Petroleum & Coal
ISSN 1337-7027

Available online at www.vurup.sk/petroleum-coal
Petroleum & Coal 56(2) 188-195, 2014

PHYSICAL AND CHEMICAL PROPERTIES OF COAL BRIQUETTES FROM BIOMASS-BITUMINOUS BLENDS

N. Gholipour Zanjani[1,2], A. Zarringhalam Moghaddam[1], S. Dorosti[1]

[1]Chemical Engineering Department, Tarbiat Modares University, Tehran , Iran P.O.Box14115-143, zarrin@modares.ac.ir;[2]Petrochemical Engineering Department, Faculty of Chemistry and Petrochemical Engineering, Standard Research Institute, Karaj, Iran P.O.B 31745-139, n.gholipour@standard.ac.ir

Received January 7, 2014, Accepted May 15, 2014

Abstract

In this work, the physical and chemical properties of briquettes of bituminous coal and sawdust/beet pulp have been investigated. Physical stability of sawdust briquettes was studied by water resistance index and compressive strength considering the effect of moisture, composition of briquettes and pressure of briquetting. The chemical tests on briquettes revealed the decrease in the sulfur content and the increase in volatile matter by increasing the amount of biomass. The higher calorific value and the lower volatile matter of the beet pulp briquettes in compare with sawdust briquettes make them more favorable as a solid fuel.

Keywords: briquetting; bituminous coal; beet pulp; sawdust; Fuel.

1. Introduction

Considering important message of world standard day, 2012, "less waste, better result", improving the ways to decrease waste is too valuable. Briquetting is a method to utilize coal fines, as a byproduct of mining, to decrease lost of this energy source. On the other hand, biomass derived from plant like sawdust, rice husk, palm kernel shell could be recycling to valuable fuels [1-3] Incorporating biomass from agricultural waste in coal briquetting formulations has proven to be an effective method to produce the better quality fuel. Using materials which are abundant in waste streams and has desirable energy and binding characteristics to provide a briquetted fuel could be an economic and ecologic viable substitute for conventional coal [4]. This method not only saves high calorific value fuels and reduces waste disposal problems of mines, but also enables to formulate clean solid fuel [3].

Sawdust, rice husk, palm fiber and shell are popular biomass to prepare briquettes [1-3]

In this research, sawdust and beet pulp were used in briquetting formulation. Beet pulp as a byproduct of sugar refining, because of its abundance and low price, can be a suitable candidate for briquette's formulation. However, limited information on utilization of beet pulp as a fuel is cited [5-7]. Sawdust is also, readily available in large quantities, as a waste from the wood processing industries. Through the last decades, the use of sawdust to improve the calorific value and better quality of low rank coal have been investigated [3,8-12]. Application of sawdust and its low temperature char as co-fuel with coal has shown better performance in terms of ignition index and burn out efficiency as compared to coal [13]. Producing the ash with high levels of silicon and aluminum and less capacity to adhere to the surfaces after co-firing its blend with bituminous make it favorable to solve the environmental problems of the fuel [14].Moreover, composite briquettes of sawdust and charred palm kernel under standard cooking procedure have shown low fuel consumed and time required for cooking [15]. Sawdust briquettes has shown better storage and stability characteristics compare with rice husk, peanut, coconut and palm briquettes [9].

There are various researches on biomass briquetting; however a better understanding of the physical and chemical properties of briquettes could lead to better performance in order to reach the high quality standards.

This work reports physical and chemical property of fuel briquettes prepared with bituminous coal (extracted from Mezino (I) coalfield), beet pulp/ sawdust, tar and calcium carbonate. The compressive strength and water resistance were studied to evaluate the ability of briquettes to withstand loads and humidity during transportation and storage. The effect of two types of biomass (sawdust and beet pulp) on the chemical properties of briquettes was also studied.

2. Experimental

2.1 Materials

The bituminous coal from Mezino I coalfield was used in this study (The major coal field in Iran with low production of ash) [16].

