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A Study on Cement Made by Partially Replacing Cement Raw Materials with Municipal Solid Waste Ash and Calcium Carbide Waste

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ABSTRACT The use of Municipal Solid Waste incinerator bottom ash (MSWI) and Calcium Carbide Waste (CCW) as a part of cement raw material was investigated. The purpose was not only to dispose of the wastes, but also to alleviate some environmental problems, by reducing resources usage, CO_2 emissions and energy consumption in cement manufacturing. The replacement of MSWI and CCW in raw meal was 5 and 10 percent. Chemical composition and general characteristics, as well as setting times and compressive strength, of the MSWI cements and CCW cements were tested and compared with conventional cement. The chemical compositions of MSWI cements and CCW cements were similar to the control cement, except that the SiO_2 component in MSWI cements was higher than that in control cement but the CaO component was lower. Setting times of cement pastes were slightly different when MSWI and CCW were used as raw materials in cement. The longer setting times of these cement pastes than those of control cement is due to lower C_3S and higher C_2S levels than in CC. Compressive strength of CCW cement mortars was closed to that of the CC cement, whereas compressive strength of mortar produced from MSWI cements was rather smaller than the control cement mortar, especially at higher MSWI percentags.

KEYWORDS: municipal solid waste (MSW) ash, calcium carbide waste (CCW), cement manufacturing, setting time, compressive strength, emission, energy conservation.

INTRODUCTION

Municipal Solid Waste (MSW) generation in Thailand is of critical concern, especially in big cities. Bangkok, alone, produced approximately 8,000 tons per day in 2002. The incineration of municipal solid waste, an effective method of volume reduction, is presently receiving widespread attention as a final disposal method of MSW in Bangkok. Like wise, Calcium Carbide Waste (CCW), a by-product from producing acetylene gas (C_2H_2) is produced in high amounts, approximately 30,000 tons in the year 2002. In general, CCW or calcium hydroxide ($Ca(OH)_2$) is obtained from the reaction between water and calcium carbide (CaC_2) as shown in the equation below:

$$CaC_2 + 2H_2O \longrightarrow C_2H_2 + Ca(OH)_2$$

CaC_2 is prepared by burning limestone ($CaCO_3$) to yield lime (CaO) and carbondioxide (CO_2) ($CaCO_3 \rightarrow CaO+CO_2$). After that, CaO reacts with coal (C) and CaC_2 together with carbonmonoxide (CO) are obtained ($CaO + 3C \rightarrow CaC_2 + CO$).

The MSW incineration process creates two general types of ash; fly ash and bottom ash. These ashes CCW from producing acetylene gas are normally disposed of by landfilling, which may create further problems, *ie* the leaching of harmful compounds and alkali to groundwater. On the other hand, if MSW ash and CCW can be used in concrete, it will not only be able to reduce the consumption of cement raw materials, but also to solve the MSW ash and CCW disposal problems simultaneously.

Sisomphon K, *et al*[1] found that MSW ash has an irregular grain surface and very high specific surface area. Other properties such as high loss on ignition, highly variable in characteristics and low reactivity were also contributing problems in the reuse of MSW ash as a pozzolan. Hamernik and Frantz[2] studied the properties of concrete containing MSW fly ash and reported that different burning conditions affected the reactivity of MSW fly ash. In addition, samples from different compositions resulted in different chemical and physical properties of the final MSW ash cement. Krammart P, *et al*[3] studied the use of CCW as cement replacing material. The results show that the setting time of paste was delayed significantly. Compressive strength of the mortars replaced with CCW was also greatly reduced when compared with the control mortar. Krammart P, *et al*[4] classified the combustible MSWs into three major types; paper, leaves, and food. After preparation, leaves, paper and food were separately burned in a ferrocement incinerator. Finally, all types of combustible MSW ashes were ground in a grinding machine fixed at 45 minutes. The weight ratio of each combustible MSW ash to total raw material was fixed at 0.05 for all of the experiments. They found that chemical composition and setting property of these cement, as well as the compressive strength of mortar, were rather close to the control cement.

