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Characteristics of residual oil fly ash and their utility for construction purposes

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Abstract

The present work investigates the physical and chemical characteristics of the residual oil fly ash (ROFA) generated as a by-product of heavy fuel oil burning in power plants. A possibility of re-utilization of the ROFA for construction purposes was estimated by activation of the ROFA with cementitious materials as a hardened paste matrix. The physical properties of the ROFA showed low bulk density, 0.21g/cm^3 and water content, 0.33%, as compared to previous studies. The particle size was less than 120μm. The chemical analysis showed that the ROFA is composed mostly of carbon (about 69%) and sulfur (7%) with residue ash. In addition, a significant amount of heavy metals, such as Sr, Ti, Zn, V, Cr and Ni were detected. The XRD data confirmed that the ROFA is mainly composed of amorphous carbon with metallic sulfates and metallic oxide materials. In order to investigate the changes of the hardened paste strength when added to the mixture, a reference mixture was developed. Also, the change of the density was estimated. The results indicated that the addition of 10, 20, and 30 wt% of ROFA shows a remarkable decrease in strength and density of the hardened paste. This study has found that ROFA has the potential to be re-utilized with cementitious material as lightweight material, or as a black pigment, thus providing beneficial changes to hardened paste color and density.

Keywords: Alkali activation; construction; residual oil fly ash; re-utilization.

1. Introduction

Residual oil fly ash (ROFA) is a by-product generated in power plants by the burning of heavy fuel oil. During combustion, the impurities fuse in suspension and float out of the combustion chamber with the exhaust gases as spherical particles in different sizes (Hwang *et al.,* 1996; Hernández *et al.*, 2011). The residual is collected from the exhaust gases by electrostatic precipitators or bag filters (Pires & Querol, 2004). The main constituent of ROFA is carbon. It also contains sulfates, silicates and nitrogen compounds, in addition to contaminants with significant concentrations of iron, vanadium, and nickel (Schroeder *et al.*, 1987; Al-Malack *et al.*, 2013; Kwon *et al.*, 2005; Mofarrah, 2014). Higher contents of metals occur in the heavy oils left after the more volatile fractions of petrol, paraffin, and diesel oil have been detailed. The chemical and physical characteristics of ROFA depend on the nature of the oil fuel (Andrew *et al.*, 2003).

The burning of one kiloliter of heavy fuel oil yields about three kilograms of fly ash (Tsai & Tsai, 1997). On average, 50-60 tonnes of fly ash are generated per day from a mid-range (i.e., 2300 MW power generating capacity) power plant (Hsieh & Tsai, 2003). ROFA is cheap and ready available in large quantities. It‘s density is light, and the particles are fine to very fine. Because of these characteristics, ROFA has the potential to travel a long distances before settling on land, water, or vegetation. Indeed, due to poor management and uncontrolled disposal, it may pose an environmental and public health hazard (Mohapatra & Rao, 2001). Yearly, over 500Mt of ROFA generated worldwide, but only a small portion is reused for productive purposes (Mõtlep *et al.*, 2010).

