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Use of selective grinding for upgrading quality of lignites from Greece and for reducing CO_2 emissions

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ABSTRACT

Lignite plays a dominant role for energy production in Greece, covering over 30% of the demand for electricity generation. Increased energy demand requires, additionally to alternative energy sources, an increase in energy supply from both old and new deposits. Due to the low quality of mined lignite, methods of upgrading in terms of power plants' efficiency and environmental performance need to be applied. In this work, the technique of Selective Size Reduction (SSR) was investigated for three deposits in North Greece, Amynteon, S. Field and Ahlada, in order to improve lignite quality through reduction of its mineral matter content. The variation of the qualitative characteristics of the grain fractions produced by SSR provided useful information on both the suitability of these fractions for combustion in the power plants of the area and the emissions of greenhouse gases. The results have shown that fractions, which were produced from the first stage of crushing, with recovery between 83% and 95%, had lower ash by 16.5–27% than the raw material and were of acceptable quality for combustion in the power plants of the area. Beneficiated fractions could give energy efficiency between 34% and 36% and a reduction in CO_2 emissions up to 29%, originated from the decomposition of the carbonate minerals, contained in the mined lignite. As a result, the cost for buying CO_2 certificates could be lowered up to 3€/MWh.

Introduction

Coal, covering 38% of global energy production, is playing a dominant role in the European Union, with a 25% share of the total installed capacity. Lignite, accounting about 30% of the primary energy consumption and over 40% of power generation, constitutes the major energy resource in Greece (Kavouridis 2008). Mineable coal deposits amount to 4.6 billion tons and are exploited by the Public Power Corporation of Greece. Greece is among the largest lignite producers in the European Union. Over the last 5 years, lignite production was decreased from 54Mt to 38Mt per annum. Despite this decrease, lignite remains a key strategic fuel for Greece, due to secure supply and the controllable cost, which give a competitive strength in Greece's fuel mix. Almost all of the output is burned in power stations, which are all, more or less, mine mouth plants, whereas approximately 1% is used to produce dry lignite and briquettes. The total electric generating capacity is 5289MWe. (Galetakis et al. 2009; Galetakis and Vamvuka 2009;

Public Power Corporation of Greece 2018; The European Association for Coal and Lignite – EURACOAL 2018; Vamvuka and Galetakis 2010).

The competitiveness of the lignite sector in Greece will greatly depend on its ability to keep a constant fuel quality and power plant efficiency, meeting at the same time CO_2 emission limits. The application of the Emission Trading Scheme has greatly affected the power generation sector, by increasing the operating cost of current units, as well as the investment cost for new units (Vamvuka 2009; Vamvuka and Galetakis 2010; Vamvuka, Galetakis, and Roumpos 2013). For the power generation sector in Greece, a cap has been set to 230 Mt of CO_2 (46 Mt of CO_2/year), which accounts for 71% of total certificates (Kavouridis 2008; National allocation plan for CO2 certificates 2006; Vamvuka and Galetakis 2010). The CO_2 emission allowance increases the variable costs of fossil-fuel power plants and thus shortens marginal costs. Lignite-produced electricity is affected much more than gas-produced electricity, because of the higher (approximately double) CO_2 emissions per unit of output (Galetakis and Vamvuka 2009; Kavouridis and Koukouzas 2006).

The emission rate for CO_2 from power plants depends upon both fuel quality and the efficiency of power production. For lignite to keep its primary role in power production, both of these parameters must be improved to meet CO_2 emission limits and keep the cost of electricity low. CO_2 emissions are particularly influenced by the carbonate minerals (e.g., calcite, dolomite, siderite) contained in the mineral matter of Greek lignites. Calcite, the most common carbonate mineral, is decomposed to lime (CaO) and CO_2 during lignite combustion by endothermic reaction, thus reducing the heat available to the boiler. This results in increased CO_2 emissions per unit of energy and, consequently, an additional cost in buying additional CO_2 rights through the ETS (Vamvuka and Galetakis 2010).

