

Characteristics of Carbonized Biomass Produced in a Manufacturing Process of Wood Charcoal Briquettes Using an Open Hearth Kiln¹

Young Min JU^{2,3} · Hyung Won LEE² · Ah-ran KIM² · Hanseob JEONG² ·
Kwang-Seok CHEA² · Jaejung LEE⁴ · Byoung-Jun AHN² · Soo Min LEE¹^{2,†}

ABSTRACT

Characteristics of carbonized biomass obtained from a Wood charcoal briquette manufacturing process using an open hearth kiln are analyzed in this research, and differences in the characteristics based on the results of a mechanical screening process and the position within the kiln. One type of biomass and five types of carbonized biomass were collected from a Wood charcoal briquette manufacturer. After screening and grinding processes were performed on samples of 1 type of biomass and 5 types of carbonized biomass extracted from a Wood charcoal briquettes manufacturer to classify by particle size, fixed carbon, ash, volatile matters, elemental composition, and high heating value (HHV) were measured. Experimental results showed that the carbonized biomass collected from the middle layer had the highest HHV, 20.4 MJ/kg, and therefore had the highest fuel quality. In terms of particle size, the carbonized biomass below 100 mesh had the lowest ash content and the highest HHV, carbon content, and fixed carbon content. Correlation analyses showed that ash content had negative correlations with HHV, volatile matters, fixed carbon, and carbon content, which suggested that ash content affected negatively on fuel quality.

Keywords: wood charcoal briquettes, open-hearth carbonization furnace, fuel characteristic

1. INTRODUCTION

Timber products refer to 15 different types of products prescribed by the Presidential Decree of the Act on the Sustainable Use of Timbers (the Timber Use Act) (Korea forest service, 2018). wood charcoal briquette is one type of timber product, and the criteria for standard dimensions and quality are defined in Annex

14 of the National Institute of Forest Science Public Notification 2018-8 “Criteria for Standard Dimensions and Quality of Timber Products” (National institute of forest science, 2018). The wood charcoal briquette defined in the Public Notification encompasses products that are shaped into certain forms after being carbonized from raw materials such as wood, woodchips from bamboo or seeds and nuts, wood shavings, or bark,

¹ Date Received November 25, 2019, Date Accepted February 28, 2020

² Division of Wood Chemistry, Department of forest Products, National Institute of Forest Science, Seoul 02455, Republic of Korea

³ Department of Mechanical Engineering, Korea University, Seoul 02841, Republic of Korea

⁴ Division of Research Planning and Coordination, National Institute of Forest Science, Seoul 02455, Republic of Korea

[†] Corresponding author : Soo Min LEE (e-mail: lesoomin@korea.kr, ORCID: 0000-0002-4214-5094)

as well as products that are shaped into forms from carbonized sawdust using additives. The wood charcoal briquette is categorized into sawdust charcoal briquettes, charcoal powder briquettes, and molded charcoal for kidling holed coal briquette, depending on the manufacturing methods. While quality criteria for ash content, fixed carbon, and high heating value are different depending on the type of wood charcoal briquette, the same criteria for heavy metal content apply to all products in this category, except for barium, which is used as a complexing agent (National Institute of Forest Science, 2017). In addition, the use of construction wood wastes, residential wood wastes, wood treated with pesticides, wood treated with preservatives, and other materials including wood, bamboo, seeds and nuts, sawdust, shavings, and barks that are suspected of contamination with harmful compounds are restricted (National Institute of Forest Science, 2017).

When it comes to logging in the Republic of Korea, cutting volume is estimated to be 7,670,000 m³ in 2016, and products collected as a result are estimated to be 5,250,000 m³, therefore leaving 2,420,000 m³ of unused forest biomass (Korea forest service, 2017). Thus, it is imperative that the unused forest biomass that makes up 32% of the total cutting volume utilized in various wood industries.

Domestic manufacturers of wood charcoal briquette have pursued diversification of the raw materials based on implementation of the Timber Use Act and National Institute of Forest Science Public Notification, and wood charcoal is currently produced primarily by using branches and roots of trees from the unused forest biomass (Korea Forest Service, 2018; Choi, 2018). Also, there had been an increased number of cases where the heavy metal content of wood charcoal briquette products is found to exceed the criteria. Current situations can be viewed as a progress towards standardization of wood charcoal briquette products, as integrated quality criteria for timber products are established and

managed; however, currently no consensus exists among industry, academia, research institutions, the authorities, and private corporations (Ahn *et al.*, 2013; Kwon *et al.*, 2018; Yang and Han, 2018).

Most of the charcoal briquettes in the Republic of Korea are manufactured using an open hearth kiln, where a pit in the ground is prepared as a carbonization chamber, and raw materials are stacked in the chamber, carbonized at 300–400 °C, and extinguished using water or soil (Yang *et al.*, 2017; Magnone *et al.*, 2019). Afterwards, a new batch of raw materials is added on top of the carbonized materials, carbonized, and extinguished and this process is repeated several times. Once the carbonized materials reach the top of the carbonization chamber, they are removed from the chamber and used for manufacturing wood charcoal briquettes. Carbonization time varies within the materials produced by this method in that the lowermost layer undergoes carbonization for the longest time and the uppermost layer for the shortest time; also, impurities such as soil are concentrated in certain layers during the process.

For this, the research aims to evaluate the quality of carbonized materials produced by carbonization of unused forest biomass in an open hearth kiln, which is used for manufacturing wood charcoal briquettes. Carbonized materials are collected based on the time in which they are added to the carbonization chamber, and they are mechanically processed and sorted by particle sizes. Their fuel characteristics before and after carbonization are compared based on factors including time of addition, carbonization time, and particle sizes.

2. MATERIALS and METHODS

2.1. Testing Materials

Materials produced in an open hearth kiln used by a wood charcoal briquette manufacturer located in

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Fig. 1. Hog (HSW) and carbonized hog (CSW) of soft wood.

Chilgok-gun, Gyeongsangbuk-do, Republic of Korea, were utilized in this research. Testing materials were the byproducts of afforestation efforts, purchased by the wood charcoal briquette manufacturer, which were composed of mixed softwoods. The raw materials were carbonized at approximately 700 °C and extinguished for approximately 80 hours. Hog of softwood (HSW) and 5 types of carbonized hog of softwood (CSW) are used in this research (Fig. 1). The carbonized hog materials were divided into 5 equal layers based on their vertical position within the kiln during carbonization, labeled as CSW1, CSW2, CSW3, CSW4, and CSW5 from the uppermost layer to the lowermost layer (Fig. 2). Samples were collected from each layer for testing, and the collected samples were double sealed and stored at room temperature for a better preservation.

