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Solidification of radio contaminated natural materials into ordinary Portland cement

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Abstract

This paper presents the result of a study on the use of natural materials(saw dust, rice husk, and sugar cane bagasse) for removing of some radionuclides from liquid radioactive waste. Contaminated natural materials used are saw dust, rice husk and bagasse which were used as untreated species and treated species (treated by KOH, HCl). These natural materials were solidified into ordinary Portland cement.Compressive strength of the final solid waste blocks has been performed. Effect of temperature, different additives types (with and without treatment) and different additives weights on the compressive strength has been carried out. At 50°C the compressive strength for samples contain untreated saw dust, rice husk and bagasse was 76, 25 and 45KN respectively, while at 200 °C compressive strength was 120, 20 and 40KN respectively.By increasing temperature from(50 to 200°C), compressive strength for solidified samples contain saw dust treated by HCl and KOH decreases from 119 to 103 KN and from 88 to 49 KN respectively. For samples contain sawdust without treatmentcompressive strength increases by increasing temperature. Compressive strength for samples contain rice husk treated by HCl increases from 77 to 99 KN as temperature increase from 50 to 200°C. For samples contain KOH treated rice husk compressive strength decrease from 65 to 30 KN as temperature increase from 50 to 200°C. For samples contain bagasse treated byHCl, KOH, compressive strength decrease as temperature increase. For samples contain untreated bagasse compressive strength increases from 45 to 64 KN as temperature increase from (50 to 200°C). The experimental results show that, for samples contain KOH treated saw dust compressive strength increases for 1gm of additives, but by increasing amount of additives (2g,3g), the compressive strength decreases. Samples contain KOH treated rice husk, the compressive strength increases by increasing amount of additives, while decreases with bagasse samples. For samples containHCl treated additives they all show decrease in compressive strength by increasing amount of additives. For samples contain untreated additives, they show increase in compressive strength by increasing amount of additives.

Keywords; Saw dust, Rice husk, Bagasse, Portland cement, Compressivestrength

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1. Introduction:

Waste Immobilization converts raw waste, usually containing mobile contaminantsinto a solid and stable form, termed a wasteform. The properties of the waste form enable it to be handled, stored and disposed of safely and conveniently, significantly reducing the potential for release of radionuclides into the environment. Forlong-term storage and disposal, waste immobilization should be an irreversible process that avoids release of contaminants from the matrix during storage and disposal [1]. The main immobilization technologies that are available commercially and have been demonstrated to be viable are cementation, bituminization and vitrification. A number of new immobilization technologies are under development [2]. Cementation of radioactive waste has been practiced for many years basically for immobilization of low and intermediate level waste. Waste cementation is based on use of hydraulic cements. Hydraulic cements are inorganic materials that have the ability to react with water under ambient conditions to form a hardened and water resistant product. The most common cements are those based on calcium silicate such as Portland cement.Portland cement can set normally in air as well as when emplaced under water [3]. The purpose of this research was to investigate the mechanical properties (compressive strength) of the final solid block of natural radio contaminated materials fixed with ordinary Portland cement.

2-Materials and characterization:

2.1-Saw dust:

It is also known as wood dust. It is the waste material of timber industry.saw dust used in this work is natural material. The chemical composition of saw dust is shown in table 1. This table clears that silicon oxide SiO_2 is the main mineral which constitutes 85% by weight of the total weight of the sample.

2.2- Ricehusk:

Rice husk used is natural material. Most farmers consider rice husk ash as a waste so they burn it. this process constitutes environmental hazards. In this research work we use natural waste (rice husk) to treat and solidify radio waste. Table 2 shows the chemical composition of rice husk where SiO_2 constitutes 91.59% by weight of the sample.

Weight Percentage
(%)
85
2.7
1.7
0.25
3.5
4.3

Weight Percentage
(%)
91.59
4.8
1.58
0.53
0.39
0.21
Trace
0.2

Table 1: Chemical composition of Saw dust [4] Table 2: chemical composition of rice husk [5] Minerals Weight Demonstrate

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2.3- Bagasse:

Sugar cane bagasse used in this work is natural material.Bagasse is a byproduct of sugar industry and is the fibrous matter that remains after sugarcane stalks are crushed to extract their juice. Bagasse is used as a renewable power generation source and for production of bio-based materials. It is used as a biofuel and in the manufacture of pulp and building materials. A typical chemical composition of bagasse isshown in table 3 and SiO₂ is the main mineral and constitutes 73% by weight of the sample.