To achieve a high output rate with low cost of mined lignite, high capacity bucket wheel excavators, conveyor belts, and spreaders are used. However, the quality of the run of mine lignite is not constant and/or uniform, but exhibits inhomogeneity and large fluctuations. The quality variation of the mined lignite main is closely related to the structure of the deposits and to applied mining techniques. The majority of the Greek lignite deposits are multi-layered, consisting of successive lignite and waste-material seams of varying thickness and chemical composition. The extreme splitting of lignite seams by the waste layers (mainly clays and marls) makes the application of the selective mining necessary. Due to the unavoidable mixing of thin waste layers with lignite, even though selective excavation techniques are applied, the quality of the run of mine lignite is inferior to the quality of the lignite layers and in addition exhibits significant short-term fluctuations. For the compensation of the short-term quality variations, large-scale homogenization-mixing methods are applied (Galetakis and Roumpos 2015).

The worsening quality of the lignite during selective mining increases its ash content and decreases its calorific value. Ash embraces dissolved salts and other inorganic substances present in coal's pore water, inorganic elements within the organic compounds of coal macerals and discrete inorganic particles representing true mineral components. The role of mineral matter on net plant efficiency is crucial (Oman, Senegacnik, and Dejanovic 2001; Ural and Akyildiz 2004). Also, the quality deterioration during mining causes problems not only in lignite handling and impurity removal facilities, but also on industrial processes such as slagging and fouling, increasing maintenance costs. Slagging and fouling deposits reduce heat transfer from the flue gas to the water-steam systems and

can cause blockage of gas flow, erosion, and corrosion with detrimental effects. As a result, the efficiency of the power plants decreases, with a significant negative impact on the CO_2 emissions and the cost of electricity produced (Vamvuka 2009; Vamvuka and Galetakis 2010; Vamvuka, Galetakis, and Roumpos 2013).

Crushing and grinding operations reduce the particle size of coal to a level suitable for feeding the power plants, improving at the same time liberation of minerals. However, the latter depends on the breakage mechanism and consequently the type of force applied. In conventional grinding processes, where high impact and attrition forces are applied, the breakage through grain boundaries is inefficient (Sriramoju, Suresh, and Dash 2019). As the process is highly energy intensive (Tromans 2008), selection of the proper method for particle size reduction required, with low energy consumption and best liberation of impurities, is very important. Previous investigations have studied the grinding properties of minerals in coal using different chemicals (Sahoo, De, and Meikap 2011; Ural and Akyildiz 2004), preheating and microwave treatment (Lytle, Choi, and Prisbrey 1992), or autogenous grinding method applying limited impact force (Sriramoju, Suresh, and Dash 2019). Furthermore, others (Austin, Kalligeris-Skentzos, and Woodburn 1994) have studied the relation between the extent of grinding and degree of liberation of ash, indicating that coal could be beneficiated by ultrafine grinding to below 45 μm, followed by froth flotation. However, generation of fines is energy intensive and creates handling problems.

Based on the above discussion and considering the limited information on the properties of the materials recovered during the grinding operation, present study aimed at investigating the effect of a simple and cheap quality control technique, selective size reduction (SSR), in improving lignite quality from three deposits in Northern Greece, through reduction of its mineral matter content. The qualitative characteristics of the grain fractions produced by SSR were determined, in order to provide information on the suitability of these fractions for combustion in the power plants of the area, the energy efficiency, as well as CO_2 emissions.

Experimental

The samples which were used in this study were delivered from Amynteon, Southern Field (S. Field) and Ahlada mines of Western Macedonia in Greece. Representative samples, after air drying, homogenization, and riffling, were ground by a jaw crusher and a ball mill to a particle size of −250 μm and subjected to qualitative analysis. Proximate and ultimate analyses were carried out according to the ASTM standards (D5142, D5373, D4239) using programmable laboratory furnaces and a CHNS Flash 2000 type analyzer. Gross calorific value (Q_g) was determined by a Leco AC-300 type calorimeter (D5865), while the chemical analysis of ashes by an energy dispersive X-ray fluorescence spectrometer (EDXRF), type Bruker S2 RANGER. Carbon dioxide emitted from carbonate minerals during combustion was determined by an asbestometer type Dietrich-Fruhling.