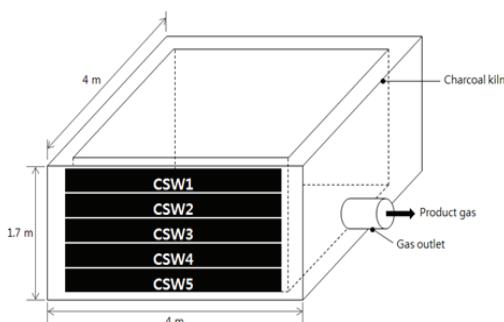


Fig. 2. Schematic view of charcoal kiln and sampling by location.

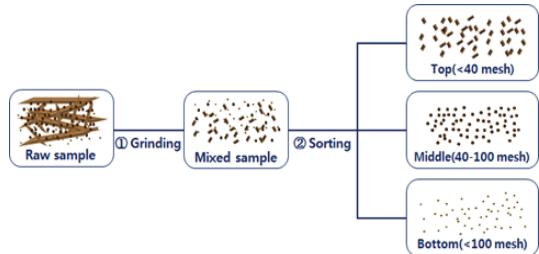


Fig. 3. Process description of mechanical pretreatments.

2.2. Mechanical screening

Carbonized materials obtained from each layer were ground and sorted based on the particle sizes to analyze the distribution of their particle sizes, as shown in Fig. 3. All samples were dried for approximately 48 hours in an oven at 65 °C prior to use to facilitate the subsequent screening process. A cutting mill (Pulverisette 19, FRITSCH, Germany) was used for the grinding process (Fig. 3-①). The cutting mill was operated using a rotational speed of 300 rpm, and a sieve with 1 mm in diameter was used. Particle size sorting was performed using a vibratory sieve shaker (analysette 3, FRITSCH, Germany), with standard sieves of 40 mesh (0.4 mm) and 100 mesh (0.149). The materials were separated into 3 categories: particles with sizes above 40 mesh, between 40 and 100 mesh, and below 100 mesh, labeled as Top, Middle, and Bottom, respectively. The yield was calculated for each category at this stage using Equation (1) in order to analyze the distribution of particle sizes.

$$Y_P (\%) = \frac{W_P}{W_S} \times 100 \quad (1)$$

Y_P =Yield of the corresponding particle size (%)

W_P =Weight of the corresponding particle size (g)

W_S =Weight of the original sample (g)

2.3. Proximate analysis

Proximate analysis was conducted using a thermogravimetric analyzer (PrepASH229, Precisa, Switzerland)

to analyze moisture content, volatile matter, ash, and fixed carbon contents of the samples. Approximately 1 g of material was used for the analysis. Analytical conditions were determined based on ISO 18122 Solid biofuels — Determination of ash content, ISO 18123 Solid biofuels — Determination of the content of volatile matter, and ISO 18134-3 Solid biofuels — Determination of moisture content — Oven dry method, published by the International Organization for Standardization (ISO). The fixed carbon content was calculated using Equation (2).

$$FC (\%) = 100 - (MC + VM + ASH) \quad (2)$$

FC: Fixed carbon content (%)
 MC: Moisture content ratio (%)
 VM: Volatile matter content (%)
 ASH: Ash content (%)

2.4. Measurement of high heating value (HHV)

HHVs were measured using a calorimeter (Parr 6400, Parr, USA), referring to ISO/DIS 18125 Solid biofuels — Determination of calorific value. Samples for analysis were prepared by accurately weighing 0.5 g of the powder and forming a pellet with a diameter of 13 mm under 10 tons of pressure, and they were completely dried in an oven at 105 ± 3 °C for more than 3 hours prior to measurement.

2.5. Elemental analysis

Elemental composition was analyzed using an elemental analyzer (vario MICRO cube elemental analyzer, elementar, Germany). Carbon, hydrogen, nitrogen, and sulfur contents were measured, and oxygen content was calculated according to Equation (3). Analysis conditions were as follows: samples underwent a complete combustion in the combustion chamber that was maintained at the temperature of 1150 ± 5 °C, and the gas generated from this process was purged using helium

flowing at 600 mL/min to the reduction tube that was maintained at 850 ± 5 °C, where they were reduced to N₂, CO₂, H₂O, and SO₂ for analysis. 50 mg of the powder was used for the analysis, and Standard Reference Material® 2693 (National Institute of Standards and Technology, USA) was used as a reference. Samples were completely dried in an oven maintained at 105 ± 3 °C for more than 3 hours prior to elemental analysis.

$$O (\%) = 100 - (C + H + N + S) \quad (3)$$

C: Carbon content (%)
 H: Hydrogen content (%)
 N: Nitrogen content (%)
 S: Sulfur content (%)
 O: Oxygen content (%)

2.6. Calculation of energy yield

The energy yield (Y_E) of the samples obtained from the screening process was calculated using Equation (4). The equation is calculated based on dry weight.

$$Y_E (\%) = \frac{HHV_p}{HHV_s} \times Y_p \quad (4)$$

Y_E = Energy yield (%)
 HHV_p = high heating value for the corresponding particle size (MJ/kg)
 HHV_s = high heating value of the original sample (MJ/kg)

3. RESULTS and DISCUSSION

3.1. Distribution of particle sizes

Weight-based yields of the samples corresponding to each particle size which are categorized by the mechanical screening process, are shown in Fig. 4. The yields of Top (>40 mesh), Middle (40–100 mesh), and Bottom (<100 mesh) samples are distributed with the ranges of 43.6%, 30.6%, and 25.9%, respectively, for

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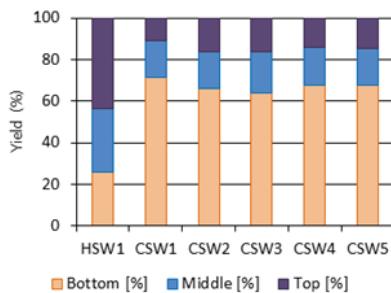


Fig. 4. Particle distribution of the wood charcoal briquettes.

the HSW sample, which is not carbonized. The yields of Top, Middle, and Bottom are in the ranges of 10.7–16.6%, 17.6–20.0%, and 63.8–71.2%, respectively, for the carbonized samples, CSW1 through CSW5. Previous studies have reported that carbonization of wood leads to thermal degradation of wood components, including hemicelluloses, celluloses, and some lignins, which causes an increase in carbon content and brittleness of wood (Lee *et al.*, 2015; Mei *et al.*, 2015). Therefore, it is understood that carbonized samples generate a greater amount of fine particles below 100 mesh when

mechanically processed than the uncarbonized sample due to the greater grindability. While the carbonized samples show slightly different yields depending on their vertical position within the open hearth kiln during the carbonization process, the differences are rather not significant, and they indicated a relatively similar tendency of particles distribution.