Researchers have carried out extensive characterizations of the chemical and physical properties of ROFA to determine potential uses (Kwon e*t al.*, 2005; Hsieh & Tsai, 2003; Mohapatra & Rao, 2001; Mõtlep *et al.*, 2010; Huffman *et al.*, 2000). Some recent studies investigated the recovery of heavy metals from ROFA. Heavy metals such as V, Ni and MO can be extracted from ROFA, which would be an effective solution to toxic metal contamination by power stations (Akita *et al.*, 1995; Vitolo *et al.*, 2001; Al-Ghouti *et al.*, 2011). Another application for ROFA is the preparation of activated carbon and using it as an adsorbent with low-cost and high pollutant-removal efficiency in wastewater treatment (Purnomo *et al.*, 2011; Ali, 2010; Danish *et al.*, 2013; Davini, 2002; Lu *et al.*, 2008). ROFA has been found to be a potential lightweight material for construction purposes since the fine ash particles have low density and high. porosity, and as a black pigment for cement mortar (Kwon e*t al.*, 2005; Camilleri *et al.,* 2013; Mofarrah & Husain, 2013; Lee & van Deventer, 2002). Recently, the stabilization potential of ROFA using an alkali activation method has been investigated to convert this by-product material into a low-cost construction material (Liu *et al.*, 2016; Jan *et al.*, 2016). Alkali activation is a chemical reaction where the solid inorganic aluminosilicate material activated by an activator such as NaOH or KOH, and sometimes with the combination of sodium silicate to form a new class of three-dimensional network alkali aluminosilicates (Davidovits, 1991). These materials are an attractive alternative to Portland cement as they have a comparable compressive strength, hardness, and chemical stability. In addition, less energy is consumed during production and less greenhouse gases are emitted (Xu & Van Deventer, 2000). Al-Degs e*t al.* (2014) studied ROFA stabilization using raw kaolinite as a binder material. The results showed a compressive strength reduction from 22 to 5 MPa with an addition of 15 to 41% ROFA. AL-Qahtani *et al.* (2014) and Al-shaaer *et al.* (2017) stabilized ROFA using treated kaolinite (metakaolin). The results showed a high compressive strength of 31MPs with the addition of 20 wt% ROFA along with high leachability for heavy metals and trace elements in neutral conditions.

In this work, an ROFA sample from power stations in Jordan has been characterized. In order to investigate the possible use of ROFA for construction purposes, a reference mixture of aluminosilicate material with pure filler was prepared. The filler was progressively reduced and replaced by adding ROFA in different mass ratios. Compressive strength and density changes were then estimated.

2. Characterization methods

ROFA sample was collected from the electrical power station at the Jordan Petroleum Refinery Company in Zarqa City, Central Jordan. The sample was collected directly from an electrostatic precipitator after the burning of heavy fuel oil. The sample was stored in an airtight container to prevent moisture absorption. The moisture content was calculated by measuring the weight in the oven at 105º C after drying (ASTM D4643-00). Its color was determined by visual observation. The bulk density and true density were measured by the standard testing methods (ASTM D6683-01 and ASTM D854-00, respectively). The particle size distribution was determined using a sieve analysis method according to ASTM D6913 / D6913M-17. A quantitative chemical analysis was performed to determine the concentration of major and trace elements in the ROFA sample by X-Ray Fluorescence (XRF)-1800, SHIMADZU Sequential X-ray Fluorescence Spectrometer. The Elemental Analyzer (An EA3000 Euro Vector, Italy) was used for carbon, sulfur, hydrogen and nitrogen analysis. The phase analysis for ROFA was carried out using a SHi-MADZU XRD-700 (MAXima-X) which was operated with Cu at 40 Kv of 2°/min. In addition, 30 mA was used for these measurements. A Netzsch Simultaneous Thermal Analyzer (Netzsch, STA 409 PC Japan) was used for DT-DSC analysis under atmospheric conditions. The temperature was increased from 5° C/min up to 550º C.

To investigate the possibility of stabilizing the ROFA for construction purposes, a reference mixture of aluminosilicate material with pure filler was prepared. The filler was progressively replaced with a mixture by 10, 20 and 30 wt% of ROFA. The prepared mixtures were reacted with an alkali activator, and the influence of using ROFA on the compressive strength and density was evaluated.

3. Materials and processing

3.1 Materials

A natural well crystallized kaolin was used as a source for silica and alumina (from the kaolin deposits in South Jordan) to develop a binder mixture. It consisted of 74% silica (SiO_2) and 26% aluminum oxide (Al_2O_3). The used grain size was less than 425µm. The kaolin used was fully characterized (mineralogical and chemical composition, thermal behavior, structural morphology and so on) in previous studies (Slaty *et al*., 2013). To improve the mixture's workability and mechanical strength, silica sand was used as inert filler (with purity about 98% and grain size between 90 and 250µm). The alkaline activator was KOH which was prepared by dissolving specific amounts of distilled water close to the plasticity limit of the mixture. The ROFA was gradually added into the cement paste to investigate the effect on the hardened paste strength.