Beet pulp and sawdust were used as biomass in briquettes. Calcium carbonate and tar were used, respectively, to reduce SOx emission and to improve physical strength. The characteristics and chemical analysis of the coal and additives are given in Table 1. The elemental analysis, calorific value, moisture, volatile matter and ash were determined at Research Institute of Petroleum Industry according to the standard methods [17-19].

Table 1 Characteristics of the coal and binders

sample	Calorific value (kJ/kg)	Moisture (w %)	Volatile matter (w %)	Ash (w %)	Elemental analysis (w %)			
					C	N	H	S
Mezino I (coal)	29018.7	2.07	9.02	13.99	71.6	1.5	2.8	2.67
beet pulp	15019.68	29.2	50.2	7.9	30.3	2.3	7.5	0.70
sawdust	8605.54	9.8	80.46	2.9	51.0	---	5.6	0.002
tar	40280.00	--	85	--	86.1	1.0	10.1	2.83

Beet pulp and sawdust were generated by Karaj sugar beet factory and Eslamshahr wood mill. Calcium carbonate was supplied by Merck Company with molecular weight equal to 100.09 g/mol and impurity of 0.005% sulfate. Tar was supplied by Tehran oil refinery. The mentioned materials were mixed by hand according to a designed formulation and pressed to prepare briquettes.

2.2. Equipment

Self-developed briquetting machine and compressive strength testing machine were used to produce briquettes and determination of compressive strength according to our previous work [16]

2.3. Samples Studied and Procedures

The raw coal was ground in the jaw crusher. It was pulverized in a ball mill to produce coal fines. One kilogram of fine coal with particle size less than 1 mm was floated in water in a mixture-tank. The purpose of the floating was to decrease ash and sulfur content of the coal for briquetting.

Then few drops of gasoline and pine oil were added to the mixture, the floated coal was collected and dried in a vacuum oven. Sawdust was screened to particle size less than 3 mm. The powdered dried coal, water, beet pulp, sawdust (with particle size less than 3 mm), Calcium carbonate and tar were mixed according to the following design. The formulated mixture was poured into a mold and heated for 15 min at about 120^{o}C in a briquetting machine. The briquettes were stored under ambient conditions and some samples were taken to be characterized.

Taguchi method was used for experimental design and the samples were formulated according to the following descriptions. There were four variables: mass percent of sawdust to tar, water and calcium carbonate and briquetting pressure. For each variable, three levels were selected, based on its range. Therefore, to investigate three level factors, L9 array including nine tests was used. Table 2 shows the selected factors and three levels. Table 3 shows the experimental design of the briquetting. Approximately 65% to 80% of each prepared sample was coal. The rest was sawdust, tar, water and calcium carbonate.

Table 2 Experimental variables and levels selected for briquetting

	variable	Level 1	Level 2	Level 3
A	mass percent of biomass to tar	10/10	15/8	20/5
B	briquetting pressure (Ton)	18	20	25
C	mass percent of water	0	3	5
D	mass percent of $CaCO_3$	0	2.5	5

Table 3 Design of experiment for briquetting according to Taguchi's L9 orthogonal array

Experimental runs	A	B	C	D
1	1	1	1	1
2	1	2	2	2
3	1	3	2	3
4	2	1	2	3
5	2	2	3	1
6	2	3	1	2
7	3	1	3	3
8	3	3	1	1
9	3	2	3	2

Similar samples were produced by substituting beet pulp instead of sawdust and analyzed.

2.4. Test Procedure

The compressive strength and water resistance index (WRI) were determined according to our previous work [16]. Each reported data in Table 4 represents the average value of two determinations.