From the previous research, the use of MSW ash and CCW as a pozzolan or cement replacing material gave undesirable properties of the cementitious materials[1,3]. Another research used MSW ash as a part of raw materials by classifying the combustible MSW in to paper, leaves and food. The results showed that the general properties were similar to Ordinary Portland Cement (OPC).[4] However, in practice it is difficult to classify MSW. Accordingly, this study presents the possibility of using MSWI and CCW as a part of raw materials in cement manufacturing without adjusting the proportion of raw meal. For real applications, if MSWI or CCW is replaced in raw materials, it may be necessary to adjust the proportion of raw meal. This new type of cement is expected to improve energy efficiency, to conserve raw materials and to reduce air pollution of the cement manufacturing, while the cement quality is expected to be the same as that of OPC.

MATERIALS PREPARATION

MSW Ash Preparation

For MSW incinerator ash, the bottom ash obtained from an incinerator in Phuket province, Thailand was used in this study. By drying the obtained bottom ash in an electrical oven at 105°C for 24 hours and grinding in the Los Angeles Abrasion machine about 45 minutes, MSWI with a Blaine fineness of approximately 1,000 cm^2/g was obtained.

CCW Preparation

To prepare CCW used in this study, the CCW were dried at 105°C for 24 hours in an electrical oven and later ground in the Los Angeles Abrasion machine for 30 minutes to a Blaine fineness of about 4,000 cm^2/g.

METHODS

The experimental program comprised of 5 series of mixes, namely control cement and each MSWI and CCW cement consisting of two different MSWI and CCW percentages in the raw material. For both MSWI cements and CCW cements, the weight ratio of MSWI and CCW to total raw material was varied at 0.05 and 0.10. The conventional raw meal composed of limestone, shale and clay was obtained from a Cement plant at Kaeng Khoi, Saraburi, Thailand. Physical properties and chemical compositions of the raw materials are shown in Tables 1 and 2, respectively.

Firstly, each MSWI and CCW were blend with conventional raw materials in a mechanical mixer and then water was added. Then, a cylindrical-shape bar was molded with a diameter of one inch and dried in an oven at 105°C for 24 hours. Next, these prepared raw meal bars were burned in a high-temperature electrical furnace with a heating rate of 20°C/minute and maintained at 1,450°C for 30 minutes. Finally, the clinker was cooled at a rate of 20°C/minute constantly and ground to cement powder. The finished products had the Blaine fineness of 3,100±100 cm^2/g with 5 percent gypsum added during grinding.

In this study, there were two sets of mortar mixes, depending on the type of sand. For the first set, sand with the gradation according to ASTM C 778 was used. Four series of mixes, representing two types of MSWI cement mortar (5 and 10 percent of MSWI in raw meal) and two types of CCW cement mortar (5 and 10 percent of CCW in raw meal), were tested in the laboratory to compare their properties with the cement made from the control raw meal. All mix proportions were determined by fixing the sand to cement ratio at 2.75 and the water to cement ratio (w/c) at 0.70 in order to produce the flow value of control mortar at 110±5. Another set uses the local, natural river sand, as the ingredient of mortar, while the series of mixes used are the same as the first set. Similarly, all mix proportions were determined by fixing the sand to cement ratio as in the first set and the water to cement ratio (w/c) equal to 0.53, in order to produce the flow value of control mortar equal to 110±5.

The initial and final setting times of all cements were determined. The compressive strength of mortar at 7, 14 and 28 days was tested. All samples were cast in 5´5´5 cm steel molds and cured in water at room temperature. The compressive strength was an average from 5 specimens.

RESULTS AND DISCUSSION

The test results are discussed on two aspects, i.e. the effects of MSWI and CCW replacement on the properties of the cement, and the benefits to environmental conservation and the cement manufacturing process.