The methodology of SSR was applied as follows: Raw samples were initially crushed with a jaw crusher and classified by sieving to particle sizes between −100 mm and −0.1 mm (first stage). Consequently, the coarser fraction was crushed and sieved to the same particle sizes as before and a new size distribution was obtained (second stage). The procedure was repeated up to the fourth stage of crushing.

Representative samples from all fractions of all stages of crushing were analyzed using the above techniques, aiming to determine the optimum cut size of the screening, in terms of recovery, ash content, net calorific value as received (Q_n), CO_2 emissions, and energy efficiency. The latter was calculated as (Vamvuka 1996):

$$n = (0.0066xQ_n) + 25.1 \qquad (1)$$

where

$$Q_n = \big[[Q_g - 5.85x9x(100 - A_d)x0.05]x(100 - M)\big]/100 - (5.85xM) \ (kcal/kg) \qquad (2)$$

A_d: ash content (%dry)
M: moisture content (%)

According to (Vamvuka 1996), Equation 1 used for energy efficiency calculation was derived by relating (linear regression) lignite calorific value and efficiency of fed power plants. Data used were from lignite basin of Western Macedonia in Greece, where current samples were taken from. Similarly Equation 2, used for the calculation of the net calorific value Q_n from gross calorific value Q_g, ash content A_d, and moisture M, was especially developed for lignite from the same area.

Results and Discussion

Qualitative characteristics of fractions of selective grinding

The proximate and ultimate analysis results of raw fuels are represented in Table 1. As can be seen, Ahlada lignite had much higher ash content than Amynteon and S. Field lignites, 45% and lower carbon content, resulting in a reduced calorific value. The concentrations of nitrogen and sulfur were low for all fuels, implying low toxic emissions of SO_2 and NO_x during combustion.

From the chemical analysis of ashes in Figure 1 it can be observed that Amynteon and S. Field ashes were very rich in CaO, while Ahlada ash in SiO_2 and to a lesser extent in CaO. Ash from S. Field was also abundant in SiO_2, whereas all ashes contained significant amounts of Fe_2O_3 and Al_2O_3.

Basic criteria, for the successful applicability of the SSR technique as a simple and cheap upgrading method of lignite, were considered the yield of improved samples, the content of ash, the calorific value, and the percentage of CO_2 emitted from inorganic substances during combustion (moisture content as received, although an important parameter too, could not be measured accurately, because the samples were air-dried). Best separation between organic and inorganic parts of the samples occurred during the first stage of crushing for all lignites. The results are presented in Table 2 and Figures 2–4.

The fractions which were characterized as upgraded, in comparison to the raw fuels, were grouped together. The cumulative qualitative characteristics were then determined

Table 1. Proximate analysis, ultimate analysis, and calorific values of the fuels (% dry).

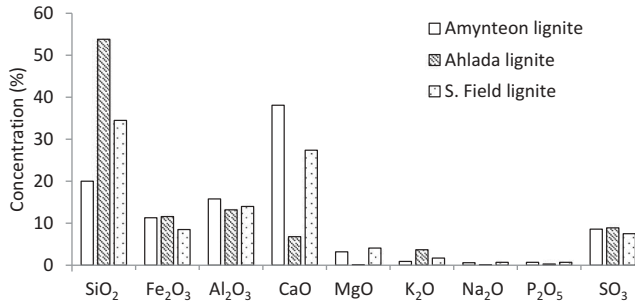| Sample	Volatile matter	Fixed carbon	Ash	C	H	N	O	S	Q_g[a]
Amynteon lignite	46.5	31.5	22.0	38.7	3.4	1.7	33.2	1.3	3073.7
Ahlada lignite	36.4	18.6	45.0	27.3	2.1	0.6	24.2	1.1	2124.8
S.Field lignite	42.66	28.1	29.3	33.4	3.0	1.4	31.8	1.1	3099.5

[a]Gross calorific value (kcal/kg).

