
CHARACTERIZATION OF VARIOUS KINDS OF REFUSED DERIVED FUEL AND THEIR EFFECTS ON CEMENT PROPERTIES

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ABSTRACT

Recently, there is an increasing demand for using refused derived fuels (RDF) as replacement for traditional fossil fuel in energy intensive industries like cement industry. However, using such sources will affect the properties and quality of the produced cement since RDF ash was mixed with OPC clinker during production. In the present study, four solid refused derived fuels namely; rubber waste, tree trimmings, rice straw and municipal solid wastes MSW are characterized and tested as RDF. Also, the effect of their ash on the setting and hardening of the hardened OPC pastes was investigated. Results showed that rubber waste has the highest calorific values followed by MSW. OPC pastes mixed with RDF ash showed reduced setting times. Besides, OPC admixed with rubber wastes ash offered the highest compressive strength values between all the studied RDF sources. Phase composition of the formed hydrates is studied by X-ray diffraction analysis and results showed that the ash within OPC matrix does not change the hydration mechanisms since the same hydration products are formed.

Keywords: Portland cement, Reused drive fuels, rubber Waste, MSW, Energy.

INTRODUCTION

Cement industry has development greatly in the last decades. The aims for such development are resource conservation and environmental protection (Madloul *et al.*, 2011). Cement industry is one of highly energy-intensive

industry with an energy typically accounting for 30-40 percent of production costs. In traditional cement kilns, different sources of fuels are used such as natural gas, coal and petroleum oil (Singhi and Bhargava, 2010). The fuel mix in the cement kiln is carbon intensive, and the calcination process produce CO₂ gas, so that in total the cement industry contributes by about 5 % of the global CO₂ emissions (Martin *et al.*, 1990 and Laszlo *et al.*, 2006). In cement plant, 60 % of the CO₂ emitted results from the calcination of limestone, 30 % from combustion of fuels in the kiln and 10 per cent from other downstream plant operations (Bosoaga *et al.*, 2009). Refuse Derived Fuel (RDF) is an expression appeared recently in many countries which refers to fuel produced from incineration of solid wastes (Alter *et al.*, 1974) or to obtain energy from waste materials. Waste derived fuels include residues from municipal solid waste (MSW), industrial/ trade waste, sewage sludge, industrial hazardous waste and biomass waste. Most of these wastes are primarily composed of carbon-based organic matter, which releases energy when it reacts, or combusts with oxygen (Seboka *et al.*, 2009). For these, production of RDF does not only eliminate wastes and find a solution to their accumulation but it will also generate energy (Schwarzböck *et al.*, 2016a). Refused derived fuels RDF are increasingly utilized in energy intensive industries such as cement manufacture and steel production (Aldrian *et al.*, 2016). Thereby, RDF could substitute conventional fossil fuels to a large extent since it represents cost saving and usually goes with lower CO₂ emission (Choy *et al.*, 2004 and Marland *et al.*, 2008). In 2006, many European countries already more than 70% of the overall energy consumption

of cement manufactures is covered by RDF (Garg *et al.*, 2009).

The main parameters on which solid waste could be chosen as RDF are its higher heating value (energy input), ash residue and the toxicity of the gas evolved. Some researchers have focused on emissions and pollution due to waste derived fuel combustion in cement kilns (Karstensen, 2008) and Zemba *et al.*, 2011). Impacts on the product quality have also been investigated by some authors. When burning RDF, or any kind of waste, the two main byproducts produced are gases and ash. During the burning process in the incinerator many of the pollutants from the exhaust gases could be removed and concentrated in the ash (Guo and Eckert, 1996 and Chatziaras *et al.*, 2014). Consequently, the ash represents a cocktail of toxins; all the pollutants captured at various stages end up here. There are two types of ash: heavy bottom ash, which comes from the furnace where the waste is burnt and light fly ash, which comes from the air pollution control equipment. Numerous studies have confirmed that a typical waste incinerator releases dioxins, lead, cadmium, mercury, and fine particles into the atmosphere (Karstensen, 2008 and Zemba *et al.*, 2011). According to the United States Environmental Protection Agency, municipal waste incinerators are one of the top four sources of dioxin and furan emissions in the USA. Heavy metals are also a major source of environmental concern (Guo and Eckert, 1996). Since these toxins are not destroyed during incineration, they end up in the incinerator ash, or they escape into the environment through the smoke stack and are transported through the air and deposited in water and soil. Recently many countries tend to use RDF as a source of energy in cement industry instead of fossil fuel or as a fraction of the fuel used in the production of cement clinker

(Kaantee *et al.*, 2004). Industry argues that the use of waste as alternative fuels has several positive impacts on sustainability, i.e. conservation of non-renewable energy sources, better energy recovery in waste, etc. In addition, the use of alternative fuels can help to reduce the costs of cement production. However this will have a large impact on the quality of clinker produced. Many reports showed that there is a rise in Cl-, Pb, Cd, Cu and Zn levels by about 2 to 5 % higher than the clinker produced without using RDF (Gendebien, *et al.*, 2003). Chlorine and phosphate content in waste are important as these can affect cement quality.