Table 4 Compressive strength and WRI for designed experiments

Experimental runs	Compressive strength(kPa)	WRI
1	282.5	94
2	493.35	96
3	493.35	95
4	536.63	83
5	471.34	95
6	612.38	90
7	460.36	75
8	629.59	70
9	629.7	77

Analysis of means (ANOM) was used to determine mean value of each factor (CS or WRI) in each level. For example, the results of the three experiments consisting of level 1 were added and then divided by three, which gave the mean values of level 1 [20-21]. Thus, the mean value of CS for factor A (mass percent of sawdust to tar) at level 1 is (282.5+493.35+ 493.35)/3= 423.399. The results of the similar calculations are shown in Table 5 and discussed in part 3.1.

Table5 The amount of CS and WRI in each level of variables

variable	Compressive strength (kPa)			Water resistance index		
	Level 1	Level 2	Level 3	Level 1	Level 2	Level 3
A	423.40	540.12	594.22	95.00	89.33	74.00
B	426.83	531.43	599.48	84.00	87.00	87.33
C	508.49	574.23	475.02	84.66	85.33	88.33
D	482.18	522.03	553.19	88.66	87.00	82.66

Analysis of variance (ANOVA) was used to investigate which factor significantly influences WRI or CS. The results of related calculations were shown in Tables 6 and 7.

Table 6 Variance analysis for compressive strength

variable		Sum of square	D.O.F	Mean Square	Variance ratio	P
A	mass percent of sawdust to tar	44121.39	2	22060.69	3.63	Not significant
B	briquetting pressure (ton)	44710.32	2	22355.16	3.68	Not significant
C	mass percent of water	20708.85	2	10354.42	1.70	Not significant
D	mass percent of $CaCO_3$	12142.54	2	6071.27	1.00	Not significant

Table 7 Variance analysis for WRI

variable		Sum of square	D.O.F	Mean Square	Variance ratio	P
A	mass percent of sawdust to tar	708.22	2	345.11	35.02	significant
B	briquetting pressure (ton)	20.22	2	10.11	1.00	Not significant
C	mass percent of water	22.89	2	11.44	1.13	Not significant
D	mass percent of $CaCO_3$	57.55	2	28.77	2.85	Not significant

The elemental analysis, calorific value, moisture, volatile matter and ash content of briquettes were determined at Research Institute of Petroleum Industry (Tehran, Iran) according to the standard methods [17-19].

3. Results and discussion

3.1 Physical stability

3.1.1 Compressive Strength

In producing briquettes the main point is to convert the fine coal waste to agglomerated structure that can be readily handled and fed to the combustion chamber. Thus the integrity and strength of the produced briquettes is of prime importance. In this study, the effect of four variables: sawdust to tar mass ratio, briquetting pressure, mass percent of water and $CaCO_3$ on the CS is presented in Table 5. It can be seen that, the decrease on the amount of the coal or the increase on the amount of the biomass leads to a high strength briquettes. The maximum CS was obtained with sawdust to tar mass ratio (20/5=) 4. The increase on the mechanical strength is attributed to the fibrous structure of sawdust [3].

The briquetting force is another influencing parameter. As Table 5 shows it increased the CS of the samples. This could be related to the binding effect of the natural lignin present in the wood which increases with the increase of the pressure [22-23]. Another effect of the pressing load is the decrease of moisture and the increase of the density of the briquette. It is predictable that commercial-scale briquetting equipment will be able to generate pressures much greater than 25 ton (of this experiment), and consequently produce stronger briquettes [4].

The effect of moisture on CS is also presented in Table 5. According to the results, the CS increased with an increase in water content from 0 to 3%, although a decrease in the fracture behavior of the briquettes was observed at moisture of 5%. It can be seen that the lowest compressive strength occurred when 5% water was added. The reason for the initial increase of CS is probably the increase of adhesion of particles with 3% water. Nevertheless with further increase of water, the porosity increases and the firming of briquettes reduces. This behavior is in the agreement with the effect of moisture on the compressive strength of Guar gum/bituminous coal/sawdust briquettes reported by D. Taulbee *et al.* [24]. Generally, coals and fuels contain a large amount of water resulting in a low calorific value and unfortunately dry coals tend to have high spontaneous combustion tendency which causes serious problems for transportation and storage [25]. Considering the amount of water percent 2.07 in crude fuel and 9.80 in sawdust, adding 3% water not only could result suitable CS but also could save the thermal properties of briquettes.