3.2. Measurement of high heating value, proximate analysis, and elemental analysis

Results of HHV measurements, proximate analysis, and elemental analysis are summarized in Table 1 for the uncarbonized HSW1 and 5 carbonized CSW samples. Samples from CSW1 through CSW5 exhibit lower volatile matter contents than the HSW by 53.5–57.7%. The significantly lower volatile matter contents of the carbonized samples are thought to be caused by the removal of most of the volatile compounds as gas during the high-temperature carbonization process. On the other hand, ash and fixed carbon contents of the carbonized samples are 20.7–34.0% and 21.1–33.1%,

Table 1. Proximate analysis, elemental analysis, and High heating value (HHV) of the hog and carbonized hog of soft woods

Sample	Proximate analysis (wt.%, dry basis)			Elemental analysis (wt.%, dry basis)					HHV (MJ/kg, dry basis)
	Volatile matter	Ash	Fixed carbon ¹⁾	C	H	N	O ²⁾	S	
HSW	68.4 (1.1) ³⁾	14.0 (1.3)	17.6 (0.2)	39.9 (0.4)	5.0 (0.4)	0.4 (0.1)	54.8 (1.8)	0.0 (0.0)	16.1 (0.2)
CSW1	14.2 (0.2)	39.1 (1.5)	46.7 (1.3)	50.5 (0.0)	1.8 (0.0)	0.9 (0.0)	46.7 (0.9)	0.1 (0.0)	19.3 (0.2)
CSW2	10.7 (0.3)	42.6 (3.1)	46.8 (2.8)	49.9 (0.0)	1.4 (0.0)	0.7 (0.0)	47.9 (2.1)	0.0 (0.0)	19.4 (0.5)
CSW3	14.7 (0.4)	34.7 (4.8)	50.7 (5.2)	50.9 (0.0)	1.7 (0.0)	0.7 (0.0)	46.6 (0.5)	0.0 (0.0)	20.4 (0.1)
CSW4	13.3 (2.3)	48.0 (10.9)	38.7 (8.6)	42.9 (0.1)	1.7 (0.1)	0.7 (0.0)	54.7 (0.3)	0.0 (0.0)	16.7 (0.1)
CSW5	14.7 (0.7)	37.7 (3.3)	47.6 (2.6)	47.1 (0.0)	1.8 (0.0)	0.8 (0.0)	50.2 (0.5)	0.0 (0.0)	18.6 (0.3)

1) Fixed carbon (%)=100-(Volatile matter+Ash) (%)

2) O (%)=100-(C+H+N+S) (%)

3) The numbers in parenthesis refer to standard deviations.

respectively, which are higher than those of the uncarbonized sample (14.0% and 17.6%, respectively). A previous study has reported that mineral components within wood become concentrated in wood charcoal by 4–5 times during a carbonization process and remain as ash (Lee and Kim, 2010). It is thought that the higher ash content of the carbonized samples than that of the uncarbonized samples in this study can be attributed to similar reasons. The carbon content of HSW samples is measured to be 39.9%, and those of CSW1 through CSW5 range from 42.9% to 50.9%, which show a small increase. Lee *et al.* (2007) has reported that the carbon contents of *Pinus Koraiensis* carbonized at 300–400 °C range from 67.7% to 72.2%, which are higher than the carbon contents proposed in this study. We hypothesize that this difference, as well as the relatively high ash contents mentioned above, has been caused by a greater amount of soil and other impurities contained in the samples used in this study, which might have been mixed in when materials were collected from the ground after trees had been cut, during transport and storage of the collected materials, or during the carbonization process in the open hearth kiln. On the other hand, the carbonized samples had lower hydrogen and oxygen contents in most cases than the uncarbonized samples, which is thought to be caused by removal of hydrogen and oxygen in the form of steam and carbon

dioxide, respectively, during carbonization. Nitrogen and sulfur contents are relatively small and do not exhibit significant changes. HHVs on average have increased after carbonization. Samples from the middle layer, CSW3, have the largest average HHV, 20.4 MJ/kg. Also, the bottom two layers (CSW4 and CSW5) exhibit smaller HHVs than the top two layers (CSW1 and CSW2) by approximately 1.7 MJ/kg. We conclude that HHVs of the carbonized materials produced in the open hearth kiln are not strongly affected by duration of carbonization but by blocked oxygen influx during the carbonization process.

Results of proximate analysis, elemental analysis, and HHV measurement for the carbonized samples (CSW) are summarized in Table 2 based on the particle sizes. Volatile matter and fixed carbon content are measured to be increased with decrease in particle size: these values for the Bottom particles, 15.5% and 52.3%, respectively, are approximately twice as large as the corresponding values for the Top particles, 7.4% and 26.0%, respectively. Ash content is observed to be decreased with decrease in particle size from 66.6% to 32.2%. It is thought that most of the impurities, such as soil, contained in the carbonized samples have sizes greater than 40 mesh, and as a result, they are concentrated on the Top layers during the grinding and sorting process. Carbon content exhibits a similar trend

Table 2. Proximate analysis, elemental analysis and High heating value(HHV) for carbonized hog of soft woods (Top, Middle, and Bottom)

Sample	Proximate analysis (wt.%, dry basis)				Elemental analysis (wt.%, dry basis)				HHV (MJ/kg, dry basis)
	Volatile matter	Ash	Fixed carbon ¹⁾	C	H	N	O ²⁾	S	
Top	7.4 (1.0) ³⁾	66.6 (7.1)	26.0 (6.4)	16.4 (5.6)	0.7 (0.1)	0.3 (0.1)	82.6 (5.7)	0.0 (0.0)	10.4 (2.4)
Middle	11.0 (1.3)	49.5 (4.4)	39.5 (3.9)	33.6 (7.4)	1.2 (0.1)	0.5 (0.1)	64.6 (7.6)	0.0 (0.0)	14.8 (2.3)
Bottom	15.5 (1.8)	32.2 (4.9)	52.3 (4.6)	58.8 (3.4)	2.0 (0.2)	0.9 (0.1)	38.2 (3.5)	0.0 (0.0)	21.3 (1.3)

1) Fixed carbon (%)=100-(Volatile matter+Ash) (%)

2) O (%)=100-(C+H+N+S) (%)

3) The numbers in parenthesis refer to standard deviations.

