
ENVIRONMENTAL IMPACTS OF SOME EGYPTIAN ELECTRIC ARC FURNACE SLAG UNDER DIFFERENT COOLING RATES

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ABSTRACT

Electric arc furnace (EAF) slag is a byproduct materials its chemical composition more closely to the cement clinker and composed mainly from different types of calcium silicate minerals. EAF slag utilization is limited due to the presence of polluting heavy metals (Cr, Ba, V, etc.) that can be dangerous to the human health and the environment. This research studies the chemical, mineralogical, and the environmental impacts of some Egyptian EAF slag with different cooling rates.

The environmental impacts of the studied slag showed that all heavy metals except vanadium and chromium was found to be very low. From the leaching test the reuse or the uncontrolled dumping of the air cooled Egyptian EAF slag without a previous treatment can have negative effects on the environment in the long term. The cooling rate affects significantly the leaching of Cr, without a significant relation of the original slag Cr content.

From the boiling test it was cleared that the percent of Free CaO and/or MgO in Egyptian EAF samples was negligible (~ 0) so that they have volume stability to be used as construction materials.

Keywords : Environmental impacts, Electric arc furnace slag, Leaching test

INTRODUCTION

Management of the natural resources and wastes are the most important goals which generate opportunities while decreasing the environmental disturbance. Preventing the depletion of natural resources and enhancing the

usage of waste materials has become a challenge to the scientist and engineers (Reinhart, 1993). The huge quantities of waste and/or by-products were produced every year. With the increasing need to recycle these industrial by-products to protect the environment, there is a successful trial to utilize large quantities of such by-product in construction projects (Ismail and Belal, 2014).

Many waste products from several industries are occurring. The most one used in this study is slag waste materials. The names of slags are based on the furnaces from which they are generated. Slags classified into two types namely, blast-furnace slag and steel-furnace slag. Steel-furnace slag was classified into basic oxygen furnace (BOF) slag, electric arc furnace (EAF) slag and ladle slag.

The steel production in Egypt was increased in the last years to reach 6.7 million tons in 2013 that resulted in an alternative increase in the steel slag, at the same time the utilization scale of the steel slag in Egypt are very limited that make the remained steel slag follow an illegal disposal methods such as dumping in old mines or nearby lands. By such methods, steel slag might pollute the natural environment (Radwan, 2015).

About 90% of the whole weight of steel slag is made of Iron, calcium, aluminum, magnesium and siliceous oxides. From a mineralogical point of view steel slag is mainly made of Larnite, Wustite and Brownmillerite . Other compounds are anhydrous calcium silicates and silico-aluminates, gehlenite, bredigite, magnetite, magnesio-ferrite, and manganese oxides (Agostinacchio and Olita, 2005).

EAF slag used in this study is a by-product generated after the melting and the primary acid refining of liquid steel. Its chemical composition is based on its content of calcium, iron and silicon oxides in a global amount of over 80% aluminum, magnesium, manganese and phosphorus oxides. EAF slag has a chemical composition close to that of the cement clinker compared to the ground granulated blast furnace slag (GGBFS) (Amin et al., 2015).

One of the most significant environmental impacts when handling the iron and steel slags is leaching of soluble components from the slag matrix, when these materials are in direct contact with soil or exposed to rainwater. Therefore, the knowledge about the chemical composition of leachates and their behavior is environmentally relevant (Miller and Donahue, 1998).

Radwan (2015) studied the metal leaching of the Egyptian EAF slag by a tank leaching test using distilled water and weak acid water in order to represent materials coming into contact with both fresh water and/or acid rains. The study showed that some metals like Cd, Pb, Cu, Ni and Mn were not detected either in the case of distilled water or using weak acidic water. On the other hand another group of metals has a low or moderate detection like Zn, Cr and Fe. Some metals like V, Ca, Mg, Al, and Si have a high detection frequency and hence the leaching test result showed that except Cr, V, Al metals and also the pH value of the EAF slag the levels of metals in EAF slag were lower than the limits of Egyptian law No. 48/1982 (Radwan (2015).

The type of the iron and steel slag cooling must be taken into consideration where rapid cooling of slag by water can result in an amorphous slag, encapsulating metals and oxides, and thereby lowering the

solubility of the heavy metals compared to rock material used in road making (Tossavainen and Forsberg, 2000). The Water quenching of steel slag results in products with a high content of glassy material. If the cooling is rapid, the slag passes from a liquid state to a solid without development of a crystalline structure. Also the study concluded that by controlling cooling conditions, the mineral transformation and consequently the solubility of elements like chromium, vanadium and other heavy metals were occurring (Tossavainen *et al.*, 2007).