Table 5 shows that the increase of the calcium carbonate amount in the briquettes has a little effect on the compressive strength. Taulbee *et al.* [24] works have shown that adding lime in the formulation of sawdust/coal briquettes with different binders (such as molasses, paper sludge and coal tar) have made higher strength briquettes than those without lime [24]. However, this has been reversed in the presence of guar gum and starch-based binders. Their results indicate that the chemical nature of the binder influences the performance of the $CaCO_3$. Thus the use of the tar in present formulations has discernible effect on the CS.

According to the results, the ideal parameter to achieve the highest CS would be the sawdust to tar content ratio 20/5; water content 3% and calcium carbonate 5% at 25 tonnes briquetting pressure.

Secondly, the compressive strength of the similar briquettes without sawdust was determined; 402.99 kPa. This value is lower than compressive strength of all sawdust briquettes studied in this article. Therefore, sawdust improves the mechanical properties of the briquettes.

Further analysis was carried out by ANOVA, to identify significant parameters. The results were summarized in Table 6. The degrees of freedom, sum of squares, mean square and the variance ratios were calculated and analyzed according to our last work [26].

It can be seen that the variance ratio of the sawdust parameter is 3.63 which is less than $F_{0.05}(2,2)$ = 19.00 and categorized as insignificant. The influences of the variation of other parameters on CS are also evaluated as insignificant.

While the effect of variation of sawdust to tar content ratio are not significant on CS, the presence of sawdust increased the CS from 402.99 kPa to 594.22 kPa. So it can be concluded that sawdust improves the CS of the briquettes.

3.1.2 Water Resistance Index (WRI)

The WRI of briquettes shows the resistance against disintegration and adsorption of water during transportation and storage. Table 5 shows the effect of four variables: sawdust to tar mass ratio, briquetting pressure, mass percent of water and $CaCO_3$ content on the WRI. According to the results, the WRI decreased with an increase in the sawdust/tar ratio. Although the presence of the tar improves the WRI of the briquettes, the cellulosic nature of the sawdust increases water absorption and subsequently the intention to disintegrate in water.

Table 5 shows the effect of briquetting pressure on the water resistance index. The slight increase of WRI is due to the high density briquettes, resulted from the increase of pressing load. Similar results reported on coal/sawdust/Guar Gum briquettes, although the reverse effect has been observed when the binder Guar Gum was replaced with Reax (lignosulfate) +Lime or Wheat starch [24]. It shows the effective role of binder (tar, Guar Gum, lime) on WRI.

According to Table 5, the influence of water content on WRI was small. It changed from 84.66 in the absence of water to a maximum 88.33 in the sample with 5% water. However, as mentioned previously, the CS of 5% water sample is much lower than sample containing 3% water. Thus, the optimum water content in the present formulation would be 3%.

The detrimental effect of $CaCO_3$ content on the WRI can be observed from data in table 5. This could be due to the solubility of this material in water, which disintegrates briquette's structure [27].

Analyzing the results relating to the effects of the different parameters on WRI suggest that the ideal composition and processing condition to achieve the highest WRI is the ratio of sawdust to tar content 10 to 10; water content 5% and 25 tonnes briquetting pressure without using calcium carbonate.

According to variance analysis in Table 7, sawdust to tar content ratio was the most effective variable on the water resistance index, among the mentioned four variables. It has the significant effect on WRI, because it's variance ratio, 35.02, is more than $F_{0.05}(2,2)$= 19.00. Thus this variable should be considered when preparing briquettes with high WRI.