The present work aims evaluating four types of solid wastes as an alternative RDF sources. This is done via studying the caloric value and ash residues of these wastes. Beside the effect of their ash residue on the properties of Ordinary Portland cement clinker and the hydration characteristics of the produced hardened OPC pastes are also investigated.

MATERIALS AND EXPERIMENTAL

Materials: Industrial clinker supplied from Helwan Company for cement production, Helwan, Egypt. It is used for the production of ordinary type I Portland cement (OPC-I, 42.5N). Four wastes include Tree trimmings (TT), Rubber waste (RW), Rice straw (RS) and municipal solid waste (MSW) are collected from local areas and used as an example of RDF. Solid wastes are burned at 1000°C for three hours and their ashes are collected and used as additive to OPC clinker.

Preparation of Dry Mixes: During the grinding of OPC clinker, the ash residues obtained from burning of various RDF wastes under investigation was added to OPC clinker with two percentage composition of 5 and 10 % from the mass of OPC clinker. A constant 5% of gypsum by weight of clinker has been added to each cement mixture.

Specimen Preparation; Curing and Testing: Different cement pastes were prepared using water/solid (W/S) ratio = 0.3. Each paste was prepared by mixing the dry mix with the required amount of water for about three minutes. After complete mixing, the resultant paste was molded into cubic specimens by using one inch cube molds. The molds, containing the pastes, were cured under 100% relative humidity for 24 hours to attain the final setting, and then the cubic specimens were removed from the molds and cured under water for time intervals of 3, 7 and 28 days.

Compressive Strength: At each time interval, the compressive strength test was performed on three cubes represented each hardened cement paste and the average value was recorded. The hydration of the hardened cement pastes was stopped using the method described in earlier publication (Abo-El-Enein *et al.*, 1972). The samples were then dried at 100°C for three hours in CO₂-free atmosphere and conserved in a desiccator containing soda lime and CaCl₂ till using for testing purposes.

Hydration Kinetics: Hydration kinetics was studied by the determination of setting times; chemically combined water and free lime contents at different ages of hydration.

Setting Time: Setting times determined Using VICAT device .after mixing the cement with the required amount of water which was tested by the penetration of the device plunger in the paste till 4 mm then after suitable time testing the penetration of the needle in the paste till 4 mm and at that time recording the initial setting time .after some times starting testing with final setting time needle till only marking the paste surface not penetrate it.

Chemically Combined Water: Chemically combined water contents (W_n , %) were determined as follow (Kondo *et al.*, 1975) :

Two representative samples of the dried specimens, exact about 1g each, were weighted in porcelain crucibles and ignited for one hour at 1000°C in an adjustable muffle furnace, cooled in a desiccator and then weighted. The chemically-combined water content (i.e., the amount of water retained after drying) was calculated as W_n (%) using the following equation:

$$W_n (\%) = [(W_1 - W_2) / W_2] \times 100$$

W_1 : is the weight of the dried sample before ignition (g) and

W_2 : is the ignited weight of sample (g).

Free Lime Content (Cao): The free CaO content was determined by the following method (Abo-El-Enein *et al.*, 1974):The sample (exact about 0.5 g) was dispersed in 40 ml of glycerol/ethanol mixture (1: 5) by volume; together with small amount of anhydrous barium chloride (about 0.5g) as catalyst, and phenol phthalein as indicator. This mixture was kept in a conical flask, fitted with an air – reflux, on a hot plate for 30 minutes (the color becomes pink). The contents were titrated with a standardized alcoholic ammonium acetate solution until the pink color was disappeared. Heat again, if the pink color

reappears, complete the titration with ammonium acetate solution until no further appearance of pink color takes place.

Phase Comostion: The phase composition of the formed hydrates was investigated using X-ray diffraction analysis using a cobalt target ($\lambda= 0.17889$ nm) and a nickel filter under working conditions of 40 kV and 30 mA.

RESULTS AND DISCUSSION

Table (1): The chemical oxide composition of OPC clinker and the four RDF ashes-clinker mix

oxide	Clinker	TT10%	RW 10%	RS 10%	MSW 10%
SiO ₂	21.076	23.33	19.53	21.99	24.43
Al ₂ O ₃	5.21	5.66	4.86	5.01	5.01
Fe ₂ O ₃	3.43	3.83	3.21	3.2	3.73
CaO	67.15	59.39	64.03	61	58.66
MgO	1.96	1.9	1.76	1.93	1.91
K ₂ O	0.04	0.33	0.06	0.5	0.17
SO ₃	0.31	2	3.05	2.53	2.19
Na ₂ O	0.36	0.33	0.35	0.46	0.4
LOI	0.34	2.35	2.8	2.36	2.74

Characterization of RDF And RDF ASH: Characterization of RDF and their residues after burning are important parameters to predict the suitability and toxicity of used wastes as RDF sources. In other word, determine their efficiency as fuel and if those sources are/or are not environmental friendly. Beside determination of the percentage of the remained ash for these wastes show how the produces OPC could be affected by these new fuel sources.

Table (2) shows CHN percent, Chloride and sulfur contents, the ash percentage, caloric values and the densities of the solid wastes residues remained after burning of various RDF sources at 1000°C for 2 hrs.