MATERIAL AND METHOD

1. Materials Processing: Three samples of EAF slag were obtained from different steel making plants in Egypt S1, S2 and S3 (air cooled EAF slags). The water cooled slag (S4 and S5) were prepared by remelting and quenching of S1 and S2 in a water tank. Where the re-melting of the EAF was carried out in a magnesium oxide crucible system, the time for melting the slag was approximately eight hours after the electrical furnace temperature raise to 1600°C. In this study as shown in [Fig. 1], where a Carbolite electrical furnace model RHF 17/25 with maximum operating temperature 1700°C was used. The liquid slag was poured into a water tank containing water with a continuous water flow in and out to discard the hot water from the tank bottom. The generated slag granules were collected at the bottom of the water tank at the end of this process. The duration of the water cooling process and granulation was approximately a few seconds duration.

2. Chemical composition: The overall chemical composition of the studied slags was analyzed by X-ray fluorescence (XRF) for major and trace elements using a Philips XRF instrument (Model PW 2404).



Fig. 1: The re-melting process using magnesium oxide crucible and Carbolite furnace.

3. The mineralogical composition: The crystalline phases were identified by X-ray powder diffraction (XRD), which carried out on a PANalytical X-Ray diffraction equipment model X'Pert PRO with secondary monochromator, Cu-radiation.

4. The scanning electron microscopy

SEM was carried out For S1 sample (air cooled EAF slag) and S4 sample (water cooled EAF slag) using SEM Model Quanta 250 FEG (Field Emission Gun) attached with EDX Unit (Energy Dispersive X-ray Analyses) with accelerating voltage 30 K.V., magnification 14x up to 1000000 and resolutions for Gun.1n (FEI Company, Netherlands).

5. Alkalinity test (pH): 10 g is taken and diluted in 75 ml distilled water by stirring. The pH value of the solution is noted with a pH meter after 1 hour, 2 hours and 24 hours and the average pH was calculated.

6. Leaching Test: The method DIN 38414-S4 (Deutsche Norm, 1984) was the compliance test method used to evaluate the performance of the slags. In this test, 100 g of the dry mass of the residue (with a particle size < 10 mm) is mixed in 1000 ml of distilled water (pH=6.92), and submitted to an upside down agitation, in a rotary agitator, during 24 h at room temperature. The solid and liquid phases are separated by a filter of 0.45 μm of pore size, and the liquid phase is then analyzed. The elemental analysis of leachates, in duplicate, was analyzed by ICP in a 720 ICP-OES, Agilent Technologies. The leachates of the five EAF slags S1, S2, S3, S4 and S5 were analyzed in order to monitor relevant parameters such as heavy metals (Al, Fe, V, and Cr, As, Pb, Cd, Cu, Hg, Ni and Zn).

7. Volume stability: In order to evaluate the expansive nature of slag the volume stability was carried out by performing Boiling test where for each EAF slag sample a twenty pieces with an average diameter (50-150 mm) and placed in a boiling water bath for 8 hours. After this time the loss in mass of the slag pieces was determined. By performing the Boiling test free CaO and/or MgO will react with water as a liquid, thus an expansion test has been carried out to reflect the slag volume stability (Radwan, 2015).

RESULTS AND DISCUSSION

The chemical composition of the five EAF slag samples presented in [Table 1]. The comparison between the five Egyptian EAF slag samples, Japanese slag and European slag showing that, significant concentrations of Fe_xO_y , CaO, SiO_2 , Al_2O_3 , MgO and MnO were detected in the Egyptian EAF slag under the study. The average of each group (Air and water cooled

groups) was also represented in the [Table 1]. The representative Egyptian EAF slag main constituents are CaO, Fe₂O₃, and SiO₂ ranging from (33.5-38.3%), (29.8-41.33%) and (12.07-15.1%) respectively. Water soluble chloride ion content was negligible in the studied samples (< 0.01%). Moreover, [Table 2] referring to the environmental impact, high concentrations of Cr, V and Sr were observed; all the other elements investigated had negligible concentrations.