Effect on Properties of Cement

The chemical composition of all cements are shown in Table 3. Setting times, flow and compressive strength of control cement (CC), MSWI cements with 5 percent MSWI (5MSWIC), 10 percent MSWI (10MSWIC), as well as CCW cements with 5 percent CCW (5CCWC) and 10 percent CCW (10CCWC) are given in Table 4.

The results indicated that the chemical compositions

Table 1. Physical properties of the cement raw meal, MSWI and CCW.

Physical Properties	Raw Meal	MSWI	CCW
Specific Gravity	n.o.	2.50	2.26
Blaine Fineness (cm^2/g)	n.o.	937	4,100

n.o.: not observed
MSWI: municipal solid waste incinerator bottom ash
CCW: calcium carbide waste

Table 2. Chemical composition of the cement raw meal, MSWI and CCW.

Chemical Composition	Raw Meal(% wt)	MSWI(% wt)	CCW(% wt)
SiO_2	12.82	48.26	6.49
Al_2O_3	3.78	4.04	2.00
Fe_2O_3	2.24	4.4 4	1.87
CaO	43.53	19.07	56.41
MgO	0.63	1.16	0.70
SO_3	n.d.	1.06	0.36
Na_20	0.09	2.41	0.18
K_2O	0.41	1.17	0.10
TiO_2	0.16	n.d.	n.d.
P_2O_5	n.d.	n.d.	n.d.
Total Cl^- Content	n.d.	0.01	n.d.
LOI	36.14	16.56	31.74

n.d.: non-detectable due to zero or very small concentration
LOI: loss on ignition

of MSWI and CCW cements are similar to the control cement, except that the SiO_2 contents of 5MSWIC and 10MSWIC, which equals to 23.34% and 24.85%, respectively, are higher than that of the CC (20.03%). On the other hand, the CaO component in CC (65.88%) is higher than in 5MSWIC (62.75%) and 10MSWIC (61.13%), as shown in Fig 1. As a result, C_3S from Bogue's composition of 5MSWCI (30.80%) and 10MSWIC (13.17%) was considerably lower than that of CC (56.02%), whereas C_2S was significantly higher (43.89% for 5MSWIC, 61.44% for 10MSWIC and 15.24% for CC), as shown in Fig 2. The water requirements of CC, 5CCWC and 10CCWC cement were almost unchanged as can be seen from the small difference in normal consistency, while the 5MSWIC and 10MSWIC cement required more water, especially when the MSWI percentage increased. The results also indicate that times of setting are slightly different when MSWI and CCW are replaced as a raw material in cement. It was noted that 5MSMIC, 10MSWIC, 5CCWC and 10CCWC cement exhibited the longer setting times than the control cement, due to the lower C_3S and higher C_2S than those in CC, according to the Bogue's composition shown in Table 3 and Fig 2. Moreover, the water requirement had an effect on the flow of mortars. The higher the water requirement of the cement, the lower the flow of mortar, as shown in Table 4. Fig 3 and Fig 4 show the compressive strength development of cement with respect to age. For mortar samples with

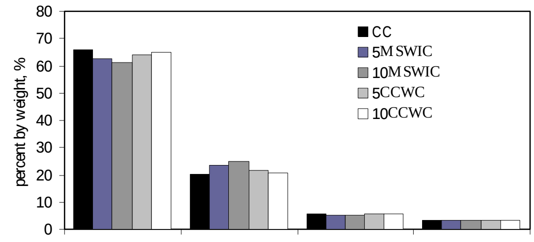

Fig 1. Comparison of four major oxides of cements.

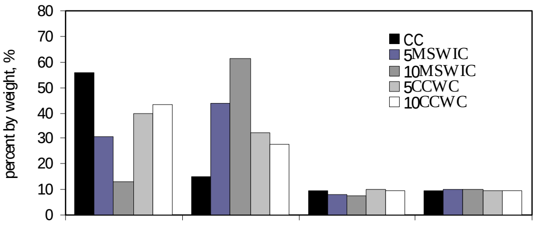

Fig 2. Comparison of oxide compound compositions of cements.