According to the listed data, the main compositions of the studied wastes are hydrocarbons with minor amine, sulfur and chloride contents. The percentage of Rubber waste (RW) and Tree trimmings (TT) offer the highest CHN and sulfur and chloride contents. This could predict the highest carbon, sulfur and nitrogen oxides emission by using these wastes as source for RDF. After firing, Rubber waste (RW) offered the highest caloric value (CV) (8600 Cal/g). While as, mixed trash (MT) showed about half the caloric value offered by (RW). Both Tree trimmings (TT) and Rice straw (RS) showed comparable caloric values (3174 and 3167 cal/g respectively) and lower densities. All the solid wastes show high ash percentage after their firing at 1000°C for 2 hrs which varies from 19.2 to 27.7 %.

Table (2): Characterization of raw RDF .

Solid residue	%C	%H	%N	%S	%Cl	% Ash	Density (g/cm ³)	Caloric Value (Cal/g)
TT	46.13	4.84	4.91	3.3	0.07	27.7	0.69	3174
RW	56.43	8.23	6.27	6.1	1.3	19.2	1.1	8600
RS	36.99	5.25	4.35	0.3	0.03	21.3	0.35	3167
MSW	26.29	4.33	3.55	0.52	0.02	22.9	0.4	4200

Setting Times: Fig. (1a-d) shows the initial and final setting times for Portland cement pastes admixed with 5 and 10 wt. % of RDF ash of various origin. All the OPC pastes admixed with 5 wt. % of RDF ash showed a reduction of both initial and final setting times compared to OPC paste free of ash, mix B. These reduction of setting times were also observed for OPC samples admixed with 10 wt % of RDF ash except those admixed with 10 wt.% of tree trimmings (TT) ash which showed a longer setting times. Short

setting times means high rates of stiffening and hardening of admixed OPC pastes. It is well established that the setting of cement is a percolation process in which isolated or weakly bound particles are connected together by the formation of hydration products when mixed with water (Bentz, 2008). The higher rate of hydration could be related to two effects occurred due to presence of RDF ash. The first is their filler effects due to their greatest fineness and low densities. Besides, the nucleation effect of the RDF ash particles, so they can act as active centers inside OPC matrix that favors the growth of the hydration products on them leading to acceleration of the hydration reaction (Marzouki, *et al.*, 2013).

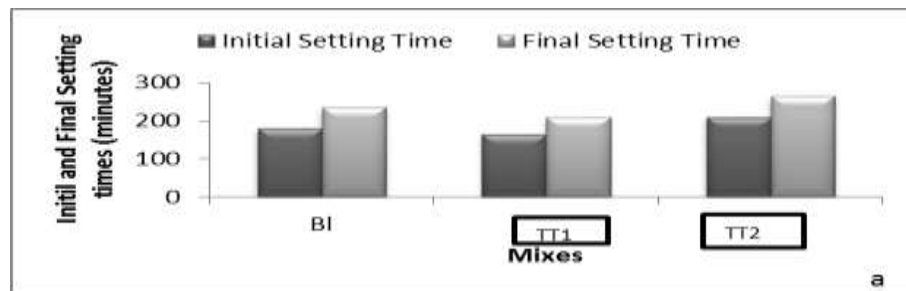


Fig. (1): Initial and final setting times of OPC pastes admixed with RDF ashes, a) tree trimmings; TT1 5% ash TT2 10% ash

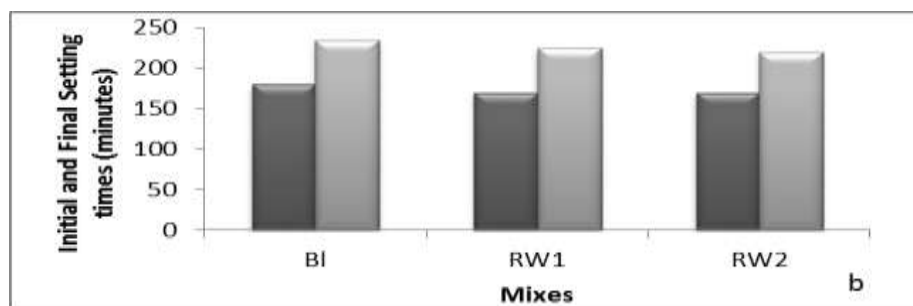


Fig. (2): Initial and final setting times of OPC pastes admixed with RDF ashes b) rubber wastes; RW1 5% ash RW2 10%

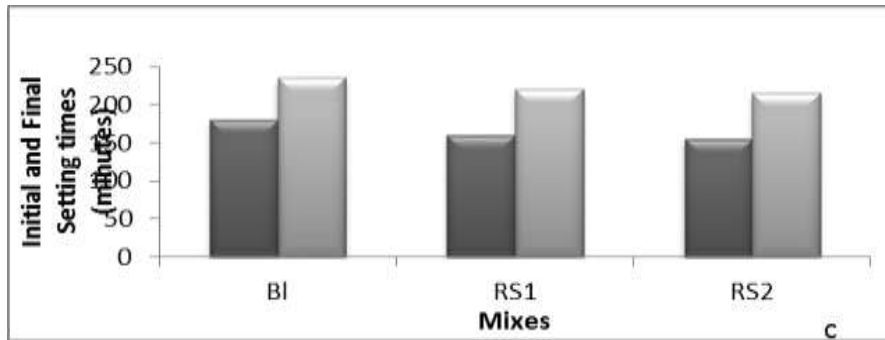


Fig.(3): Initial and final setting times of OPC pastes admixed with RDF ashes
c) rice straw; RS1 5% ash RS2 10%