3.2 Mixing process

The sample mass composition of the reference mixture was 100 kaolin, 50 silica sand, 16 KOH, and 22 H_2O. This composition was determined to give the best compressive strength. The raw materials (kaolin and silica sand) were weighed according to the given ratio and mixed for five minutes. Then the alkaline activator (KOH) was added. The mixture was mixed for 10 minutes and then poured into the mold and compacted a load pressure of 4KN. The molded samples were cured at 80º C for 48 hours and then taken to air dry at room temperature for seven days. The compressive strength was tested by a CBR device, and the density was calculated by measuring the sample mass and volume.

3.3 Adding ROFA

The ROFA was added to the cement mixture of 100 Kaolin, 16 KOH, and 22 H_2O at 10, 20, and 30 wt%, whereas the silica sand was progressively reduced from 50% (the reference sample) to 40, 30, and 20 wt%. The effect of the ROFA addition was estimated by measuring the compressive strength and density. The purpose was to investigate the effectiveness as an admixture in hardened construction cement. At least three samples were prepared from each mixture. The average compressive strengths and densities were obtained.

4. Results and discussion

4.1 Characterization of ROFA sample

The physical measurements of the ROFA, as found in the literature are indicating that, the color of ROFA is close to carbon black; however, it depends on the burning process and the characteristics of the heavy oil (Kwon *et al.*, 2005; Mofarrah, 2014). The bulk density of the ROFA varies from 0.20 to 1.50g/cm^3 and the true density is around 2.15g/cm^3. The particle size ranged from 10 to 120μm (Al-Malack *et al.*, 2013; Kwon *et al.*, 2005; Mofarrah, 2014). The studied sample was black, lightweight, and composed of fine particles. The moisture content was 0.33%. The bulk density (the mass of dry particles per unit bulk volume) was 0.37g/cm^3. The true density (the ratio of mass to true volume) was 1.08g/cm^3. The grain size distribution indicated that, 76% of the particles were ranged from less than 120μm up to 106μm and the other particles were below 45μm. In general, the particle size of fly ash up to 45μm it is more suitable as fillers in construction materials or as an adsorbent (Sarkar *et al.,* 2005).

The chemical analysis of ROFA, as given in Table 2, shows that the ROFA has a composition of about 69% C and 7% S. Other major inorganic substances were CaO, SiO_2, and Al_2O_3. Minor elements were Cr, Ni, Pb, Se, Sr, Ti, V, and Zn. Which are significantly elevated in the ROFA sample. According to research, the chemical composition of ROFA mainly depends on heavy oil characteristics and the burning environment.

Table 1. Physical measurements of ROFA

Bulk density (g/cm^3)	0.37
True density (g/cm^3)	1.08
Moisture content %	0.33
Particle size (μm)	< 120

The typical chemical composition of ROFA as found in the literature indicated that, carbon is the main composition of ROFA. The range of carbon composition is between 50 to 90% (Kwon *et al.*, 2005; Tsai & Tsai, 1997). In the addition of some compounds such as SiO_2, CaO, Fe_2O_3, and Al_2O_3. A valuable metallic compounds such as vanadium and nickel; and heavy metals generally exist in crude petroleum oil (Hwang *et al.,* 1996).

Figure 1 shows that ROFA sample had an amorphous nature in the X-ray with some distinct minerals and compounds. The diffractogram consists of sharp lines superposed on strong peak at 26° due to amorphous carbon. The other sharp lines represent inorganic compounds and have been identified as metallic sulfates and sulfides of Ca, Zn, V, and Ni. High sulfur content tends to produce higher levels of sulfates (Huffman *et al.*, 2000; Manivannan & Seehra, 2000; Babu e*t al.*, 1995). The metallic oxides were identified as SiO_2, which accords with previous studies (Mofarrah & Husain, 2013; Mofarrah, 2014; Huffman *et al.*, 2000; Babu e*t al.*, 1995).

The DT-DS analysis of the ROFA is shown in Figure 2. According to the TG curve, the weight begins to decrease at around 100º C and at a declination of about 17% at 540° C. This represents the degradation of the organic compound. The DSC data show an endothermic peak as the weight starts to decrease.