Figure 1. Chemical analysis of lignite ashes in major oxides.

Table 2. Qualitative characteristics of lignite fractions produced by selective size reduction.

Particle size (mm)	Cumulative weight (%)			Cumulative ash (%)			Cumulative Q_n (kcal/kg)		
	Amynteon lignite	S. Field lignite	Ahlada lignite	Amynteon lignite	S.Field lignite	Ahlada lignite	Amynteon lignite	S. Field lignite	Ahlada lignite
+100	8.1	20.3	2.2	19.4	15.3	18.2	1622.2	1707.4	1645.0
−100 + 50	23.2	37.9	5.2	19.2	20.2	19.6	1625.3	1659.9	1617.3
−50 + 25	57.5	54.5	18.4	15.7	19.9	20.2	1668.9	1645.0	1605.7
−25 + 16	72.6	62.2	27.0	15.4	21.3	26.3	1676.3	1637.4	1476.6
−16 + 8	88.7	74.0	50.9	17.5	23.3	30.0	1674.0	1621.9	1400.3
−8 + 4	92.6	80.4	70.3	17.9	24.2	32.3	1673.5	1617.9	1359.2
−4 + 2	94.5	86.0	82.5	18.0	24.5	33.0	1673.0	1614.0	1336.0
−2 + 1	95.8	89.9	89.3	18.6	25.8	34.5	1670.4	1606.4	1325.0
−1 + 0.5	96.7	93.1	94.1	19.4	26.4	36.2	1668.1	1597.3	1318.7
−0.5 + 0.3	97.5	95.0	96.5	20.2	26.9	38.7	1665.7	1594.7	1309.8
−0.3 + 0.1	98.8	97.9	99.3	20.9	28.3	40.3	1664.2	1590.6	1306.8
−0.1	100	100	100	21.6	28.8	42.9	1661.0	1586.0	1302.3