5CCWC and 10CCWC cement, compressive strength was close to that of the CC cement. Whereas the compressive strength of mortars produced from MSWI cements, both 5MSWIC and 10MSWIC, was lower than that of control cement mortar, especially at the higher MSWI percentage. Since MSWI cements have lower CaO and higher SiO_2 than those in CC, C_3S is lower than CC. Therefore it is recommended here that raw meal

Table 3. Chemical composition of control cement, MSWI cement, and CCW cement.

Chemical Composition	CC (% wt)	5MSWIC (% wt)	10MSWIC (% wt)	5CCWC (% wt)	10CCWC (% wt)
SiO_2	20.03	23.34	24.85	21.61	20.89
Al_2O_3	5.69	5.17	5.00	5.75	5.60
Fe_2O_3	3.12	3.29	3.34	3.20	3.17
CaO	65.88	62.75	61.13	64.00	64.85
MgO	0.80	0.93	0.99	1.05	1.04
SO_3	2.33	2.01	2.26	2.14	2.14
Free CaO	2.59	0.50	0.48	1.72	3.40
Total Cl^- content	n.d.	n.d.	n.d.	n.d.	n.d.
LOI	0.57	1.44	1.41	1.74	1.25
Blaine fineness (cm^2/g)			3,100±100		
Specific Gravity	3.04	3.11	3.12	3.10	3.06
Bogue's Composition					
Computed C_3S	56.02	30.80	13.17	39.93	43.07
Computed C_2S	15.24	43.89	61.44	32.07	27.65
Computed C_3A	9.81	8.14	7.61	9.83	9.48
Computed C_4AF	9.48	10.00	10.15	9.73	9.64

CC: control cement, 5MSWIC: MSWI cement with 5 percent MSWI, 10MSWIC: MSWI cement with 10 percent MSWI,5CCWC: CCW cement with 5 percent CCW, 10CCWC: CCW cement with 10 percent CCW
n.d. :non-detectable due to zero or very small concentration

Table 4. Normal consistency and setting time of paste and compressive strength of mortar.

Type of Cement	Normal Consist.	Setting Time (hr:min)		Mortar Using Sand I, w/c = 0.70				Mortar Using Sand II, w/c = 0.53			
				Flow	Compressive Strength (MPa)			Flow	Compressive Strength (MPa)		
		Initial	Final		7-day	14-day	28-day		7-day	14-day	28-day
CC	0.245	2:25	3:05	105	19.20	22.05	26.70	105	40.31	45.91	49.97
5MSWIC	0.265	3:00	3:50	92	9.96	12.25	13.85	93	27.26	31.21	36.16
10MSWIC	0.290	2:25	3:20	88	3.72	4.86	7.54	88	8.32	11.66	16.28
5CCWC	0.240	2:40	3:35	105	18.24	22.30	27.05	105	38.04	48.28	51.25
10CCWC	0.230	2:25	3:20	107	18.71	21.02	26.89	106	38.76	43.51	44.49

Sand I: meeting ASTM C 778 gradation requirement.
Sand II: local, natural river sand with a fineness modulus of approximately 3.29.
w/c: water to cement ratio

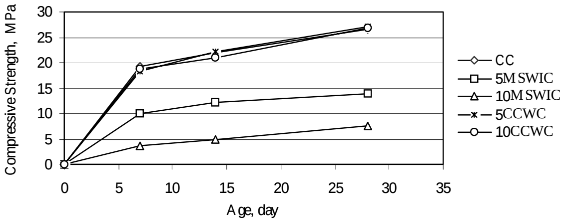

Fig 3. Compressive strength development of cement with w/c 0.70.