Table 3: Chemical composition of bagasse [6]

Weight percentage
(%)
73
6.7
6.3
2.8
3.2
4
1.1
2.4

2.4- Cement:

Ordinary Portland cement used is from**CEMEX EGYPT**company.Cements are binding agents in concretes and mortars. Concrete is an artificial rock-like material, basically a mixture of coarse aggregate (gravel or crushed stone), fine aggregate (sand), cement, air, and water. The term Portland cement is a general term used to describe a variety of cements used today. Portland cements are hydraulic cements, which means they will set and harden by reacting chemically with water through hydration. It is manufactured through the blending of mineral raw materials at high temperatures in cement rotary kilns. Rotary kilns produce an intermediate product called "clinker." Clinker is ground to produce cement. By modifying the raw material mix and, to some degree, the temperature of manufacture, slight compositional variations in the clinker can be achieved to produce Portland cements with varying properties. When Portland cement is mixed with water to make paste, it becomes gradually less plastic and finally becomes a hard mass. When it loses plasticity, it is sufficiently rigid to withstand a definite amount of pressure. Cement is widely used for solidification of Low level waste and Intermediate level waste. Table 4 shows the chemical composition of Portland cement.

1	· · · · · · · · · · · · · · · · · · ·
Minerals	Weight percentage (%)
SiO_2	23.77
$Fe_2 O_3$	6.2
$Al_2 O_3$	5.41
CaO	57.14
MgO	2.28
SO_3	2.25
Na ₂ O	0.64
K ₂ O	0.25
Cl	0.07
Cr(Hexavalent)	1.10

 Table 4:Chemical composition of Portland cement[7]

30% HClandKOH used fromEL Nasr pharmaceutical chemicals company.

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3-Morter preparation and testing of specimens:

Natural materials used in this work were soaked in Hydrochloride acid (HCl)and in Potassium hydroxide(KOH)1M solution for week then being squeezed and rinsed by 500ml of tap water twice and being dried at 60 $^{\circ}$ C for 24 hours then drilled to size of1mm.Untreated species also drilledto size 1mm and water to cement ratio used is 30:70.Nine cylindrical samples of diameter 5cm and thickness of 1cm were prepared manually bymixing 70g of cement with 30g water and (1g,2g,3g) of treated and untreated species.Curing time is one month. Compressive strength tests carried by Power Pack Compression Testing Machine figure (1) for three times and average of values are calculated. The compressive strength is defined by the amount of force on specimen can receive before it cracks according to following equation:

Stress= Force/ Area=F/A

Where F: Loaded forceon specimen, (Newton). A: Area of specimen, (Cm^2) .



Figure(1): Power pack Compression testing machine

4-Results and Discussion:

4.1- Effect of temperature:

For studying effect of high temperatures on compressive strength, the samples were exposed to different temperatures (50, 100, 150, 200°C) and compressive strength measured at each degree for three times and average calculated. Tables 5, 6, 7 represent the experimental results obtained for effect of temperature on the compressive strength for cement samples contain natural additives untreated or treated by HCl or KOH.

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Tuble 5. Experimental auta obtained for cement samples contain sulv aust					
Temperature	Compressive strength (KN/Cm ²)				
(°C)	untreated	Treated by HCl	Treated by KOH		
50	76	119	88		
100	85	110	80		
150	88	150	53		
200	120	103	49		

Table 5: Experimental data obtained for cement samples contain saw dust

Table 6: Experimental data obtained for cement samples contain rice husk

Temperature(°C)	Compressive strength (KN/Cm ²)		
	untreated	Treated by HCl	Treated by KOH
50	25	77	65
100	22	80	40
150	20	84	38
200	20	99	30

Table 7: Experimental data for cement samples contain bagasse

Temperature(°C)	Compressive strength (KN/Cm ²)			
	untreated	Treated by HCl	Treated by KOH	
50	45	92	57	
100	54	70	46	
150	64	59	44	
200	40	50	20	

4.1.1-For untreated natural additives:

The effect of temperature on the compressive strength for cement saw dust samples is shown in figure (5). From this figure it is clear that by increasing temperature from 50 to 200°C, the compressive strength increases from 76 to 120KN/Cm². For cement-bagasse blocks, compressive strength increase from 45 to 54KN/Cm²as temperature increases from 50 to 100°C. The observed improvement in compressive strength can be attributed to the hydration of water out of the concrete mass. At (100°C), the compressive strength starts to diminution as a result of the evaporation of the free water in concrete mass. The water continues to drive out of the specimen and start to configure paths to get out the water stream inside the pores of the specimen, this naturally caused internal stresses lead to wreaking of the concrete mass (may create a micro-crack in the paste). After that, the links between the concrete particles begin to weaken as a result of the redistribution of particles, which explains the weakness of concrete compressive strength at the temperature of (150°C) [7]. For cement-rice husk samples, compressive strength decreases gradually as temperature increase. This may be due to that, burning of rice husk lead to two forms of silicon dioxide crystalline and amorphous. Burning at temperature at about 700°C lead to amorphous phase which is responsible for the pozzolanic activity of rice husk and down this degree the crystalline phase is obtained [8] as shown in figure(2).