for each beneficiated material, in order to investigate whether the methodology produced fractions of specific quality, acceptable for use in the power plants of the area of Western Macedonia. Table 2 shows that the increasing distribution of ash from the coarser toward the finer fractions was raised up to 40% for Amynteon lignite, 88% for S. Field lignite and up to 120% for Ahlada lignite and was most probably related to the more brittle lignite partings. Figure 2 shows that the highest degree of liberation of organic material from minerals occurred for particle sizes of −25 + 16 mm in the case of Amynteon lignite, while +100 mm in the case of S. Field and Ahlada lignites (Figs. 3 and 4 respectively). For Amynteon lignite, the lowest ash content was obtained for −25 + 16 mm fraction, when cumulative weight was 75%. At this size, Qn was the highest, 1676 kcal/kg. In the case of S. Field and Ahlada lignites, the coarsest fraction +100 mm presented the lower ash content 15% and 18%, respectively, which resulted in a Qn 1707 kcal/kg and 1645 kcal/kg. For Amynteon lignite, as an upgraded fraction was selected the −100 + 2 mm, which was recovered by 95%, had 18% ash on a dry basis and a Q_n of 1673 kcal/kg (Fig. 2). The reduction of ash with respect to the initial sample was 18%, whereas the increase in lower heating value was ~1%. For S. Field lignite, as upgraded fraction was also selected the −100 + 2 mm, which was recovered by 86%, had 24.5% ash on a dry basis and a Q_n of 1614 kcal/kg (Fig. 3). In this case, the reduction of ash with respect to the initial sample was 16.4%, whereas the increase in the lower heating value was ~1.9%. On the other hand,
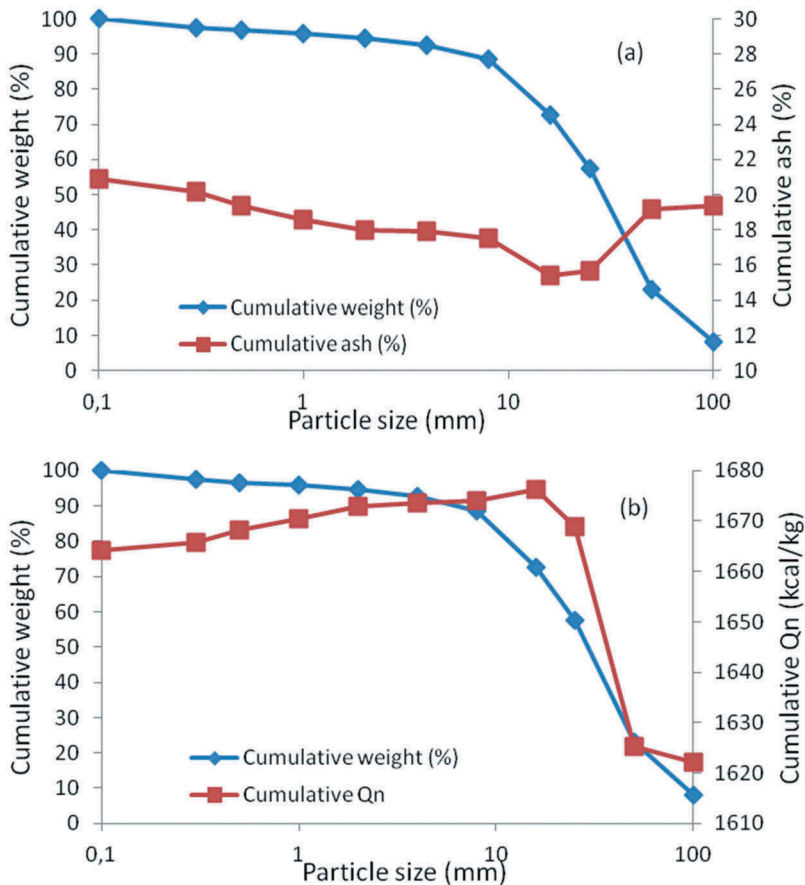
Figure 2. Variation of cumulative weight and cumulative ash (a), cumulative weight and cumulative Q_n (b), as a function of particle size for Amynteon lignite.

for Ahlada lignite, as upgraded fraction was selected the −100 + 2 mm, with a recovery of 83%, ash content on a dry basis 33%, and Q_n of 1336 kcal/kg (Fig. 4). For this fuel, the reduction of ash with respect to the raw sample was much greater (27%), while the increase in lower heating value was 2.8%.

The considerable improvement of ash content and Q_n of the samples obtained from the SSR method is important for industrial units, as ash can cause operational problems (slagging/fouling, corrosion) and environmental pollution (SO_2, toxic heavy metals), while the lower heating value determines the degree of loading of the units, for which a specified power can be achieved. Both these parameters define the efficiency and availability of the facilities and thereby the power cost. The results of Table 2 show that the selected upgraded fractions meet the fuel specifications for the power plants of the area, i.e., ash content 13–55% on a dry basis and lower heating value 1,300–2,030 kcal/kg.