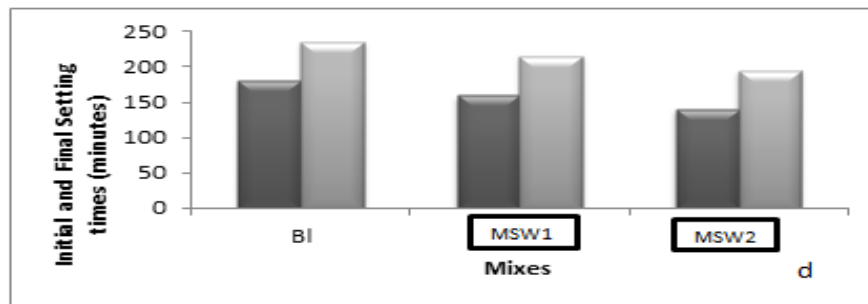


Fig.(4): Initial and final setting times of OPC pastes admixed with RDF
ashes, d) municipal solid waste; MSW1 5% ash MSW2 10%

COPRESSIVE STRENGTH: The variation of the compressive strength values for OPC and OPC pastes admixed with 5 and 10% of RDF ash with the hydration ages are shown in Fig. (2 a-d). The control OPC paste, B mix, shows three stages of strength development with increasing the hydration ages up to 90 days. A rapid increase in the compressive strength values is noticed during the first 24 hours of hydration which represent the acceleration stage. This is followed by a continuous and gradual increase up to 28 days which represent the dormant stage. A slow increase in the compressive

strength values noticed during the hydration period from 28 to 90 days which represent the state of equilibrium is reached.

OPC specimens admixed with RDF ash with different origin, showed the same trend of variation of compressive strength values with the hydration ages. However there is a variation in the compressive strength values compared to OPC pastes depending on the hydration period and the origin of RDF ash and its percent in the OPC paste. For OPC pastes admixed with rubber wastes (5 & 10 % RW) there is an improvement in the compressive strength values at all hydration ages compared to the control OPC paste (B mix) .In fact these mixes (RW1 and RW2) offered the highest compressive strength values between all the studied RDF sources. Such results are in agreement with the works that using of crushed rubber could improve the OPC and concrete properties (Girskas *et al.*, 2017 and Bisht and Ramana, 2017).

For OPC pastes admixed with other RDF ash (mixes TT, RS and MSW) show higher compressive strength values than blank (neat OPC, mix B) during the early ages of hydration; up to 3 days and sometimes this extended to 7 days as for ash derived from tree trimmings (TT mixes). This confirms that the presences of RDF ash within OPC pastes improve the early hydration reaction as indicated from the setting time results. At later ages of hydration, after 28 and 90 days, the ash admixed OPC pastes show relatively lower or comparable values of the compressive strength to those offered by OPC pastes. Such results are in agreement with those of Katare and Madurwar who used RDF of bio-origin. (Katare and Madurwar, 2017 and Kaminskas *et al.*, 2015).

OPC pastes admixed with Tree trimmings ash (TT) showed an improvement in the compressive strength values by increasing their percentage in OPC pastes from 5 wt.% to 10 wt.%. Whiles as both rice straw (RS) and mixed municipal solid waste (MSW) ash showed reverse results.

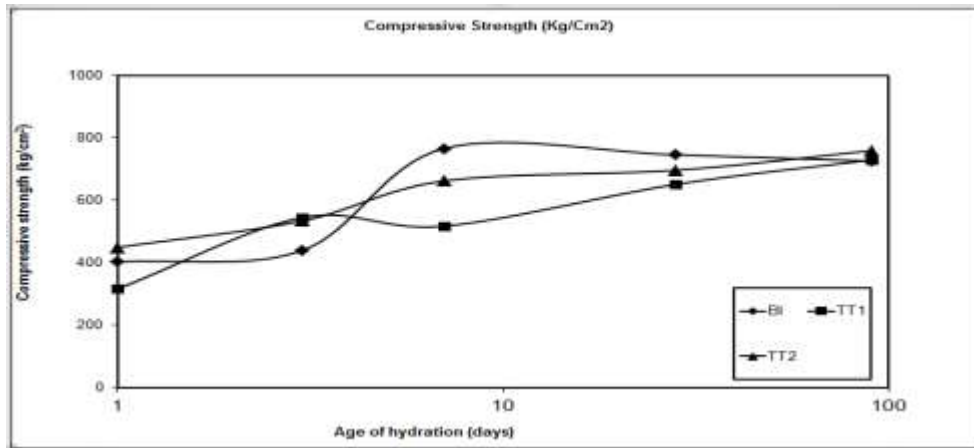


Fig.(2-1): Compressive strength of OPC pastes admixed with RD ashes at various hydration ages; a) tree trimmings; TT1 5% ash TT2 10% ash