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with the fixed carbon content, which increases with decreasing particle sizes; the Bottom particles are found to have the highest carbon content, 58.8%. On the other hand, oxygen content decreases with decrease in particle size, with a minimum of 38.2% exhibited by the Bottom particles. Hydrogen and nitrogen contents show a small increase as particle size decreases, and sulfur content does not show any significant changes. The Bottom particles also have the highest HHV, 21.3 MJ/kg, which is above the quality criteria for wood charcoal briquettes, 17.6 MJ/kg, prescribed in Annex 14 of the National Institute of Forest Sciences Public Notification 2018-8 “Criteria for Standard Dimensions and Quality for Timber Products.” A previous study has also reported that materials with smaller particle sizes exhibited a higher carbon content and high heating value when carbonized (Hwang *et al.*, 2012), and that energy in the carbonized samples was concentrated in the Bottom particles (Hwang *et al.*, 2012). Considering these results, it is concluded that selecting carbonized materials with smaller particle sizes in grinding and sorting processes will increase the quality of wood charcoal briquette, and this pre-processing is expected to be more effective for materials containing a large amount of impurities such as soil.

3.3. Energy yield

Results of energy yield calculations based on HHVs of each sample are shown in Table 3. The energy yield

in this study considers both weight-based yield from the mechanical screening process and the measured HHVs, and it represents the ratio of the final energy that can be recovered through the screening process to the total energy contained in the original sample. The energy yields of the uncarbonized samples, HSW, are measured to be 0.46, 0.31, and 0.21 for the Top, Middle, and Bottom particles, respectively. The decreasing trend in energy yield with decreasing particle size is thought to be attributable to low grindability of the HSW samples, resulting in the lower yield of smaller particles, as shown in Fig. 4. On the other hand, the energy yields of the carbonized samples are measured to be 0.05–0.12, 0.12–0.17, and 0.70–0.82 for the Top, Middle, and Bottom particles, respectively, which indicate that the energy yields dramatically increase with decreasing particle size. It is understood that the carbonized samples generate more smaller particles which have larger HHVs, thus resulting in significant differences in energy yield in which these two factors are multiplied. The highest energy yield is observed in CSW1 Bottom, 0.82; the highest HHV is observed in CSW3 Bottom, 20.67 MJ/kg, while its energy yield (0.71) is on the lower side due to its relatively low yield. This is considered to be due to having a relatively lower weight-based yield when compared to its high HHV. While the total HHVs of HSW and CSW4 are similar, making it difficult to believe that the carbonization was successful, mechanical screening helps confirm successful

Table 3. high heating values (HHV) and energy yields (EY) for the hog and carbonized hog of soft woods

Particle size	HSW†		CSW1		CSW2		CSW3		CSW4		CSW5	
	HHV	EY	HHV	EY	HHV	EY	HHV	EY	HHV	EY	HHV	EY
Total	16.11 ¹⁾ (0.15)	1.00	19.27 (0.21)	1.00	19.40 (0.54)	1.00	20.41 (0.11)	1.00	16.73 (0.12)	1.00	18.60 (0.32)	1.00
Top	16.84 (0.68)	0.46	9.73 (0.58)	0.05	13.51 (0.77)	0.12	10.83 (0.69)	0.09	6.81 (0.41)	0.06	10.99 (0.66)	0.09
Middle	16.40 (0.33)	0.31	15.98 (0.55)	0.15	14.31 (0.72)	0.13	16.90 (0.29)	0.17	11.08 (0.15)	0.12	15.53 (0.28)	0.15
Bottom	13.12 (0.19)	0.21	22.24 (0.30)	0.82	20.67 (0.14)	0.70	22.81 (0.19)	0.71	19.65 (0.03)	0.79	20.89 (0.06)	0.76

1) The numbers in parenthesis refer to standard deviations.

carbonization of the CSW4 layer, shown by a large increase in HHV of the Bottom particles from CSW4 compared to those from the uncarbonized samples.

3.4. Correlation analysis

Correlations among HHV, volatile matter, ash content, fixed carbon, and carbon content of the Top, Middle,

and Bottom particles of the carbonized samples CSW1 through CSW5 are presented in Fig. 5. Ash content in the carbonized samples is statistically highly correlated with the fixed carbon content, exhibiting the largest value of $R^2=0.9886$; this is thought to be related to the clearly decreasing trend in ash content with decreasing particle size observed in this research. HHVs

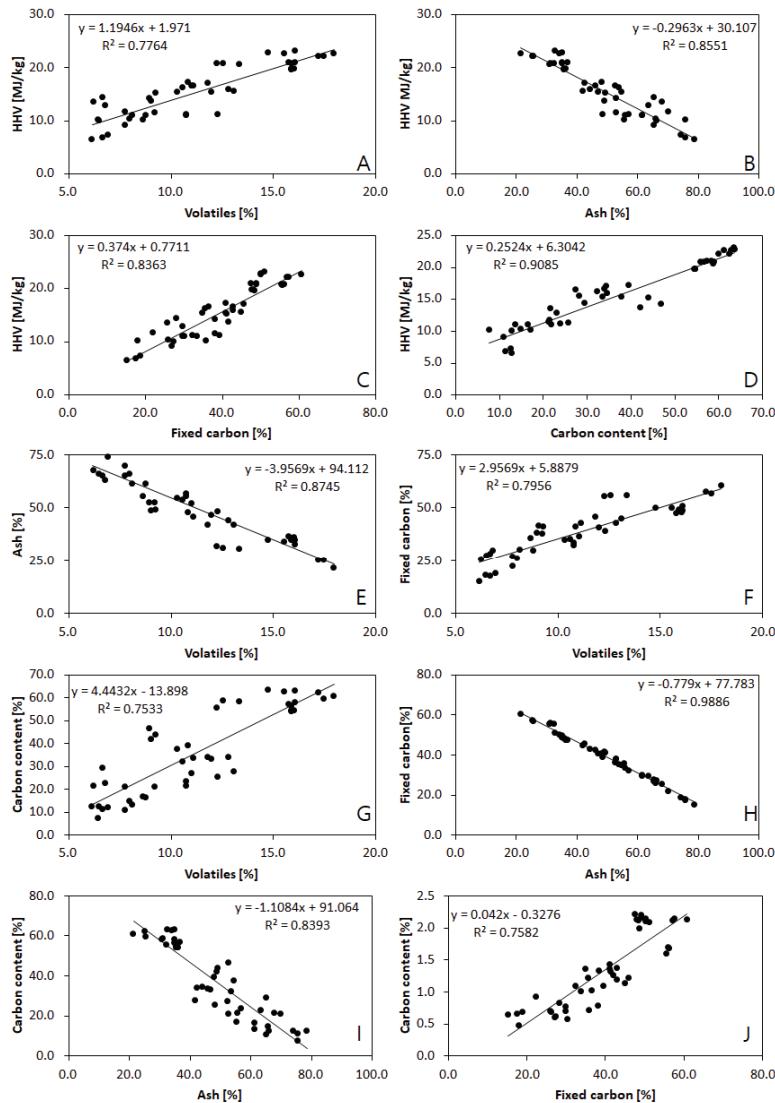


Fig. 5. Relationship between ash, volatile matter, fixed carbon, carbon content and HHV of carbonized hog of soft wood