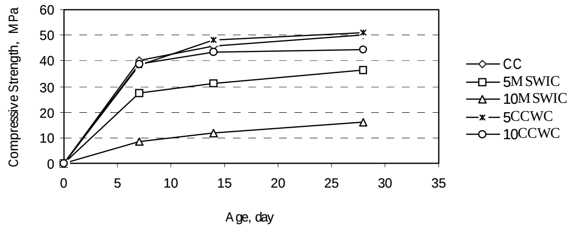

Fig 4. Compressive strength development of cement with w/c 0.53.

adjustment is needed if MSWI is replaced in order to obtain cement of similar quality to CC.

With respect to the chloride content in the derived products, most chlorides in the ash were vaporized together with alkali contained in raw meal in the clinkerization process.[5] So, high chloride content, which is a typical problem of using MSW ash as a direct cement replacing material in concrete, was not problematic. In MSWI, the chloride content was 0.01% by weight, while in the case of CCW, chloride was not detectable due to zero or very small concentration. The total chloride contents in the MSWI cements and CCW cements shown in Table 3 are far below the allowable limit defined, for example, by the British standard (0.10 % by weight of cement as the most serious case for prestressed concrete).[6]

Benefits to Environmental Conservation and the Cement Manufacturing Process

Reduction of Landfill Space and Trace Harmful Elements

Not only due to large amount of MSW and CCW but also due to limited landfill space and issues of groundwater contamination, landfilling of either MSW and CCW or their residues are facing strong opposition from the public. If all wastes are used in the cement production, the need for landfill space for these wastes will be reduced. Moreover, after MSW ash cements and CCW cements are used to produce concrete, the trace harmful elements, *ie*, Cd, Cr, Mn, Zn, Cu, or Pb contained in the waste are dissolved in the hydration process and are converted to insoluble forms with hydroxides in the alkaline environment given by the hydration of cement. Arsenic is fixed as a solid solution in calcium sulfoaluminate hydrate (ettringite) by substitution of SO_4^{2-} and Hg is adsorbed to the surface of C-S-H. Thus, these elements are fixed in concrete, so they are hardly released into the environment.[7]

Raw Material Conservation

Quarrying the raw meal for cement manufacturing, especially limestone involves an intervention in nature and the landscape. Because of higher awareness of environmental and natural conservation, raw material acquisition in many countries either by extending existing quarries or starting new ones has become more difficult. Apart from the raw material conservation by direct replacement of the raw meal by MSWI and CCW, the use of MSWI and CCW indirectly leads to an extra raw material conservation by increasing the production yield due to the lower LOI values of MSWI and CCW than the raw meal. In Table 5, the amounts of final cement products were calculated based on LOI values of raw meal, MSWI and CCW as follows:

$$Wc = W_{RM}(1 - LOI_{RM}) + W_{MC}(1 - LOI_{MC})$$

where

- W_C is the weight of final clinker product
- W_{RM} is the weight of conventional raw meal used as a raw material
- W_{MC} is the weight of MSWI or CCW used as a raw material
- LOI_{RM} is the loss on ignition of conventional raw meal
- LOI_{MC} is the loss on ignition of MSWI or CCW

From Table 5, the LOI value of raw meal is 36.14%, while the LOI of MSWI and CCW, are only 16.56% and 31.74%, respectively. This result explains raw material conservation of the cement process when MSWI and CCW are replaced as raw materials. Based on the properties of raw meal, MSWI and CCW used in this study, the relationship between raw meal conservation and MSWI and CCW replacing percentage is shown in Fig 5.

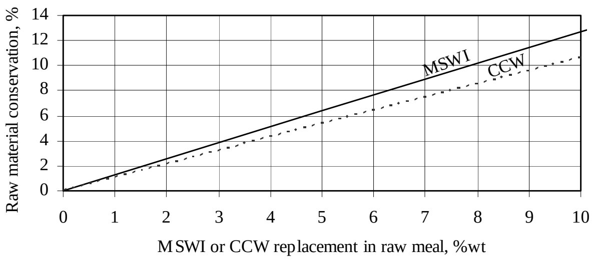

Fig 5. The relationship between raw meal conservation and MSWI and CCW replacement percentage.