A comparison between the qualitative characteristics of the beneficiated materials and those of the raw fuels is made in Table 3 and Figure 5. As can be seen, for Amynteon lignite the reduction of CO_2 produced from carbonate minerals of the selected fraction was 19.7% and the energy efficiency 36.1%, remaining practically unchanged. For S. Field
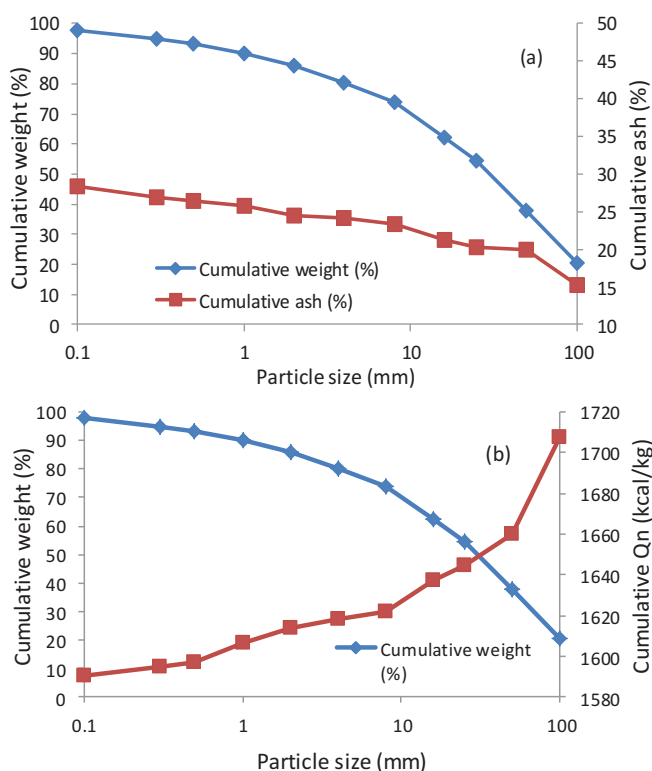
Figure 3. Variation of cumulative weight and cumulative ash (a), cumulative weight and cumulative Q_n (b), as a function of particle size for S. Field lignite.

lignite the corresponding CO_2 reduction was 17.5% and the energy efficiency was improved by 0.84%, attaining a value of 35.8%. For the upgraded fraction of Ahlada lignite, CO_2 emissions were lowered by 29%, while the energy efficiency increased by ~1%, reaching a value of 34%.

Concerning the refuse (fractions −2 mm), which constitutes approximately 5%, 10%, and 17% of the initial samples of Amynteon, S. Field and Ahlada lignites, respectively, with an ash content of 42–52%, as shown in Table 4, it could be worth to investigate its use as additive raw materials of the cement industry, or alternatively its blending with other waste materials for use in soil amendment and amelioration.

Effect on CO₂ Emissions Cost

The decrease of carbonate minerals, and more specifically the decrease of calcite, which is the most abundant carbonate mineral in Greek lignite, in the upgraded fractions of the lignite results in the reduction of CO_2 emissions, as well as, in an increase of the heating value and the power plant efficiency. These factors lead to lower CO_2 emissions (tones of CO_2 per generated MWh) and consequently to lower cost for buying CO_2 rights. The reduction cost resulting from the decrease of CO_2 emissions is given by (Vamvuka and Galetakis 2010):
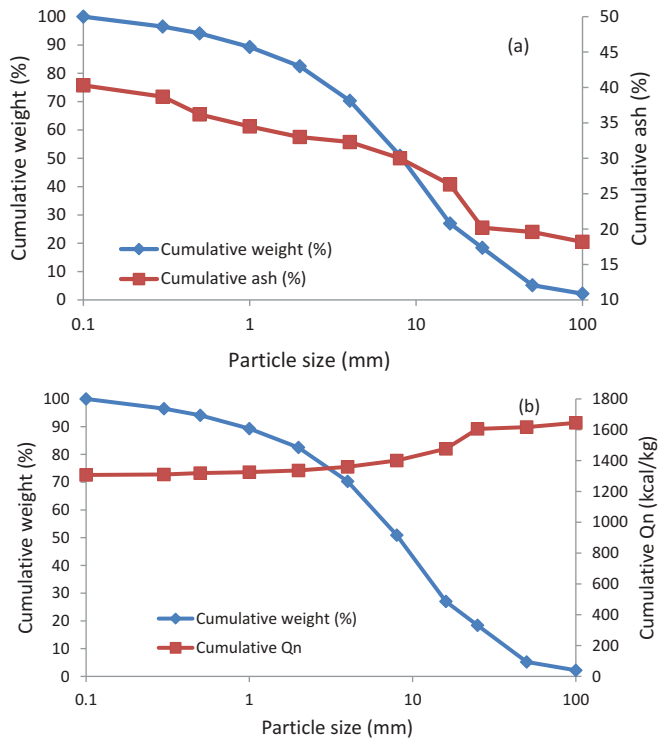
Figure 4. Variation of cumulative weight and cumulative ash (a), cumulative weight and cumulative Q_n (b), as a function of particle size for Ahlada lignite.