From the results of IB₂, M_b and pH that presented in [Table 3] it was clear that the EAF slag is an Alkaline by product with pH ranging from 11.1 to 11.8, and the samples S4 and S5 (water cooled EAF slag) have a Basicity indexes (IB₂ and M_b) higher than those S1, S2 and S3 (air cooled EAF slag). The results in the increased of the basicity index result in a more hydraulic slag (Amin et al., 2014).

From the XRD chart [Fig. 2] and [Table 4], the identified main minerals of the 3 air cooled EAF slag and the 2 water cooled are Larinite (β -Ca₂SiO₄) and Wustite (FeO). On the other hand Magnetite and Mayenite are characteristic in water cooled EAF slag [Fig. 2] and [Plate 1].

In [Fig. 2], the chart of S1 (air cooled EAF slag example) showing that the major minerals are Larnite and a Wustite-type solid solution. Also trace amounts of Wollastonite, Gehlenite, Monticellite, Srebrodolskite, and Brownmillerite are also observed. In [Plate 1] (A) showing small amounts of Cr. S4 chart (water cooled EAF slag example), the main phase is the Wustite and Wayenite phase. Also Larnite phase and Magnetite phase are constituted in minor amounts. From the SEM and EDXs results [Plate 1] (C), there are also many small grains of Cr-rich oxide. S5 chart (water cooled EAF slag

example) showing that the Wustite and Mayenite are still abundant with minor amount of Larnite phase. From the XRD diffraction pattern the absence of Gehlenite was noted in the water cooled EAF slag.

Table 1: The chemical composition of the 5 EAF slag sample the major element (%)

Oxide	Air cooled EAF slag Samples			Water cooled EAF slag mSamples		European EAF slag *	
	S1	S2	S3	S4	S5	Low MgO	High MgO
SiO ₂	14.2	12.87	12.07	13.89	12.31	8-17	25-35
Al ₂ O ₃	8.1	6.61	7.12	8.2	6.44	4-7	4-7
CaO	38.3	36.9	33.5	38.41	36.63	30-40	25-35
Na ₂ O	0.01	0.03	< 0.01	< 0.01	0.01	—	—
K ₂ O	0.02	< 0.01	< 0.01	0.01	< 0.01	—	—
MgO	4.2	2.52	2.36	4.1	2.42	4-8	8-15
Fe _x O _y	29.8	36.61	41.33	29.92	37.66	18-28	20-29
MnO	2.8	2.67	1.2	2.77	2.81	< 6	< 6
TiO ₂	0.44	0.34	0.43	0.41	0.31	—	—
P ₂ O ₅	0.31	0.21	0.23	0.33	0.18	< 1.5	< 1.5
Cl	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	—	—
SO ₃	0.1	0.07	0.06	0.07	0.04	—	—
LOI	0	0	0	0	0	—	—

* : Radwan (2015)

Table 2: The chemical composition of the five Egyptian EAF slag sample the trace element unit by (ppm).

Element	Air cooled EAF slag Samples			Water cooled EAF slag Samples	
	S1	S2	S3	S4	S5
V	690.7	436.4	184.2	640.4	420.7
Cr	11687.6	5627.8	10947.0	10474.7	5421.3
Ni	10.7	19.2	17.2	11.1	18.2
Cu	260.5	191.1	210.3	250.9	187.4
Zn	316.3	160.6	109.4	309.8	147.8
Co	120.4	99.2	100.3	107.1	89.4
Sr	1027.1	475.4	680.8	1007.8	472.2
Y	7.4	6.1	7.1	6.5	5.6
Zr	137.1	200.6	159.2	137.3	178.8
Nb	76.5	56.8	61.1	77.0	49.7
Ba	110.4	160.1	178.3	108.7	124.2
La	31.5	32.8	28.6	27.0	28.7
Pb	129.1	141.2	74.5	125.1	140.5

Table 3: IB2, Mb and pH of the 5 EAF slag samples with an average for air cooled group and water cooled group.