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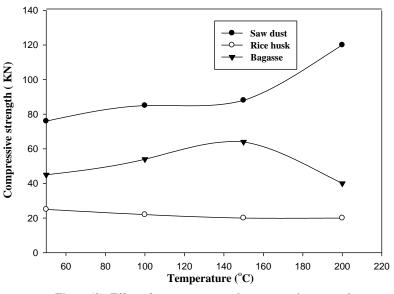


Figure (2): Effect of temperature on the compressive strength for cement samples contain 1gm of untreated additives(saw dust, rice husk, bagasse)

4.1.2- For HCl treated specimens:

For cement samples contain HCl treated bagasse or saw dust, compressive strength decrease by increasing temperature from 50 to 200 °C as shown in figure (3) which shows the effect of temperature on compressive strength for cement samples contain HCl treated additives. From the figure it is clear that at 50 °C compressive strength for sample contain saw dust treated by HCl was 119KN/Cm² and at 200 °C was 103KN/Cm². For samples contain HCl treated bagasse compressive strength at 50°Cwas 92KN/Cm² and at 200°C was 50KN/Cm². For cement- rice husk blocks, compressive strength increase by increasing temperature from 50 to 200°C and this may be due to that rice husk pretreated with HCl has a great amount of amorphous SiO₂ which result in an increase in pozzolanic activity of rice husk leading to increase in compressive strength [9-10]as shown in figure(3).

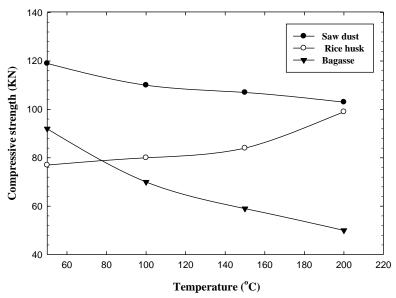
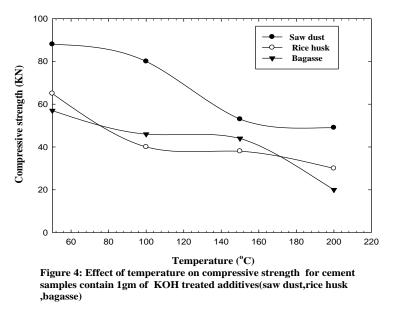


Figure 3: Effect of temperature on the compressive strength for cement samples contain 1gm of HCl treated additives(saw dust, rice husk, bagasse)

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4.1.3- For KOH treated specimens:

By increasing temperature from 50 to 200°C,for cement blocks contain KOH treated saw dust,compressive strength decrease from 88 to 49KN/Cm²as shown in figure(4) that shows effect of temperature on compressive strength of cement samples contain KOH treated additives. The same happens for cement samples contain KOH treated bagasse or rice husk where compressive decrease from 57KN/Cm² to 20KN/Cm² for cement-bagasse samples and from 65 to 30KN for cement-rice husk samples as shown in figure (4). The decrease in compressive strength may be due to reduction in pozzolanic activity of additives by increasing temperature.



4.2-Compressive strength tests:

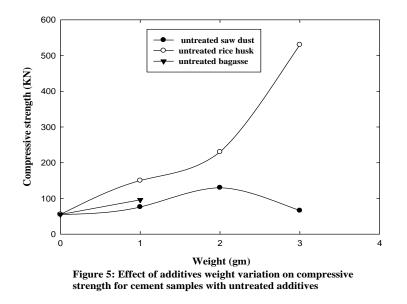
Compressive strength is the important factor on the mechanical stability of the final solid waste block.