Table 3. Qualitative characteristics of lignite upgraded fractions.

Sample	Particle size (mm)	Cumulative ash (%)	Cumulative Q_n (kcal/kg)	Lignite CO_2 (%)	Energy efficiency (%)
Amynteon lignite (raw)	+100-0.1	22.0	1,660.0	6.6	36.0
Amynteon lignite(upgraded)	+100 + 2	18.0	1,673.0	5.3	36.1
S. Field lignite (raw)	+100-0.1	29.3	1,584.0	6.3	35.5
S. Field lignite (upgraded)	+100 + 2	24.5	1,614.0	5.2	35.8
Ahlada lignite (raw)	+100-0.1	45.0	1,300.3	2.4	33.6
Ahlada lignite (upgraded)	+100 + 2	33.0	1,336.0	1.7	33.9

Table 4. Content of ash of refused lignite fractions produced by selective size reduction.

	Cumulative weight (%)			Cumulative ash (%)		
Particle size (mm)	Amynteon lignite	S. Field lignite	Ahlada lignite	Amynteon lignite	S. Field lignite	Ahlada lignite
−2 + 1	23.0	29.1	41.8	35.7	41.6	46.2
−1 + 0.5	40.7	51.7	70.4	37.3	41.8	48.0
−0.5 + 0.3	55.3	64.9	82.2	38.2	42.4	49.1
−0.3 + 0.1	79.5	85.3	96.3	40.3	43.6	50.9
−0.1	100	100	100	41.7	44.2	51.6

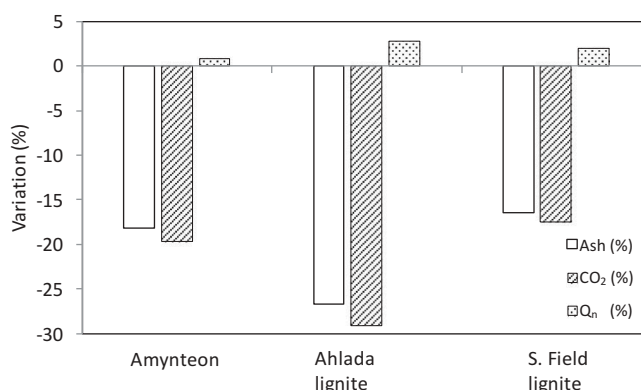$$S_a = \frac{880.45 P \delta C}{n(Q_n - 942.22 \delta C)} \quad (3)$$

Figure 5. Variation of qualitative characteristics of upgraded samples, as a function of those of raw fuels.

where: S_a is the cost reduction in €/MWh

δC is the difference in CO_2 content (%) between the upgraded and the raw lignite

P is the price for CO_2 emissions rights in €/t

Q_n is the net heating value as received of the upgraded lignite in kcal/kg

n = the power plant efficiency (%)

According to (Vamvuka and Galetakis 2010), estimation of S_a was based on the stoichiometric calculation of CO_2 release, due to the decomposition of calcite during lignite combustion and on the decrease of the specific consumption of lignite (t of lignite per generated MWh). The decrease of specific consumption was associated with heating value and power plant efficiency improvement.