of the carbonized samples are highly correlated with the carbon content ($R^2=0.9085$). This is thought to be caused by enrichment of C-C bonds, which have high bond energy, within the carbonized samples as the hydrogen and oxygen are removed during carbonization (Lee *et al.*, 2016). In addition, HHVs exhibit a statistically significant positive correlation with fixed carbon content ($R^2=0.8363$). HHVs of the carbonized samples with different particle sizes have a statistically significant negative correlation with ash content ($R^2=0.8551$). This is thought to be related to the trend that ash content of the carbonized samples decreases as particle size decreases, while HHV increases, as shown in Table 2. The correlation between HHVs of the carbonized samples and volatile matters exhibits a scattered distribution and is therefore statistically insignificant. Ash, volatile matter, and carbon content have statistically significant negative correlations ($R^2>0.83$). Volatile matters and fixed carbon have a positive correlation ($R^2=0.7956$); likewise, carbon content, volatile matter, and fixed carbon have statistically significant positive correlations ($R^2>0.75$).

4. CONCLUSIONS

In this study, characteristics of carbonized samples produced using a wood charcoal briquette manufacturing process based on an open hearth kiln are analyzed. For the carbonized samples collected based on their vertical position within the kiln, it was originally expected that the bottommost layer would exhibit the highest quality, but the experimental results suggest that the quality decreases in the order of middle layer, top layers, and bottom layers. Fuel quality of the carbonized materials produced using the open hearth kiln is observed to be not related to carbonization time; the middle layer exhibits the highest quality due to a relatively uniform carbonization reaction arising from heat transfer from both directions, i.e., from the layers above and below.

When characteristics of the carbonized samples are analyzed based on particle size, HHV, carbon content, and fixed carbon content increase as the particle size decreases. In particular, the carbonized samples below 100 mesh exhibit an HHV, carbon content, and fixed carbon content of 21.3 MJ/kg, 58.8%, and 52.3%, respectively, which are directly related to an increase in fuel quality. A relatively large increase in quality can be expected if carbonized samples below 100 mesh are selected after carbonization and grinding, in a case where screening is not feasible prior to carbonization in existing processes.

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APPENDIX

(Korean Version)

평로탄화로를 이용한 성형목탄 제조공정에서 생산된 탄화 바이오매스의 특성

초록 : 본 연구는 평로탄화로 이용한 성형목탄 제조과정에서 얻은 탄화된 바이오매스의 특성을 분석하였으며, 기계적 전처리 및 평로탄화로 내 위치에 따른 특성 차이를 비교하고자 하였다. 성형목탄 제조업체에서 채취된 바이오매스 1종과 탄화 바이오매스 5종의 시료를 대상으로 선별(screening) 및 분쇄(grinding)를 통해 분석시료의 입자크기 범위별로 분류한 후, 고정탄소, 회분, 휘발성 화합물, 원소 함량, 발열량을 측정하였다. 실험 결과, 평로탄화로의 위치에 따라서는 중간층의 탄화 바이오매스 발열량이 20.4 MJ/kg으로 가장 높은 연료적 특성을 나타내었다. 선별 입자 크기에 따라서는 100 mesh 이하의 탄화 바이오매스에서 회분함량이 가장 낮았고 발열량, 탄소 함량, 고정탄소 함량은 높았다. 상관관계 분석 결과 회분 함량은 발열량, 휘발성 화합물, 고정탄소, 탄소 함량과 모두 음의 상관관계를 나타내어 회분 함량이 연료적 특성에 부정적인 영향을 미치는 것을 확인하였다.

1. 서 론

목재제품이란 목재의 지속가능한 이용에 관한 법률(목재이용법)에서 대통령령으로 지정한 15개 제품을 말한다(Korea forest service, 2018). 성형목탄은 목재제품의 한 종류로, 국립산림과학원 고시 제2018-8호 목재제품의 규격과 품질기준의 부속서 14에서 규격과 품질기준이 정의되었다(National institute of forest science, 2018). 해당 고시에서 정의된 성형목탄은 목재, 대나무나 종실의 삽면, 대팻밥, 수피 등을 원료로 하여 일정한 형태로 성형 후 탄화한 것과, 탄화한 톱밥숯에 결합제 등 보조첨가제를 넣고 성형한 것을 말하며, 제작 방식에 따라 톱밥성형탄, 숯가루성형탄 및 구멍탄착화용 성형탄으로 구분된다. 성형목탄의 종류에 따라 회분함량, 고정탄소, 고위발열량 등 품질기준에 차이가 존재하지만, 착화제로 사용되는 바륨을 제외한 중금속 함량은 동일한 기준치가 요구된다(National institute of forest science, 2017). 이외에도 건설폐목재, 생활폐목재, 살충소독처리목재, 방부처리목재와 그 밖의 유해물질 오염이 우려되는 목재나 대나무, 종실, 톱밥, 대팻밥, 수피 등의 사용이 제한된다(National institute of forest science, 2017).

우리나라의 별채 현황을 살펴보면 2016년 국내 별채량은 767만m³이며, 이 중 별채산물 수집량이 약 525만m³으로 242만m³의 미이용 산림바이오매스가 발생하고 있다(Korea forest service, 2017). 따라서, 전체 별채량의 약 32%를 차지하는 미이용 산림바이오매스는 다양한 목재산업 분야에서 활용되어야 한다.

현재 국내 성형목탄 제조업체들은 목재법과 국립산림과학원 고시의 시행으로 사용원료 변화를 추진하였고, 현재 미이용 산림바이오매스(Korea forest service, 2018) 중 임목의 가지 및 뿌리를 주원료로 목탄을 생산하고 있다(Choi, 2018). 또한, 국내에서 생산되어 유통 중인 성형목탄이 중금속 기준치를 초과하여 적발되는 경우가 늘어나고 있는 실정이다. 이러한 상황은 목재제품에 대한 품질기준이 통합되고 관리되면서 성형목탄이 제도화되기 위한 과정으로 볼 수 있지만, 현재에는 관련된 산학연·관·민간의 다양한 의견이 분분한 상황이다(Ahn et al., 2013; Kwon et al., 2018; Yang and Han., 2018).