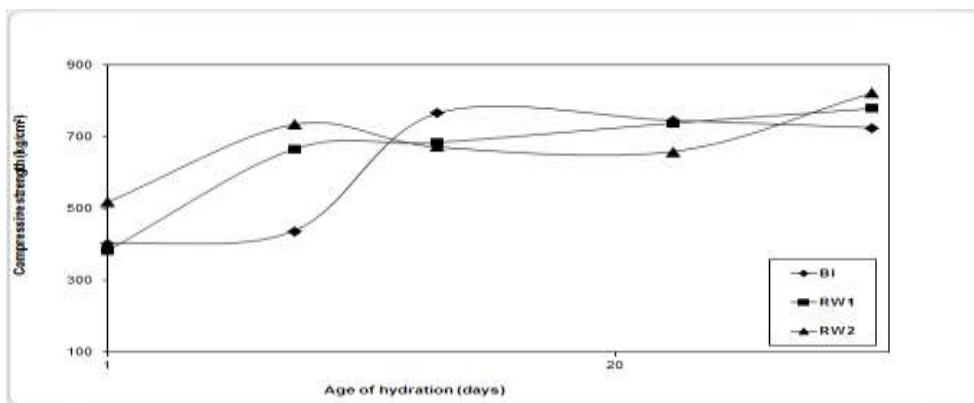


Fig.(2-2): Compressive strength of OPC pastes admixed with RD ashes at various hydration ages; b) rubber wastes; RW1 5% ash RW2 10%

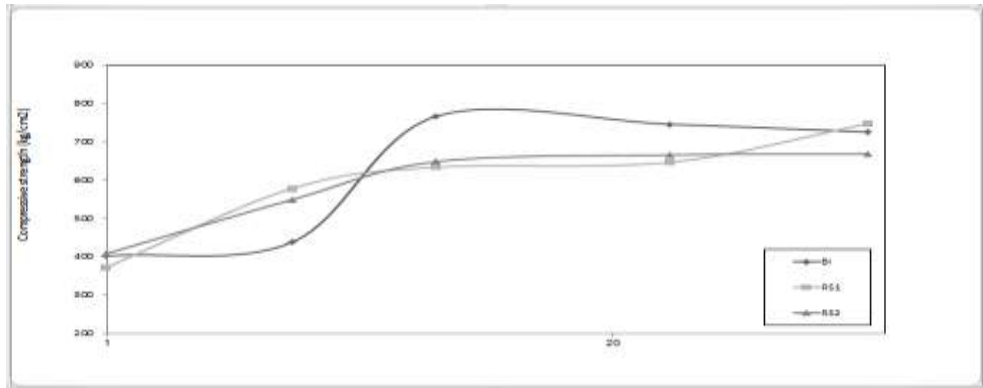


Fig.(2-3): Compressive strength of OPC pastes admixed with RD ashes at various hydration ages; c) rice straw; RS1 5% ash RS2 10%

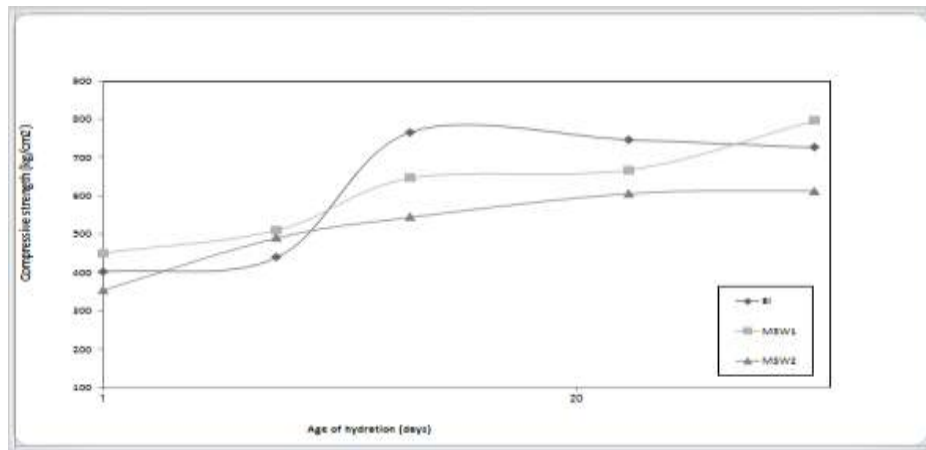


Fig.(2-4): Compressive strength of OPC pastes admixed with RD ashes at various hydration ages d) municipal solid waste; MSW1 5% ash MSW2 10%

HYDRATION KINETICS: Hydration kinetics of the investigated pastes was carried out by determining the free lime (CaO, %) and chemically combined water (W_n, %) contents at different ages of hydration.

COMBINED WATER (W_n %): The results of non-evaporable (chemically combined) water content, (W_n , %), obtained for specimens made from neat OPC and those admixed with 5 and 10 wt.% of RDF ash (mixes TT, RW, RS and MSW) are represented graphically as a function of hydration age in Figs.(3, 4) respectively.