4.2.1- For untreated natural materials

The effect of additives weight variation on compressive strength for cement samples contain untreated natural additives (saw dust, rice husk, bagasse) is shown in figure 5. For cement blocks containing saw dust, compressive strength increase by increasing amount of sawdust added(1 gm-2 gm) form 55 to 130KN/Cm² and above 2gm compressive strength decreases(66KN/Cm²) as amount of sawdust added increase(3gm). This may be due to the increase in porosity resulting from the penetration of water into the fibers and saw dust particles [11]. For cement blocks contain rice husk, compressive strength increases gradually from 55 to 530KN/Cm² by increasing amount of additive from 1 to 3gm. The same happens for cement blocks contain bagasse where compressive strength increase from 55 to 96KN/Cm² by increasing amount of additives, but for further additives added the samples begins to jogging. The increase in compressive strength can be explained due to physical and chemical aspects. The physical effect is that ultra-fine particles fill the voids in cement which make micro structure of cement paste denser. The chemical effect may be due to formation of additional calcium silicate hydrates from the pozzolanic reaction of additives. The reduction in compressive strength with increasing amount of additives added may be due to saturation of cement mix with oxides such as MgO, K_2O present in additives which form composites that may inhibit the formation of strength giving calcium silicate hydrates from cement hydration, also the further replacement of cement by additives decreases the cement content in the paste and increase the amount of water needed for the excess additives which lead to

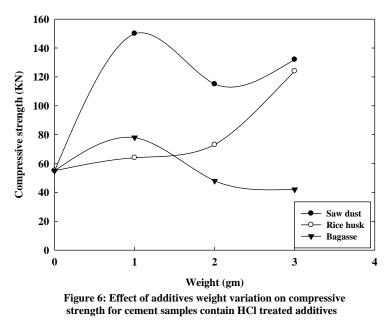
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that water unavailable for cement hydration which leads to reduction in hydration reaction and decrease in compressive strength of the block [12].



4.2.2- For HCl treated specimens:

For cement blocks contain saw dust treated by HCl, compressive strength increase (for 1gm added) from 55 to 150 KN/Cm², but for further addition of saw dust, compressive strength decreases as shown in figure 6 which clear the effect of additives weight variation on compressive strength for cement- HCl treated saw dust blocks. For cement blocks containing rice husk treated by HCl, Compressive strength increases gradually from 55 to 124KN/Cm²by increasing amount of additives from 1 to 3gm. This increase may be due to that rice husk treated byHCl contain fewer metal impurities such as K,Mg resulting in a higher pozzolanic activity of rice husk and a better strength performance of the paste. The pore diameter of rice husk- cement paste after HCl pretreatment tends to be smaller which may be one of reasons for the strength increase [13]. 1 gmHCl treated bagasse added to the cement samples, compressive strength increase from 55 to 78KN/Cm²while, for more addition of bagasse (2,3gm), compressive strength decreases as shown in figure (6).



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4.2.3- For KOH treated specimens:

Figure 7 shows the effect of additives weight variation on compressive strength of cement samples contain KOH treated additives (saw dust, rice husk, bagasse). From this figure it is clear that, for cement blocks contain 1 gmsaw dust treated by KOH, compressive strength increases from 55 to 100KN/Cm², but by increasing additives amount (2gm, 3gm) the compressive strength decreases. The increase in compressive strength may be due to the increase in saw dust content in the cement paste need more water for normal consistency of cement paste and this lead to decrease in compressive strength of the block. For rice husk additives sample, compressive strength decreases from 55 to 30KNCm². Treatment of rice husk by KOH lead to increase in amount of alkalis present which act as an accelerator for crystallization of SiO₂ leading to reduction in pozzolanic activity of rice husk and decrease in compressive strength. For cement-bagasse blocks, compressive strength increases from 55 to 140KNCm² by increasing amount of additives. This may be due to that bagasse fiber consists of crystalline cellulose connected to hemicellulose and lignin and by treating with KOH, KOH react with OH⁻ of hemicellulose leading to destruction of structure and fiberssplit into filaments[14]. This lead to increase in surface area and increases the contact and adhesion to the cement, but for further increase in amount of additives added lead to decrease in compressive strength (100KNCm^2) as shown in figure (7).

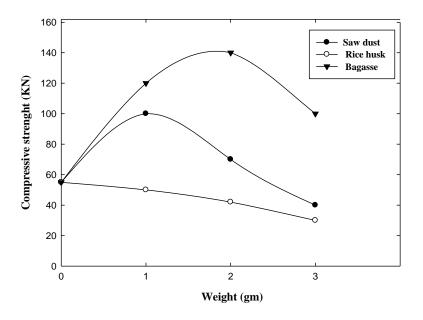


Figure 7: Effect of additives weight variation on compressive strength for cement samples contain KOH treated additives

5-Conclusion:

The conclusion of this study can be summarized as follow:

Sawdust ash can be used for improving mechanical properties of Portlandcement. Untreated sawdust shows good results in improving compressive strength of cement and for higher temperature. For treated sawdust by HCl and KOH there was variation in results depending on amount added. For bagasse its results were not satisfied to improve mechanical properties of Portland cement. For rice husk both treated and fresh one give good results in improving mechanical properties of Portland cement.

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