성형목탄 제조를 위해 국내에서 가장 많이 사용하고 있는 방식은 평로탄화로를 이용한 방법으로 땅속에 일정한 공간을 파내어 탄화실을 만든 후 적당한 높이로 미처리 원료를 쌓아 약 300~400°C의 온도로 탄화시키고 물이나 흙을 뿐려 소화시킨다(Yang et al., 2017; Magnone et al., 2019). 이후 새로운 미처리 원료를 소화된 탄화물 위로 재투입하여 앞서 말한 방법과 같이 탄화 후 소화시키는 과정을 수차례 반복하여 수행한다. 최종적으로 탄화물이 탄화실 높이까지 다다르면 탄화물을 꺼내어 성형목탄의 원료로 사용된다. 이러한 탄화 방식을 통해 생산된 원료는 가장 오래 탄화가 진행된 최하단층의 원료와 마지막에 탄화된 최상단층의 원료의 탄화 시간이나 탄화 과정 동안 흙과 같은 이물질이 특정한 층에 퇴적되는 현상이 발생한다.

이에 본 연구에서는 미이용 산림바이오매스를 평로탄화로에서 탄화하여 생산된 탄화물을 투입 시기에 따라 채취하여 기계적 전처리를 통해 입자 크기별로 분류한 후 탄화처리 전·후와 투입 시기, 탄화 시간, 입자 크기 등에 따른 탄화시료에 대한 연료적인 특성을 비교하고 이를 통해 평로탄화로에서 생산된 성형목탄 원료에 대한 품질을 평가하고자 하였다.

2. 재료 및 방법

2.1. 공시재료

본 연구에서는 경상북도 철곡군에 위치한 성형목탄(wood charcoal briquette) 제조업체가 사용하는 평로탄화로에서 생산된 원료를 활용하였다. 공시재료는 숲가꾸기로부터 발생된 별채부산물을 성형목탄 생산자가 구매한 것으로 침엽수 혼합수종이다. 탄화원료는 약 700°C의 온도조건에서 탄화되어 약 80시간 동안 소화되었다. 본 연구에서는 침엽수 호그 (Hog of Soft wood, HSW)와 탄화된 침엽수 호그 5종 (Carbonized hog of soft woods, CSW)을 이용하였다(Fig. 1). 탄화된 호그는 평로탄화로의 탄화 위치에 따라 최상단으로부터 일정한 간격으로 5등분으로 분류하여 채취하였으며, 최상층으로부터 CSW1, CSW2, CSW3, CSW4, CSW5을 채취하여 공시재료로 사용하였다(Fig. 2). 채취된 시료는 시료 상태를 잘 보존하기 위해 2중으로 밀봉하여 상온에서 보관하였다.

2.2. 기계적 선별

각각의 층으로부터 얻어진 성형목탄에 대한 입자 크기의 분포 분석을 위해 Fig. 3에 제시된 것처럼 분쇄하여 입자 크기별로 선별하였다. 모든 시료는 선별의 용이성을 위해 65°C 오븐에서 약 48시간 건조시킨 후 사용하였다. 분쇄(Fig. 3-①)는 절단밀(Pulverisette 19, FRITSCH, Germany)을 사용하였다. 절단 밀의 회전 속도는 300 rpm으로 가동시켰고 1 mm의 직경을 가진 체를 사용하였다. 입자 크기 선별은 진동식 체진동기(analysette 3, FRITSCH, Germany)와 40 mesh (0.4 mm), 100 mesh (0.149 mm)의 크기를 갖는 표준체를 사용하여 40 mesh 이상(Top), 40~100 mesh(Middle), 100 mesh 이하(Bottom)로 분류하였다. 선별 과정에서 입자 크기 분포를 분석하기 위해 입자 크기별 수율을 계산하였으며 계산식은 Equation (1)과 같다.

$$Y_P (\%) = \frac{W_P}{W_S} \times 100 \quad (1)$$

Y_P = 해당 입자크기의 수율(%)

W_P = 해당 입자크기의 무게(g)

W_S = 원시료 무게(g)

2.3. 공업분석

공업분석은 수분회분측정기(PrepASH229, Precisa, Swiss)를 사용하여 시료의 함수율, 휘발성 화합물, 회분, 고정탄소를 분석하였다. 분석에는 약 1 g의 시료가 사용되었다. 분석 조건은 국제표준화기구(ISO, International Organization for Standardization)의 회분 함량 측정(ISO18122 Solid biofuels-Determination of ash content), 휘발성 화합물 함량 측정(ISO18123 Solid biofuels-Determination of the content of volatile matter), 함수율 함량 측정(ISO18134-3 Solid biofuels-Determination of moisture content-Oven dry method)을 참고하여 수행하였다. 고정탄소의 경우 Equation (2)를 통해 계산되었다.

$$FC(\%) = 100 - (MC + VM + ASH) \quad (2)$$

Fixed carbon (FC) = 고정탄소 함량(%)

Moisture content (MC) = 함수율(%)

Volatile matter (VM) = 휘발물 함량(%)

Ash = 회분 함량(%)

2.4. 발열량 측정

발열량은 발열량 측정기(Parr 6400, Parr, USA)를 사용하여 측정하였으며, ISO/DIS 18125 Solid biofuels – Determination of calorific value를 참조하여 수행하였다. 분석 시료는 약 0.5 g의 분말을 정착하여 10 t의 압력으로 직경 13 mm의 웰릿 형태로 성형하여 측정에 사용하였으며, 시료는 105±3°C 조건의 오븐에서 3시간 이상 전진시킨 후 측정에 사용하였다.