Table 2. Chemical composition of ROFA

Major oxides %	**Al_2O_3**	**CaO**	**Fe_2O_3**	**K_2O**	**MgO**	**Na_2O**	**P_2O_5**	**SiO_2**	**LOI**		
	3.34	4.72	1.49	0.40	0.82	0.10	3.12	2.09	4.98		
Organic elements %	**C**	**H**	**N**	**S**							
	69.16	1.15	1.19	7.32							
Trace elements ppm	**Ba**	**Co**	**Cr**	**Cu**	**Ni**	**Sc**	**Sr**	**Ti**	**V**	**Zn**	**Zr**
	101.3	4.8	320.2	32.0	190.0	92.0	1132.0	861.5	237.4	532.8	54.6

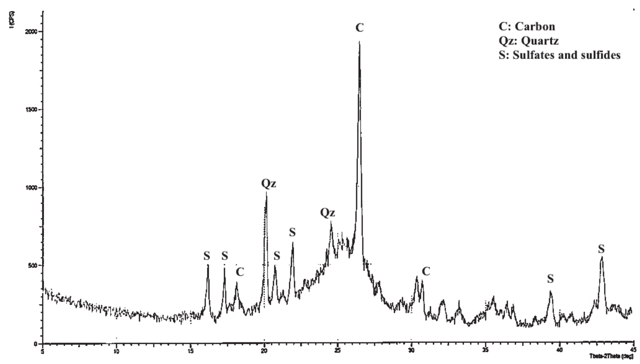

Fig. 1. XRD pattern of ROFA

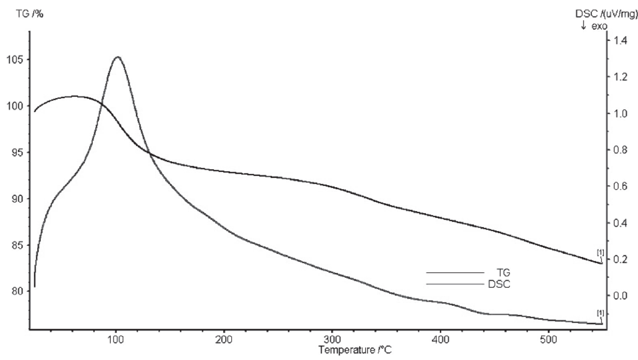

Fig. 2. TG/DSC trend of ROFA

The change of compressive strength by the addition of 10, 20, and 30% of ROFA is shown in Figure 3.

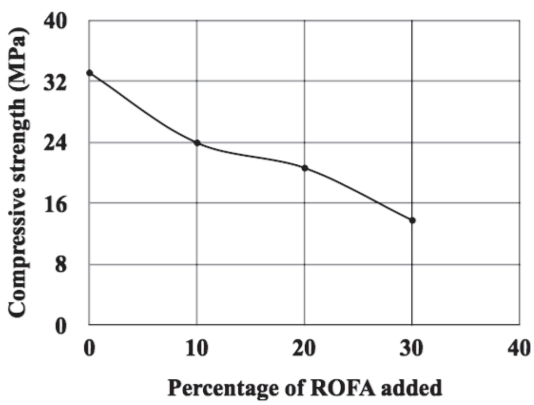

Fig. 3. Change of strength with addition of ROFA

The results show a remarkable decrease in compressive strength of cement mixture. Reference sample shows value of compressive strength about 33.06MPa. The gradual addition of 10, 20, and 30% of ROFA by replacing the weight ratio of silica sand from 50 to 40, 30, and 20%, decreases the compressive strength to 23.88, 20.57, and 13.70MPa, respectively. The change in sample strength is due to decreases in silica content. In addition, the used kaolinite was impure. The ROFA ash content could also dissolve by alkali solution. Density sample changes are shown in Figure 4. This figure demonstrates that the addition of ROFA causes a significant difference in density compared to the reference sample. This is due to the low density and high porosity of the fly ash particles. The results are in good agreement with previous studies (Al-Degs e*t al.*, 2014; Al-Qahtani *et al.,* 2014; Al-shaaer *et al.,* 2017). Although a good reduction was observed for samples containing ROFA compared to the reference sample, the product is still suitable for use in the application of lightweight composite material (Martina *et al.*, 2016).