Table 5. Raw meal requirement of MSWI cements and CCW cements as compared with the conventional cement for a ton of clinkers.

Types of cement	Raw materials requirement (ton)		Raw material conservation	
	Raw meal	MSWI or CCW	%wt	ton
CC	1.5659	0	-	0
5MSWIC	1.4652	0.0771	6.4344	0.1007
10MSWIC	1.3674	0.1519	12.6774	0.1985
5CCWC	1.4825	0.0780	5.3262	0.0834
10CCWC	1.3997	0.1555	10.6159	0.1662

Reduction of Carbon Dioxide Emission

In the cement production, air pollution is one of the environmental problems of greatest concern. The gases from the kilns are normally identified as CO, CO_2, and NO_x. In Thailand, cement production is one of the major producers of CO_2. The CO_2 emission comes from fuel burning for clinkerization and from the dissociation of the raw materials ($CaCO_3$ and $MgCO_3$). Here, only CO_2 emitted from the dissociation of raw materials will be considered. In conventional cement, $CaCO_3$ is the major source of CaO and also carbon dioxide. In this study, MSWI and CCW serve as sources of CaO that emit less CO_2 than limestone or other conventional calcareous sources because their calcium oxide is usually not in the carbonate form. However, unburned carbon in MSWI also contributes to the emission of CO_2. Since CCW is a by-product from producing acetylene gas, which uses $CaCO_3$ and coal in the process, CO_2 emitted from CCW is expected to be from the unburned carbon and some $CaCO_3$ left in the CCW. The CO_2 emission from the conventional raw meal, MSWI, and CCW, investigated by the ultimate analysis test, are shown in Table 6. The samples were burnt in a high temperature furnace at the temperature of about 1,100°C, and a CO_2 absorber was used to measure the amount of CO_2 emitted from the material.

From Table 6, the amount of CO_2 emitted from cement manufacturing depends on the properties of the raw material especially LOI and the amount of carbon contained in the raw materials. Table 6 shows that the CO_2 emission varies with LOI values of the raw materials. So, the advantages of lower LOI of MSWI and CCW than the conventional raw meal not only improve the production yield but also reduce CO_2 emission significantly.

Based on the CO_2 and LOI data in Table 6, CO_2 emissions from different cement types are computed as in the following equation and summarized in Table 7.

$$CO_2 = \frac{\text{weight of } CO_2 \text{ emission per unit weight of raw mill}}{\text{weight of final product per unit weight of raw mil}}$$

$$CO_2 = \frac{(1-\frac{r}{100})(\frac{CO_{2,RM}}{100}) + (\frac{r}{100})(\frac{CO_{2,MC}}{100})}{(1-\frac{r}{100})(1-\frac{LOI_{RM}}{100}) + (\frac{r}{100})(1-\frac{LOI_{MC}}{100})}$$

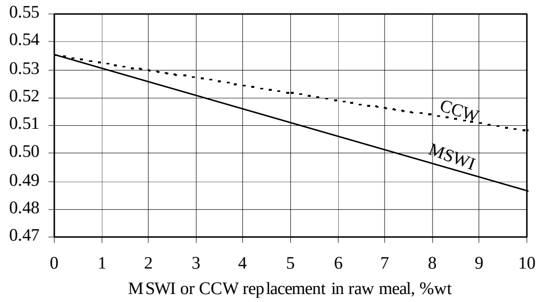

Fig 6. The relationship between CO_2 emission per ton of final clinker and MSWI and CCW replacement percentage.