2.5. 원소함량 분석

원소함량은 원소분석기(vario MICRO cube elemental analyzer, elementar, Germany)를 사용하여 분석하였으며, 탄소, 수소, 질소, 황 함량을 측정하였다. 산소 함량은 Equation (3)에 의해 계산되었다. 분석조건은 1150±5°C로 유지되는 연소관에서 완전연소시켜 발생된 가스를 600 ml/min의 유량으로 헬륨을 흘려 850±5°C로 유지되는 환원관으로 이동시킨 후 N₂, CO₂,

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H_2O , SO_2 로 환원시켜 분석하였다. 분석에는 50 mg의 분말을 정침하여 사용되었다. 표준물질에는 2693(NIST, USA)가 사용되었다. 분석에 사용할 시료는 $105 \pm 3^\circ\text{C}$ 조건의 오븐에서 3시간 이상 전진시킨 후 원소함량을 측정하였다.

$$\text{O} (\%) = 100 - (\text{C} + \text{H} + \text{N} + \text{S}) \quad (3)$$

$$\begin{aligned}\text{C} &= \text{탄소 함량} (\%) \\ \text{H} &= \text{수소 함량} (\%) \\ \text{N} &= \text{질소 함량} (\%) \\ \text{S} &= \text{황 함량} (\%) \\ \text{O} &= \text{산소 함량} (\%)\end{aligned}$$

2.6. 에너지 수율 계산

선별공정을 통해 획득한 시료들의 에너지 수율(Y_E)은 Equation (4)을 이용하여 계산하였다. 에너지 수율 계산식은 전진중량을 기준으로 계산한다.

$$Y_E (\%) = \frac{\text{HHV}_P}{\text{HHV}_S} \times Y_P \quad (4)$$

$$\begin{aligned}Y_E &= \text{에너지 수율} (\%) \\ \text{HHV}_P &= \text{해당 입자크기의 발열량} (\text{MJ/kg}) \\ \text{HHV}_S &= \text{원시료의 발열량} (\text{MJ/kg})\end{aligned}$$

3. 결과 및 고찰

3.1 입자 분포

성형목탄 시료의 기계적 전처리를 통해서 분류된 입자 크기별 중량 수율은 Fig. 4와 같다. 미처리시료인 HSW의 경우 Top (>40 mesh), Middle ($40\sim100$ mesh), Bottom (<100 mesh)이 각각 43.6%, 30.6%, 25.9%의 범위로 분포하였다. 탄화시료인 CSW1~CSW5의 경우 Top, Middle, Bottom 수율이 각각 약 10.7~16.6%, 17.6~20.0%, 63.8~71.2%의 범위로 나타났다. 선행연구들에 따르면 탄화 처리는 목재의 구성성분인 헤미셀룰로오스, 셀룰로오스, 일부 리그닌의 열분해를 야기하며, 이에 따라 목재의 탄소 함량을 증가시켜 결과적으로 목재의 취성을 향상시키는 것으로 보고되었다(Lee et al., 2015; Mei et al., 2015). 따라서 동일한 분쇄 처리를 가하였음에도 분쇄성이 뛰어난 탄화시료가 미처리시료 보다 상대적으로 100 mesh 이하의 미세입자를 많이 획득할 수 있었던 것으로 사료된다. 탄화시료의 경우, 탄화 시 평로탄화로 내에서의 위치에 따라서 Top, Middle, Bottom 수율의 소폭 변화하였지만 유의미한 차이를 나타내지는 않았고 비교적 유사한 입자분포 경향을 나타냈다.

3.2. 발열량, 공업분석, 원소분석

미처리시료인 HSW과 5종의 탄화시료 CSW에 대한 발열량, 공업분석, 원소분석 결과는 Table 1과 같다. 휘발성 화합물의 경우 HSW에 비해 CSW1~CSW5에서는 최소 53.5%에서 최대 57.7%까지 낮게 나타났다. 고온의 탄화 과정 중 시료 내 휘발성 화합물이 대부분 기체로 유리되면서 탄화시료의 휘발성화합물 함량이 미처리시료에 비해 크게 감소한 것으로 사료된다. 이에 반해 회분 및 고정탄소의 함량은 미처리시료(14.0%, 17.6%)에 비해 각각 높은 20.7~34.0%, 21.1~33.1%의 범위로 나타났다. 선행연구에 따르면 목재 내의 미네랄 성분이 탄화과정을 통해 4~5배정도 목탄에 농축되어 회분으로 남는다고 보고되었다(Lee and Kim, 2010). 따라서 이와 같은 이유로 탄화시료가 미처리시료에 비해 높은 회분 함량을 나타내는 것으로 사료된다. 탄소 함량은 HSW은 39.9%로 나타났으며, CSW1~CSW5의 경우에는 HSW에 비해 소폭 상승한 42.9~50.9%의 범위로 나타났다. Lee 등(2007)의 연구결과에 의하면 탄화온도 $300\sim400^\circ\text{C}$ 에서 처리된 잣나무의 탄소 함량은 67.7~72.2%의 범위로 본 연구결과에서 제시된 탄소 함량보다 높았다. 이는 원목의 벌채 후 땅에 놓여있는 원료를 수거하는 과정이나 채취한 원료의 운반 및 보관과정 또는 평로탄화로에서 탄화하는 과정에서 흙과 같은 불순물이 많이 포함되어있기 때문에 예상되며, 상기 높은 회분 함량 결과도 이와 같은 원인에 기인하는 것으로 사료된다. 한편, 수소와 산소의 함량은 미처리시료에 비해 탄화시료에서 대부분 감소하였는데, 이는 수소 및 산소가 탄화 중 수증기와 이산화탄소의 형태로 분리되었기 때문으로 판단된다. 질소와 황 성분은 상대적으로 미량으로서 유의미한 변화 양상을 보이지 않았다. 발열량은 평균적으로 탄화에 의해 증가되었다. 탄화 위치에 따라 중간층인 CSW3에서 20.4 MJ/kg으로 가장 높은 발열량을 보였다. 또한, 하단층(CSW4, CSW5)은 상단층(CSW1, CSW2)

에 비해 발열량이 약 1.7 MJ/kg 높게 나타났다. 본 결과에서 평로탄화로에서 생산된 탄화시료의 발열량은 탄화 시간은 크게 영향을 주지 않지만, 탄화 과정에서 산소 유입의 차단이 영향을 주는 것으로 판단된다.

탄화시료(CSW)에 대해 분쇄 후 입자 크기에 따른 공업분석, 원소분석, 발열량 측정 결과는 Table 2와 같다. 휘발성 화합물 및 고정탄소 함량은 입자 크기가 작아질수록 높아지는 경향을 나타냈으며 각각 7.4%, 26.0%(Top)에서 15.5%, 52.3%(Bottom)으로 함량이 약 2배 증가하였다. 회분의 경우 입자 크기가 작아질수록 66.6%에서 32.2%까지 감소하는 경향을 나타냈는데, 이는 분쇄공정 시 탄화시료와 섞여있는 흙 등 불순물 대부분이 40 mesh 보다 입자 크기가 커서 표준체를 통과하지 못하고 Top에 집적된 결과로 사료된다. 탄소 함량의 경우 Bottom에서 58.8%로 가장 높게 나타났으며, 입자 크기가 작아질수록 증가하여 고정탄소와 유사한 경향을 나타냈다. 이와 반대로 산소함량은 입자 크기가 작아짐에 따라 소폭 상승하였으며, 황 함량은 변화폭이 상대적으로 거의 없었다. 발열량의 경우에도 Bottom에서 21.3 MJ/kg으로 가장 높은 발열량을 나타내었으며, 국립산림과학원 고시 제2018-8호 목재제품의 규격과 품질기준의 부속서 14의 성형목탄 기준에서 숯가루성형탄의 품질기준(17.6MJ/kg)을 상회하는 결과를 보였다. 선행연구에서도 입자 크기가 작은 조건에서 탄화된 경우 탄소함량이 높고 발열량 또한 증가된다고 보고되어(Hwang *et al.*, 2012), 탄화시료의 경우 입자 크기가 작은 Bottom에 에너지가 집약됨을 확인할 수 있었다(Hwang *et al.*, 2012). 이와 같은 결과들을 고려하면, 탄화시료의 분쇄 및 선별 시 입자 크기가 작은 시료를 선택하여 야 성형목탄 품질이 향상될 것으로 사료되며, 또한 흙과 같은 불순물이 다량 포함된 시료일수록 보다 우수한 전처리 효과를 볼 수 있을 것으로 기대된다.