Considering the effect of variables on physical properties, WRI and CS follow the opposite trend with the change of sawdust to tar content ratio that it may be due to the intention of adsorption of water by sawdust. It is noteworthy that the ratio of sawdust to tar causes significant effect on WRI, but has insignificant effect on CS. On the other hand, increase of briquetting pressure shows positive but insignificant effects on both WRI and CS.

Based on the above analysis , the best composition and process condition for high WRI and CS would be sawdust to tar content ratio 15/8, water content3% ,calcium carbonate 2.5% and 25 tonnes briquetting pressure.

3.2. Comparison of Sawdust / Coal and Beet Pulp / Coal Briquettes

Similar biomass/coal briquettes were prepared with beet pulp instead of sawdust, to compare their chemical and physical properties (Figures 1-4). The sulfur is an undesirable content of the coal and its reduction has been the subject of numerous researches [20,28]. Figure 1 shows that the amount of sulfur in sawdust briquettes is lower than beet pulp briquettes. This could be due to the lower sulfur content of sawdust (Table 1). Figure 1 also depicts that with the increase of biomass, the amount of sulfur was reduced. Besides, Table 1 shows that, beet pulp and sawdust have lower percent of sulfur than coal. Therefore, the amount of SO_x release from the combustion of biomass/coal briquettes will always be less than that of coal.

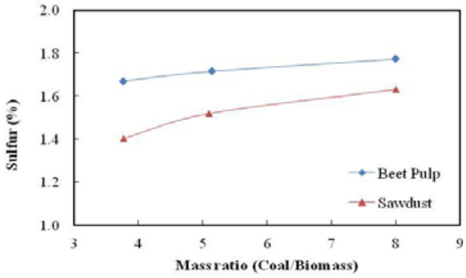

Fig.1. Effect of natural binders on the amount of sulfur

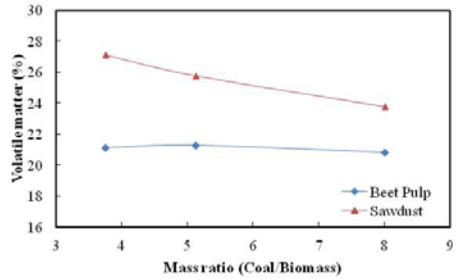

Fig.2. Effect of natural binders on the amount of volatile materials

The volatile matter decreased with the increase of the amount of coal/biomass mass ratio (or decrease of the amount of biomass in briquettes) as shown in Fig. 2. This could be because thermal decomposition of biomass starts at lower temperatures and proceeds faster within a relatively narrower temperature range than that of coal does [7]. The interesting point is that biomass combustion alone could produce fine particulates with a size of less than 2 micrometer but, burning the biomass with coal shifts the particle size distribution from fine particles to coarse particles, which can be easily captured by dust collection systems [29]. As it can be observed the VM of sawdust briquettes is more than that of beet pulp briquettes (Fig. 2). This could be related to provide enough hydrogen-donors at the sawdust blends to prevent the recombination reactions [30]. Although there are the higher volatile content of the sawdust, the studies show that co-firing sawdust (with a larger mean particle size than 0.3 mm as used in this work) and coal has little effect on NO emissions comparing with firing coal under air-fired conditions [31]. However, lower conversions of fuel-N to NO are usually shown for the blended fuels in comparison to coal, especially with an increasing biomass percentage in the blends [32].

One of the important features of a solid fuel is its calorific value (C.V.). It also determines the commercial value of the fuel. The C.V. of briquettes is compared in Fig. 3. In present set of experiments, the minimum C.V. was 25499.7 KJ/Kg related to 75/20/5 coal/sawdust/tar and the maximum value was 28100.5 KJ/Kg relating to 80/10/10, coal/beet pulp/ tar. As it is expected the presence of biomass reduced the C.V. than that of raw coal. However, the C.V. of the produced briquettes containing sawdust or beet pulp was above 24540 KJ/Kg, which belongs to high rank fuel category [33].