The chemically combined water contents of the hardened specimens made of neat OPC showed three stages of variation of W_n -values with increasing hydration age. An initial stage of rapid increase in W_n values; up to 3 days, which show a fast rate of hydration reaction between water and unhydrated OPC phases ;(El-Gamal *et al.*, (2012)). This was followed by a slow stage of increase of W_n value during the period of 3-28 days; this represent the "dormant period" (Morsy *et al.*, (1998)). This was followed by an "acceleration stage" with a gradual and continuous increase in the chemically combined water contents with increasing hydration ages up to 90 days. This increase is attributed to the formation and later accumulation of hydration products mainly as CSH.

Obviously, all the OPC mixes admixed with RDF ash showed lower W_n , % values than the neat OPC mix at all the hydration ages. However such results are in contrary with the setting times and compressive strength results since the admixed mixed showed a reduced setting times and higher strength than the neat OPC. Such finding could be related to the formation of hydration products in OPC-ash mixes with low water contents and improved strength properties (Abo-El-Enein, *et al.*, 2016).

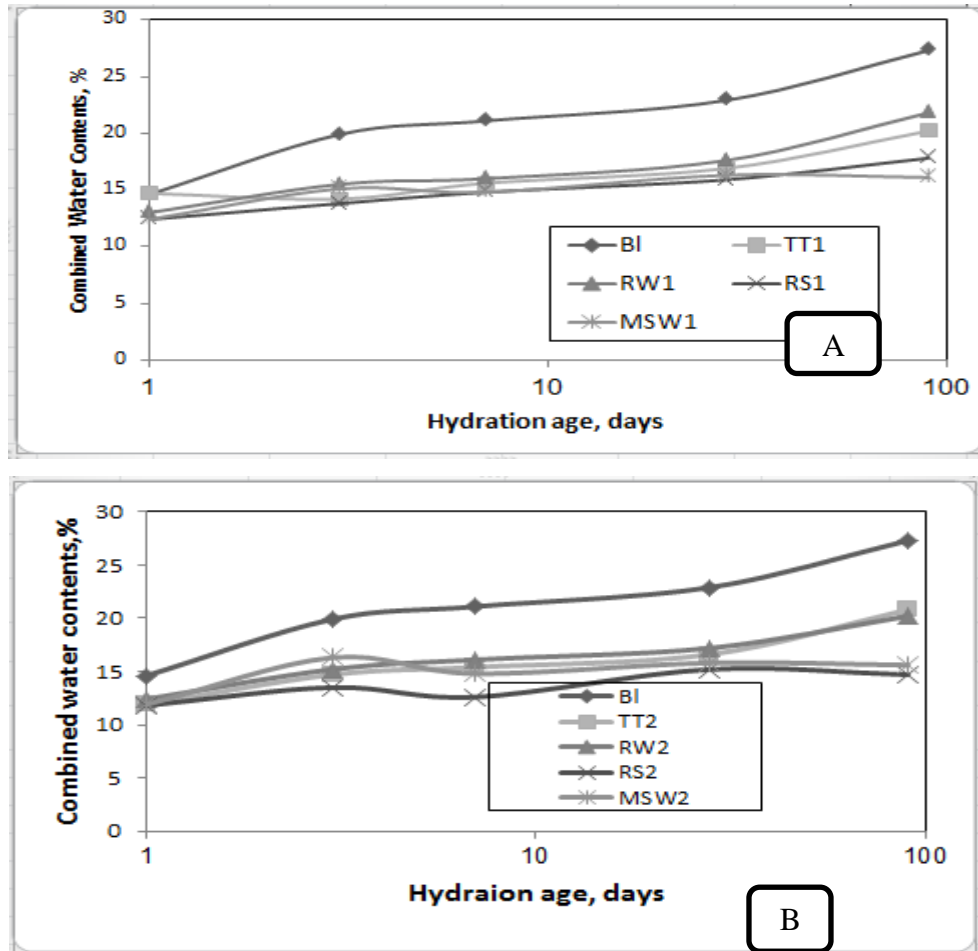


Fig.(5): Combined water contents (%) o OPC pastes admixed with RD ashes
 a) 5 wt. % b) 10 wt. %

TT1 5% ash TT2 10% ash RW1 5% ash RW2 10% RS1 5% ash
 RS2 10% MSW1 5% ash MSW2 10%

FREE LIME (CaO): Fig.(4 -a,b) showed the results of the variation in free lime content (CaO,%) with age of hydration of the hardened neat OPC pastes, B, and those made of admixing OPC with RDF ash. Obviously, B mix

showed a gradual and continues increase in free lime contents up to 90 days indicating the progress of the hydration reaction and accumulation of Ca (OH)₂ which is one of the hydration products of OPC pastes beside calcium silicate hydrates CSH. For OPC- ash mixes, low values of CaO% are recorded in case of admixed mixes compared to B mix at all ages of hydration. Beside, increasing the percentage of RDF ash in OPC mixes from 5 to 10 wt.% results to more decrease the CaO % values. Besides, there are variations in the trend of increasing CaO % with the ages of hydration in these admixed mixes. At early ages of hydration, up to 3 days, CaO % values increased with the time of hydration. However at later ages of hydration, up to 28 days, CaO% values may be decreased or remained unchanged while at 90 days increased. This variation in CaO % values could be attributed to pozzolanic properties of RDF ash and their presence inside OPC matrix leads to their reaction with Ca (OH)₂ and formation of additional amounts of calcium silicate hydrates, CSH. Such additional CSH precipitate inside the pore system of OPC pastes leading to lower porosity and improve mechanical properties.