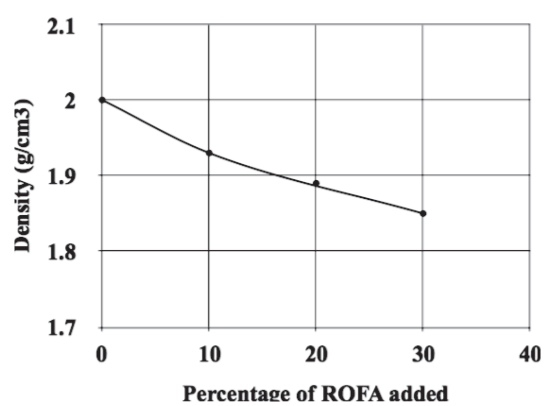

Fig. 4. Change of density with addition of ROFA

5. Conclusion

In this study the physical and chemical properties of ROFA were analyzed and a possible utilization of ROFA for construction purposes was investigated. The results showed that the sample ROFA was mainly composed of carbon, sulfur and residue ash. The particle size was less than 120µm, and XRD data confirm that it composed of amorphous carbon with some distinct minerals and compounds. When considering the utilization of ROFA for construction uses, this particular ROFA can change compressive strength and density. The results of the addition of 10–30% ROFA in a cementitious material mixture showed a significant decreasing of compressive strength due to decreasing in silica sand part as a filler.

The data show that the sample ROFA can be used in construction materials or a black pigment. It should not be used as a filler.

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خصائص الرماد المتطاير من حرق الزيوت الثقيلة وإمكانية الاستفادة منه في البناء

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خلاصة

يهدف هذا البحث إلى دراسة الخصائص الفيزيائية والكيميائية للرماد المتطاير الذي ينتج كمخلفات صناعية لحرق الزيوت الثقيلة في محطات توليد الطاقة الكهربائية. ولغايات البحث في إمكانية إعادة استخدام هذه المادة لأغراض البناء، فقد تمت مفاعلة هذه المادة مع تركيبة من المواد الأسمنتية للحصول على خليط قوي ومتماسك. وقد دلت القراءات الفيزيائية لمادة الرماد المتطاير عندما قورنت بدراسات سابقة، على إنها ذات كثافة منخفضة (٠،٢غ/ سم٣) وبمحتوى رطوبي قليل (٠،٣٣٪). و حجم حبيبات المادة أقل من ١٢٠ ميكروميتر. أما التحاليل الكيمائية فقد أشارت إلى إحتواء هذه المادة ٦٩٪ من عنصر الكربون و ٧٪ من عنصر الكبريت. بالإضافة إلى وجود تراكيز من العناصر الثقيلة مثل Sr, Ti, Zn, V, Cr, Ni والتي تعتبر إلى حد ما مرتفعة إذا ما قورنت بدراسات سابقة. وبالنسبة لدراسة التركيب المعدني للرماد لمتطاير فقد أشارت إلى وجود مادة الكربون الغير متبلورة ، إضافة إلى معادن مركبات الكبريت والأكاسيد. ولدراسة تأثير إضافة مادة الرماد المتطاير إلى تركيبة من المواد الأسمنية والمستخدمة في التطبيقات البنائية، فقد تم قياس التغيير الحاصل في قوة وكثافة الخليط بعد إضافة نسب مختلفة من هذه المادة (١٠٪ و ٢٠٪ و ٣٠٪). وقد أشارت النتائج إلى حدوث إنخفاض تدريجي في قوة وكثافة العينات مع إزدياد نسبة هذه المادة فيها. وقد أوصت الدراسة بأن امكانية استخدام هذه المادة مع خليط من المواد الاسمنتية كمادة مالئة يعتبر غير مجدي لأنه يقلل من قوة مادة البناء إلا أنها من الممكن الأستفادة منها لتصنيع مواد البناء ذات الوزن الخفيف وكذلك كصبغة سوداء لتغيير مظهر مواد بناء لأغراض الزينة.