Table 5 and Figure 6 show the estimated cost reduction for the upgraded lignites. According to CO_2 market data (www.investing.com/commodities/carbon-emissions-historical-data), the last 3 years the CO_2 certificates have shifted to much higher values, and it is foreseen that this is going to increase further in future. Consequently, the prices of 10, 15, 20, 30, and 40€/t were considered in equation 3. As can be seen, the highest reduction was achieved for Amynteon lignite (0.76 to 3.02€/MWh). For S. Field lignite the corresponding reduction varied from 0.49 to 1.96€/MWh, while for the Ahlada lignite ranged from 0.41 to 1.10€/MWh. Despite the fact that the resulting reduction cost per MWh is quite low, the expected annual savings are considerable as the electricity produced

Table 5. Qualitative characteristics of raw and upgraded lignite and cost reductions for upgraded lignites (three different prices of CO_2 emission rights were considered).

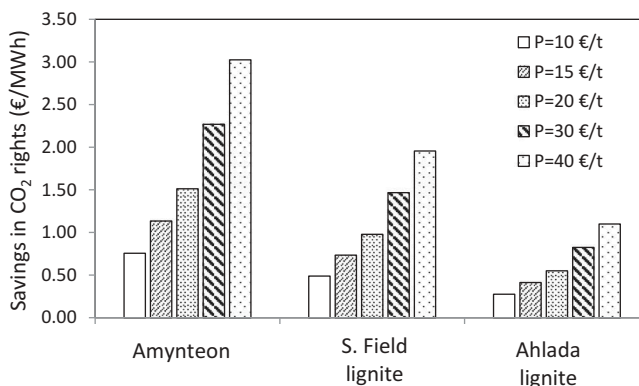|					S_a (€/MWh)				
Sample	Q_n (kcal/kg)	CO_2 (%)	n (%)	δC (%)	P = 10 €/t	P = 15 €/t	P = 20 €/t	P = 30 €/t	P = 40 €/t
Amynteon lignite (raw)	1,660.0	6.6	36.0						
Amynteon lignite (upgraded)	1,673.0	5.3	36.1	1.3	0.76	1.13	1.51	2.27	3.02
S. Field lignite (raw)	1,584.0	6.3	35.5						
S. Field lignite (upgraded)	1,614.0	5.2	35.8	1.1	0.49	0.73	0.98	1.47	1.96
Ahlada lignite (raw)	1,300.3	2.4	33.6						
Ahlada lignite (upgraded)	1336.0	1.7	33.9	0.7	0.27	0.41	0.55	0.82	1.10

Figure 6. Cost reduction per MWh generated from the upgraded lignite for different prices of CO_2 emission rights.

annually in Greece from lignite exceeds 3TWh. Therefore, present results could be valuable for the power companies in order to decide through a feasibility study whether the method of SSR proposed is economically sound.

Conclusion

The application of SSR, a simple method with no use of chemical additives, to low-quality lignites, proved to be successful for the beneficiation of the samples, without any optimization of the process what so ever. Upgraded materials produced from the first stage of crushing presented improved combustion characteristics with respect to raw fuels and were of acceptable quality for combustion in the power plants of Western Macedonia. As upgraded fraction was selected the −100 + 2 mm. For Amynteon lignite, this fraction was recovered by 95% and had 18% ash on a dry basis and a Q_n of 1673kcal/kg. For S. Field lignite, it was recovered by 86%, had 24.5% ash on a dry basis and a Q_n of 1614kcal/kg, while for Ahlada lignite the recovery was 83%, ash content on a dry basis was 33%, and Q_n 1336kcal/kg.Ash reduction varied between 16.5% and 27%, revealing decreasing deposition problems in boilers and less environmental pollution. CO_2 emissions from minerals were lowered up to 29%, whereas the lower heating value of beneficiated samples was improved up to 3%. As a result, the cost of buying CO_2 certificates could be lowered up to 3€/MWh with a concomitant reduction in electricity cost.

SSR could be combined with other applied quality control methods, such as selective mining and homogenization/blending of lignite. In this way, the quality of mined lignite will be higher and more stable, the content of calcite reduced and the cost of associated CO_2 emissions minimized.

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