where

- CO_2 is the weight percentage of CO_2 emission from cement raw material during cement manu-facturing process, %wt
- r is the MSWI or CCW replacement percentage, %wt
- $CO_{2,RM}$ is the weight of CO_2 emitted from raw meal, %wt (34.21%)
- $CO_{2,MC}$ is the weight of CO_2 emitted from MSWI or CCW, %wt (12.36% for MSWI and 18.95% for CCW)
- LOI_{RM} is the loss on ignition of conventional raw meal, %wt (36.14%)
- LOI_{MC} is the loss on ignition of MSWI or CCW, %wt (16.56% for MSWI and 31.74% for CCW)

Based on the properties of the raw meal, MSWI and CCW used in this study, the relationship between the amount of CO_2 emission from the raw material, MSWI and CCW replacing percentage is shown in Fig 6.

Thermal Energy Conservation

Cement is an energy intensive product with about 475 kcal/kg for $CaCO_3$ dissociation and has the highest thermal energy consumption in clinkerization process[8]. So, reducing $CaCO_3$ in raw meal means reduction of the total energy consumption of the process. Using MSWI and CCW, which usually have CaO in other forms than carbonate form, is also expected to reduce the fuel or thermal energy requirement for changing $CaCO_3$ into the reactive CaO.

Table 6. LOI and potential releases of CO_2 for different types of materials in weight percentage.

Types of Materials	CO_2 emission (%wt)	LOI (%wt)
Conventional Raw Meal	34.21	36.14
MSW incinerator bottom ash	12.36	16.56
Calcium Carbide Waste	18.95	31.74

Table 7. Amount of CO_2 emission of final clinker products for CC, MSWI and CCW cement.

Types of cement	CO_2 emission of final clinker (%wt)	CO_2 emission reduction of final clinker	
		Percent of reduction when compared with CC	CO_2 reduction per ton of clinker (kg)
CC	53.57	0	0
5MSWIC	51.08	4.66	24.9
10MSWIC	48.66	9.17	49.1
5CCWC	52.20	2.57	13.7
10CCWC	50.83	5.11	27.4

Table 8. Free lime content in control clinker, MSWI clinkers and CCW clinkers.

Chemical Properties	CC	5MSWIC	10MSWIC	5CCWC	10CCWC
Total CaO (% wt.)	65.88	62.75	61.13	64.00	64.85
Free CaO (From wet analysis, % wt.)	2.59	0.50	0.48	1.72	3.40

Burnability of the raw meal is one of the important factors in cement production, especially due to its relation with thermal energy consumption of the production. The burnability of cement raw meal is defined as the amount of mass transfer of its constituents with ease or difficulty to the clinker phases. Burnability is popularly assessed on the basis of the amount of free lime in the product. Raw meal with high burnability can be burned at a lower temperature or for a shorter period of burning. Therefore, if the amount of input thermal energy for clinker burning is controlled, burnability improvement, as reflected by reduction of free lime content in the final cement products, can be obtained as in Table 8.

The excess free lime usually hydrates very slowly, causing unsoundness of the cement paste in the hardened state. So, the reduction of free lime content not only results in burnability improvement of the cement, but also in the volume stability of the concrete. In regards of thermal energy consumption, replacing with 10% CCW may not be beneficial.

Conclusion

All tested municipal solid waste incinerator ash replacement cement and calcium carbide waste replacement cement had slightly different setting properties from the control cement. The compressive strength of mortars using the calcium carbide waste cements was not much different when compared with that of the control cement. However, the compressive strength of mortar produced from the municipal solid waste incinerator ash cement was lower than that of the control cement mortar, especially when the ash percentage increased.

Natural resources are conserved by the direct replacement of municipal solid waste incinerator ash and calcium carbide waste in the raw meal. The production yield is improved due to lower LOI of both the incinerator ash and calcium carbide waste than the conventional raw meal. Because the incinerator ash and calcium carbide waste mainly contain non-carbonate CaO sources, the production of the incinerator ash and calcium carbide waste cements generates less CO_2 than the conventional cement process and also leads to lower energy consumption in cement manufacturing.

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