Item	Air cooled EAF slag Samples				Water cooled EAF slag Samples		
	S1	S2	S3	Average	S4	S5	Average
IB2 [*]	2.7	2.87	2.78	2.78	2.77	2.98	2.87
Mb ^{**}	1.91	2.02	1.87	1.93	1.93	2.08	2.00
pH	11.2	11.4	11.1	11.23	11.5	11.8	11.65

* :IB2 = CaO/SiO₂,

** :Mb= (CaO + MgO)/ (SiO₂ + Al₂O₃)

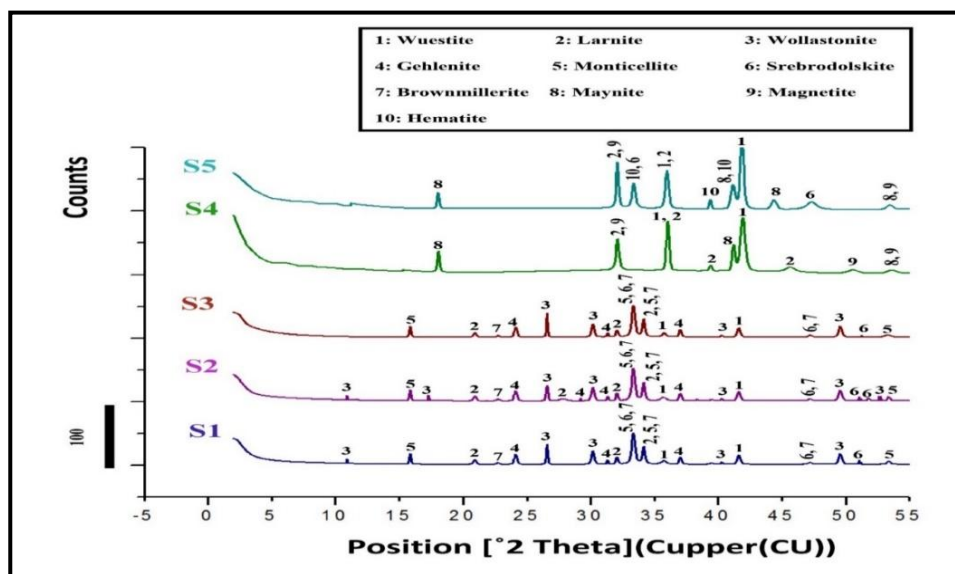


Fig. 2: XRD patterns of the investigated five Egyptian EAF slags at different cooling conditions.

Table 4: The mineral constituents of the five Egyptian EAF slag.

Item	Formula	S1	S2	S3	S4	S5
Wustite	FeO	Y	Y	Y	Y	Y
Larnite	Ca ₂ SiO ₄	Y	Y	Y	Y	Y
Wollatonite	CaSiO ₃	Y	Y	Y	X	X
Gehlenite	Fe ₂ SiO ₄	Y	Y	Y	X	X
Monticellite	CaMgSiO ₄	Y	Y	Y	X	X
Srebrodolskite	Ca ₂ Fe ³⁺ ₂ O ₅	Y	Y	Y	X	Y
Brownmillerite	Ca ₂ (Al,Fe) ₂ O ₅	Y	Y	Y	X	X
Maynite	Ca ₁₂ Al ₁₄ O ₃₃	X	X	X	Y	Y
Magnetite	Fe ₃ O ₄	X	X	X	Y	Y
Hematite	Fe ₂ O ₃	X	X	X	X	Y

Y : The mineral is present

X : The mineral is absent

From S4 and S5 XRD charts [Fig. 2], the clear broadening of the peak width were a result of the rapid water cooling of the EAF slag, suggested that the clear broadening of the peak width may be caused due to a decreased in the crystallite size (Suryanarayana and Grant, 1998).

The volume stability in [Table 5] was carried out using boiling test for all the five Egyptian EAF slag samples. The results range from 0 to 0.03 %, proving that the percent of the free CaO and/or free MgO is very low or

