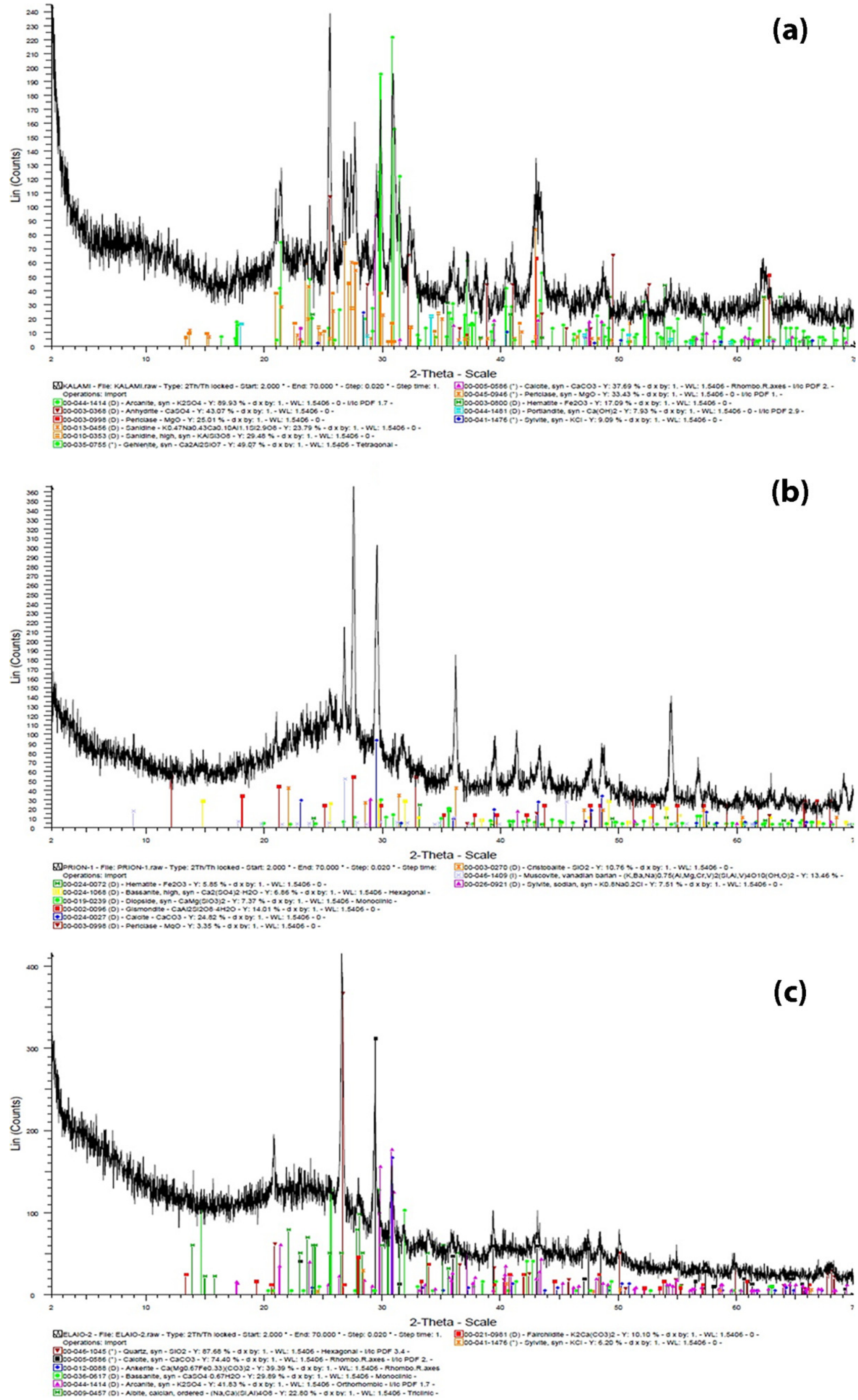


Figure 3. X-ray diffraction patterns of the biomass ash samples: (**a**) reed ash, (**b**) sawdust ash, (**c**) olive kernel residues ash.

4. Conclusions

All biomass samples exhibited higher calorific values not only from the studied lignite, but also from many other Greek coals.

Despite its lower carbon content, sawdust sample exhibited the highest calorific values among the biomass samples, probably due to its lower total moisture content. It also exhibited a lower sulfur and chlorine content.

As far as the ash content, reed, and olive kernel presented higher values than the sawdust.

An amorphous phase was detected in all the ashes of the studied samples. Calcite was detected in all the ashes, both from biomass and coal, but sylvite was detected only in the biomass ashes.

The presence of sylvite in those samples indicates the abundance of potassium in the organic matter that bonds with chlorine and sulfur during combustion. This explains the formation of arcanite that was detected in the reed and the olive kernel ashes. Fairchildite has been detected in olive kernel ash, while cristobalite, diopside, and gismondite were found in sawdust ash.

Clay minerals were determined only in the coal-ash samples.

Further research is needed in order to investigate the reported differences in the composition of the studied ashes in order to define the best environmental management or potential industrial applications for those ashes.

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