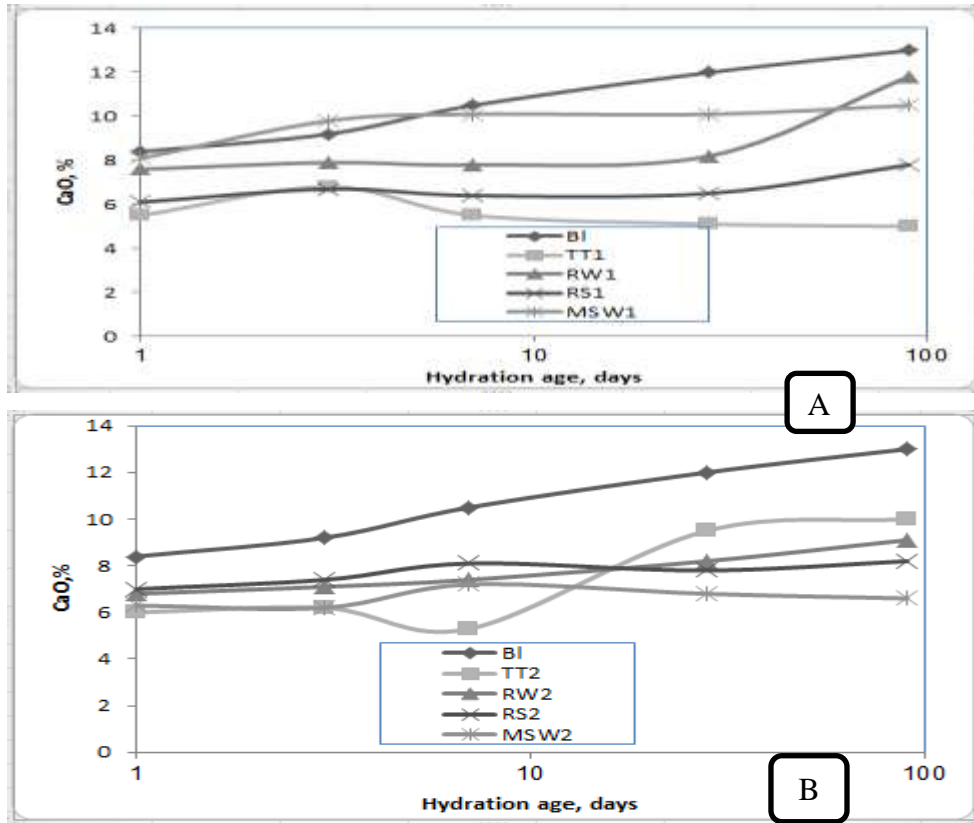


Fig.(6): Free lime contents (%) o OPC pastes admixed with RD ashes a) 5 wt. % b) 10 wt.

%TT1 5% ash TT2 10% ash RW1 5% ash RW2 10% RS1 5% ash RS2 10% MSW1 5% ash MSW2 10%

PHASE COMPOSTION: X-ray diffraction patterns of mixes B, TR2, RS2, RW2 and MT2 after 3 days of hydration are shown in Fig.5. XRD of blank mix, B, displays peaks which they are characterized to hydration products of OPC mainly; $\text{Ca}(\text{OH})_2$ (CH) and Calcium silicate hydrates (CSH). The appearance o these peaks indicates the onset of the hydration reaction. Peaks

due to unhydrated phases, C_2S , C_3S and gypsum, are also identified in XRD of mix B. The persistence of peaks due to C_2S and C_3S after 3 days of hydration is due to incomplete hydration reaction (El-Gamal, and Hashem, (2017). OPC pastes admixed with RDF ashes show the same hydration products as blank mix indicating that mixing of Portland cement with RDF ashes does not change the hydration phases. However the acceleration effect due to admixing of OPC with RDF ashes is clearly observed in XRD patterns especially for mixes containing rubber ash (sample RW1) and municipal solid wastes (sample MSW1). There are distinct increases in intensity of peaks due to CH and CSH are noted accompanying the prevailing of peaks due to C_2S and C_3S with lower intensity than the case of mix B.

After 28 days of the hydration reaction, XRD patterns of the blank mix show an increase in the intensities of the peaks characterized to the hydration products, CH and CSH, see Fig 6, this accompanied with a notable decrease in the intensities of reactants peaks. For OPC-ash mixes, again, no different hydration products are identified in their XRD patterns. However, the acceleration effect due to incorporated of RDF ashes in the hydrated matrix of OPC is nearly recovered as indicated by the similarity in the intensities of the peaks due to CH and CSH and those of the unhydrated phases as those of mix B.