3.3. 에너지 수율

각 시료의 발열량에 따른 에너지 수율 계산 결과는 Table 3과 같다. 본 연구에서의 에너지 수율은 기계적 선별 공정의 중량수율과 발열량을 고려한 값으로 원시료의 총 에너지 중 선별 공정을 통해서 최종적으로 얻을 수 있는 에너지 비율을 나타낸다. 미처리시료인 HSW의 경우 에너지 수율은 각각 Top (0.46), Middle (0.31), Bottom (0.21) 순으로 높게 나타났다. 이는 HSW의 분쇄성이 상대적으로 우수하지 못하여 초기 결과(Fig. 4)처럼 입자 크기가 작을수록 중량 수율이 낮아지기 때문에 결과적으로 에너지 수율 또한 입자 크기가 낮을수록 줄어든 것으로 사료된다. 반면, 탄화시료의 경우 Top, Middle, Bottom 시료의 에너지 수율은 각각 0.05~0.12, 0.12~0.17, 0.70~0.82의 범위로 나타났으며, 입자 크기가 줄어들수록 에너지 수율이 급격히 증가하는 경향을 나타냈다. 이는 탄화시료의 경우 입자 크기가 작은 그룹이 중량수율도 높았을 뿐만 아니라 발열량도 높아 이를 모두 고려한 에너지 수율에서 보다 큰 차이를 보인 것으로 사료된다. CSW1의 Bottom에서 0.82로 가장 높은 에너지 수율을 나타냈고 CSW3의 Bottom에서는 20.67 MJ/kg으로 가장 높은 발열량을 보였지만 0.71로 비교적 낮은 에너지 수율을 보였다. 이는 Bottom의 높은 발열량에 비해 낮은 중량수율을 가지기 때문으로 판단된다. CSW4의 Total과 미처리원료의 발열량을 보면 탄화가 이루어졌다고 판단하기 어려웠지만 선별을 통해 Bottom에서 발열량이 크게 상승한걸 살펴볼 수 있다.

3.4. 상관관계 분석

탄화시료인 CSW1~CSW5의 입자 크기에 따라 분류된 Top, Middle, Bottom의 발열량, 휘발성 화합물, 회분함량, 고정탄소, 탄소함량의 상관관계 분석 결과는 Fig. 5에 제시하였다. 탄화시료의 입자 크기에 따른 회분함량은 탄화시료의 고정탄소와 통계적으로 높은 양의 상관관계를 보였으며, 가장 높은 결정계수를 보였다($R^2=0.9886$). 이러한 이유는 본 연구의 탄화시료의 입자 크기가 작아질수록 회분함량이 줄어드는 경향이 뚜렷하기 때문으로 판단된다. 탄화시료의 발열량은 탄소함량과 밀접한 상관관계($R^2=0.9085$)를 보였으며, 이는 탄화가 진행되면서 목재 내의 수소와 산소 함량이 감소하면서 탄화시료 내에 높은 결합 에너지를 가지는 C-C 결합의 증가하기 때문으로 판단된다(Lee *et al.*, 2016). 또한 발열량은 고정탄소와 통계적으로 유의한 양의 상관관계($R^2=0.8363$)를 나타냈다. 입자 크기에 따른 탄화시료의 발열량은 회분과 통계적으로 유의한 부의 상관관계($R^2=0.8551$)를 나타내었다. Table. 2에 제시한 것처럼 탄화시료의 입자 크기가 작아짐에 따라 회분함량도 감소하였으나 발열량은 증가되는 경향을 보이기 때문에 판단된다. 탄화시료의 발열량과 휘발성 화합물은 분포로 통계적으로 유의하지 않은 결과를 보였다. 회분과 휘발성 화합물, 탄소함량의 상관관계를 살펴보면 통계적으로 유의한 음의 상관관계($R^2>0.83$)를 보였다. 휘발성 화합물과 고정탄소는 양(+)의 상관관계($R^2=0.7956$)를 보였으며, 마찬가지로 탄소함량과 휘발성 화합물, 고정탄소와의 상관관계도 유의한 양(+)의 상관관계($R^2>0.75$)로 나타났다.

Characteristics of Carbonized Biomass Produced in a Manufacturing Process of Wood Charcoal Briquettes Using an Open Hearth Kiln

4. 결 론

본 연구에서는 평로탄화로를 통한 성형목탄 제조공정을 통해 생산되는 탄화시료의 특성을 분석하였다. 평로탄화로의 위치에 따라 채취한 탄화시료의 경우 탄화 시간이 가장 긴 하단층에서 높은 품질을 나타낼 것으로 예상하였지만 중간층, 상단층, 하단층 순으로 높은 품질을 나타내었다. 이는 평로탄화로를 통한 탄화 방식의 경우 연료의 품질은 탄화 시간과는 연관성이 없고 상단층과 하단층 양방향에서의 열전달로 인해 중간층에서 비교적 균일한 탄화가 진행되어 가장 높은 품질을 보였다. 탄화시료의 입자 크기에 따라 분류하여 분석한 결과로 입자 크기가 작아질수록 발열량, 탄소함량, 고정탄소함량이 높아졌으며, 특히 100 mesh 이하의 탄화시료에서 연료의 품질을 향상시키는 발열량, 탄소함량, 고정탄소함량이 각각 21.3 MJ/kg, 58.8%, 52.3%로 나타났다. 기 가동 중인 공정상 탄화 전 시료를 선별할 수 없는 경우, 탄화 후에 분쇄 및 100 mesh 이하의 탄화시료를 선별할 때 비교적 높은 품질 향상을 기대할 수 있다.