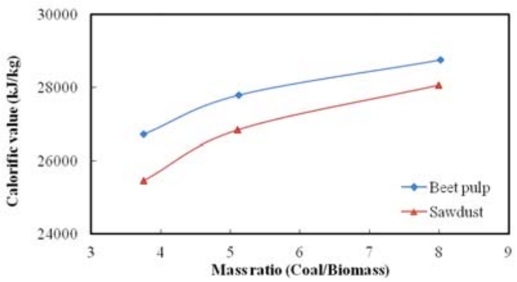

Fig.3. Effect of natural binders on the calorific value

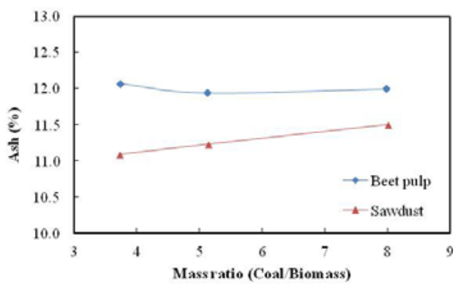

Fig.4. Effect of natural binders on the ash percent

Figure 4 shows that with the increase of the sawdust, the remained ash percent decreased. The ash from beet pulp briquettes was more than that of sawdust briquettes; it could be because of considerable amounts of volatile matter that remain in the char of beet pulp and could evolve in further heating more [7]. As it can be observed the ash produced from firing the briquettes of beet pulp-coal is akin to olive stones-bituminous coal with more than that of sawdust-coal briquettes [14]. This was partly expected from the chemical specification of the materials in Table 1 as compared with the pure coal firing. Generally, the presence of high content of silicon and aluminum in coal (approximately 17.74% SiO_2 and 10.86% Al_2O_3) suggests the existence of ashes with high fusion temperatures and, therefore, with less capacity to adhere to the surfaces [12,27]. Moreover, adding sawdust produced not only less ash but also easily removable deposits (ash) comparing with pure coal that was in good agreement with researches [12].

According to the overall results, the beet pulp briquettes are more favorable than sawdust briquettes, mainly due to their higher C.V. and lower volatile matter.

4. Conclusions

Fuel briquettes were prepared using coal and sawdust/ beet pulp, as main components. Their CS, WRI and chemical properties were determined. The results showed that the best composition and process condition to achieve high physical stability of sawdust briquettes would be sawdust to tar content ratio 15/8, water content 3%, calcium carbonate 2.5% and 25 tonnes briquetting pressure.

The effect of sawdust to tar content ratio was significant on WRI, but was insignificant on CS. The increase of briquetting pressure shows positive but insignificant effects on both WRI and CS.

The comparison of the sawdust samples with identical beet pulp briquettes showed that the presence of these natural binders decrease sulfur and residual ash of briquettes, while they make a negligible decrease in calorific value. By considering higher calorific value and lower volatile matter in the beet pulp briquettes, they can be considered more favorable than sawdust briquettes.