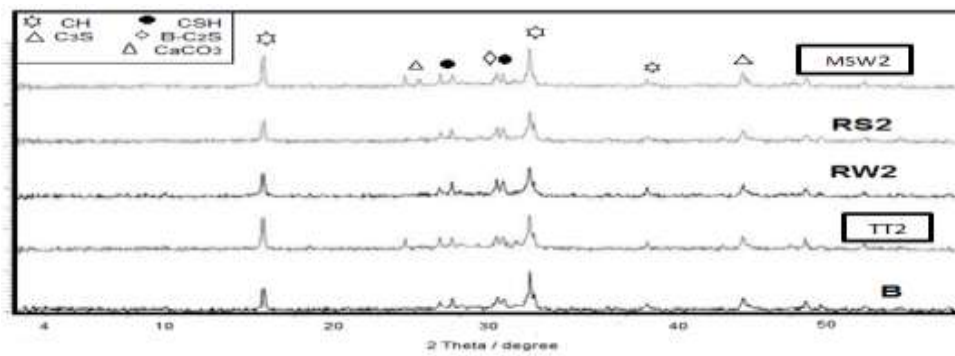


Fig.(7): XRD patterns of various mixes after 3 days of hydration

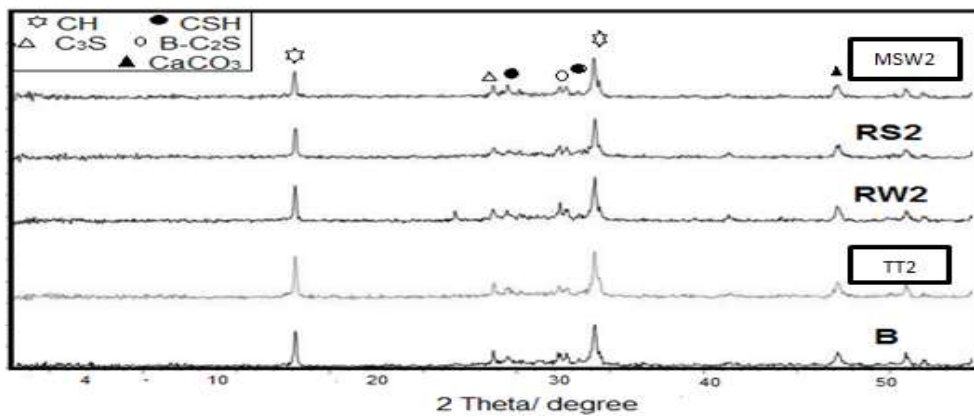


Fig. (8): XRD patterns of various mixes after 28 days of hydration. TT1 5% ash TT2 10% ash RW1 5% ash RW2 10% RS1 5% ash RS2 10% MSW1 5% ash MSW2 10%

CONCLUSION

On the basis of our studies the following conclusions could be derived:

- Rubber waste, tree terminals, rice straw and mixed trash can be used as a source of RDF fuel for cement industry to replace the traditional fossil fuel.
- Rubber waste has the highest caloric value and density among the studied RDF sources.
- Mixing of the ash residue of these RDF sources with the cement clinker during manufacture lead to reduce the setting times and accelerate the early hydration reaction of the resultant cement.
- Mixing of OPC clinker with rubber waste ash increase the obtained compressive strength of OPC.
- Presence of RDF ash does not change the hydration mechanism of the produced cement.

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توصيف أنواع مختلفه من الوقود البديل وتأثيرها على خواص الاسمنت

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المستخلص

ازداد مؤخرًا الطلب على استخدام الوقود المرفوض كبديل عن الوقود الاحفوري التقليدي في الصناعات كثيفه الطاقه مثل صناعه الاسمنت لكن استخدام هذا الوقود سوف يؤثر على خصائص وجوده الاسمنت المنتج وذلك مع خلط الرماد الناتج من حرق ذلك الوقود مع الكلنكر أثناء الانتاج. في هذه الدراسه أربعة أنواع من هذا الوقود وهي المخلفات الجلديه؛ قصاصات الأشجار؛ قش الارز والمخلفات البلديه الصلبه تم توصيفها واختبارها كوقود مرفوض. وأيضاً فحص تأثير الرماد المتبقي في زمن الشك وتصلب معجون الاسمنت البورتلاندي العادي. أظهرت النتائج أن المخلفات الجلديه لها أعلى سعرات حراريه ثم بعد ذلك المخلفات البلديه الصلبه. أظهرت عجائن الاسمنت البورتلاندي العادي المخلوط مع الرماد المتبقي من الوقود المرفوض

انخفاض زمن الشك، كما ظهر أن الأسمنت البورتلاندي العادي المخلوط مع الرماد المتبقي من المخلفات الجديه أعطى أعلى قوة تحمل للضغط بين كل أنواع الوقود محل الدراسه. تم عمل دراسه لكل الأطوار من المواد المتهدرته بإستخدام تحليل حيود الأشعه السينيه وأظهرت النتائج أن خلط الأسمنت البورتلاندي العادي مع الرماد المتبقي لم يغير ميكانيكيه الهيدره حيث تكونت نفس نواتج الهيدره.

كلمات مفتاحيه: الأسمنت البورتلاندي العادي؛ الوقود المتبقي؛ المخلفات الجديه؛ المخلفات البلديه الصلبه؛الطاقه.