References

[1] Husain, Z., Zainac, Z. and Abdullah, Z.: Biomass and Bioenergy, 2002, 22(6): 505.
[2] Steel, K.M. and J.W. Patrick: Fuel, 2001. 80(14): 2019.
[3] Blesa, M.J., Miranda, J.L., Moliner, R,. Izquierdo, M.T., Palacios, J.M.: Journal of Analytical and Applied Pyrolysis, 2003. 70(2): 665.
[4] Massaro, M.M., S.F. Son, and L.J. Groven: Fuel, 2014. 115(0): 62.
[5] Demirbaş, A., Energy Conversion and Management, 2001, 42(11): 1357.
[6] Dumanli, A.G., I. Gulyurtlu, and Y. Yürüm, Renewable and Sustainable Energy Reviews, 2007, 11(9): 2058.
[7] Yılgın, M., Deveci Duranay, N. and Pehlivan, D.: 2010, 51(5): 1060.
[8] Chaiklangmuang, S., Supa, S., Kaewpet, P.: Chiang Mai Journal of Science, 2008, 35(1).
[9] Chin, O.C., Siddiqui, K.M.:Biomass and Bioenergy, 2000. 18(3): 223.
[10] Chou, C.-S., Lin, S.-H., Peng, C.-C., Lu, W.-C.: Fuel Processing Technology, 2009, 90(7–8): 1041.
[11] Blesa, M.J., Fierro, V., Miranda, J.L., Moliner, R., Palacios, J.M.: Fuel Processing Technology, 2001, 74(1): 1.
[12] Kupka, T., Zajac, K. and Weber, R.:Energy & Fuels, 2009. 23(7): 3429.
[13] Sarkar, P., Sahu, S.G., Mukherjee, A., Kumar, M., Adak, A.K., Chakraborty, N., Biswas, S.: Applied Thermal Engineering, 2014. 63(2): 616.
[14] Abreu, P., Casaca, C. and Costa, M.: Fuel, 2010, 89(12): 4040.
[15] Kuti, O.A.: AU J.T., 2009, 12(4): 284.
[16] Zarringhalam-Moghaddam, A., Gholipour-Zanjani, N., Dorosti, S.: Journal of Coal Science and Engineering (China), 2011, 17(4): 434.
[17] ASTM, Standard Test Method for Sulfur in Ash from Coal and Coke, in D-1757. 1996.
[18] ASTM, Standard Test Method for Volatile Matter in the Analysis Sample of Coal and Coke, in D3175 - 11 2007.

[19] ASTM, ASTM, Standard Test Method for Moisture in the Analysis Sample of Coal and Coke, in D-3173-03. 2007.
[20] Alam, H.G., Moghaddam, A.Z. and Omidkhah, M.R.: Fuel Processing Technology, 2009, 90(1) 1.
[21] Sun, H.-W., Liu, J.-Q., Chen, D., Gu, P.: Microelectronic Engineering, 2005, 82(2): 175.
[22] Yaman, S., SahanŞahan, M., Haykiri-Açma, H., Şeşen, K., Küçükbayrak, S.: Fuel Processing Technology, 2001, 72(1): 1.
[23] Marsh, R., Griffiths, A.J. and Williams K.P.: Fuel, 2008, 87(8–9): 1724.
[24] Taulbee, D., Patil, D.P., Honaker, R.Q., Parekh, B.K.: International Journal of Coal Preparation and Utilization, 2009, 29(1): 1.
[25] Morimoto, M., Nakagawa, H. and Miura, K.: Energy & Fuels, 2010. 24(5): 3060.
[26] Gholipour Zanjani, N., A. Zarringhalam Moghaddam, and S. Dorosti, Energy Sources, Part A: Recovery, Utilization, and Environmental Effects, 2013, 35(22): 2173.
[27] Dorosti, S., M.S. Thesis, Determination of Process Conditions and Formulation in Producing Coal Briquette from Iranian Antheracite, in Chemical Engineering. 2003, Tarbiat Modares University: Tehran.
[28] Abdollahy, M., Moghaddam, A.Z. and Rami, K.: Fuel, 2006, 85(7–8): 1117.
[29] Gani, A., Morishita, K., Nishikawa, K., Naruse, I.: Energy & Fuels, 2005, 19(4): 1652.
[30] Park, D.K., Kim, S.D., Lee, S.H., Lee, J.G.: Bioresource Technology, 2010, 101(15): 6151.
[31] Skeen, S.A., Kumfer, B.M. and Axelbaum, R.L. :Energy & Fuels, 2010, 24(8): 4144.
[32] Dong, L., Gao, S., Song, W., Li, J., Xu, G.: Energy & Fuels, 2008, 23(1): 224.
[33] ISIRI, I.N.S., Classification of Hard Coals by Type, in 1271

Corresponding author. A. Zarringhalam Moghaddam, *Department of Chemical Engineering, Tarbiat Modarres University, Tehran, Iran. Tel./fax: +98 21 88005040, E-mail addresses: zarrin@modares.ac.ir*