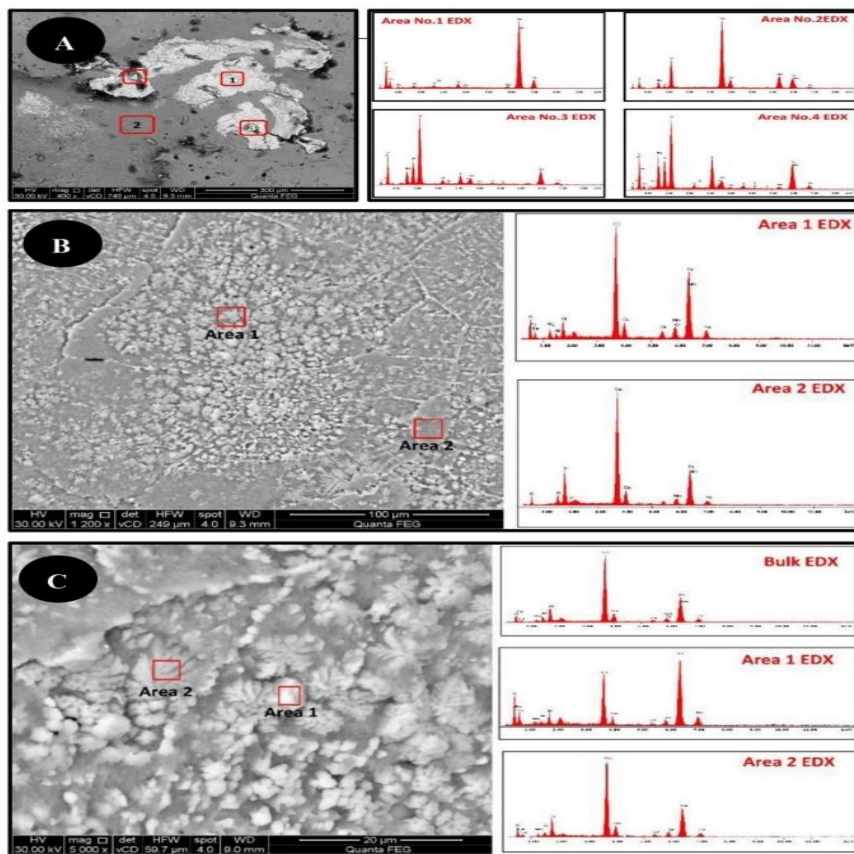


Plate 1: Micrographic appearance of slag samples where (A) represents the air cooled sample S1 and (B) and (C) represent the water cooled EAF slag sample.

approximately not founded that confirmed by XRD pattern [Fig. 2], where there was no presence of CaO and/or MgO peaks. Hence the Egyptian EAF slag under the study have a volume stability.

Table 5: Result of volume stability (boiling test)

Item	S1	S2	S3	S4	S5
Mass Loss (%)	0.03	0.01	0.02	0	0

The leaching characteristic of Egyptian EAF slag, in the present study was evaluated using DIN 38414-S4 method (Gomes and Pinto, 2006) using distilled water. Although the five Egyptian EAF slags (Air cooled and water cooled groups) show similar chemical compositions [Tables 2 and 3] and a similar basicity indexes IB2 or Mb with small differences as shown in [Table 3]. The obtained results of leaching tests are presented in [Table 6] was completely different.

Gomes and Pinto (2006) reported the information that, except for Cr, the leaching of other elements is not relevant in terms of environmental impact.

In the present study the leaching test results [Table 6] showing that, except Cr, V and pH of leachate, the leaching of other heavy metals are found to be very low. Vanadium metal has a high detective frequency. It is detected in the five EAF slag samples ranging from 0.007 to 0.03 ppm. The air cooled slag S1 release the greatest quantity of Cr (627 ppb), and when comparing these results with [Table 7] it was clear that the leachate Cr concentration was well above the limit of the Egyptian law No. 48/1982 and also the limit prepared by Italian regulation for the recovery of steel slags. The leaching of Cr is significantly smaller for water cooled EAF slag S4 (11 ppb) and

practically zero (< 5 ppb) for S5. The higher Cr leaching of S1 (627 ppb) could be explained due to its greater Cr content (11687.6 ppm). However the same hypothesis has no signification for S4, which represent a high content of Cr (10474.7 ppm) and the lowest Cr release (< 5ppb). It seems to be clear that the cooling rate affects significantly the leaching of Cr, modifying the phases in the slags and their solubility, without a significant relation of the original slag Cr content.

pH values using distilled water (pH=6.9) of leachate from the studied samples ranged from 11 to 11.7 Although the results of pH values are shown to be well above the limits of the Egyptian law no. 48/1982, and it was clear that the pH level is very near to be corrosive to aluminum or the galvanized steel pipes with direct contact to the slag. And so the uncontrolled dumping of the air cooled and/or the water cooled of the Egyptian EAF slag without a previous treatment can have negative effects on the environment in the long term due to its high leachate pH.

Table 6: Chemical composition results of V, Cr, As, Pb, Cd, Co, Hg, Ni and Zn (ppm) and pH in the leachate resulted from leaching test according to DIN 38414-S4 for the five EAF slag samples

Element (ppm)	Air cooled EAF slag Samples			Water cooled EAF slag Samples	
	S1	S2	S3	S4	S5
V	0.01	0.03	0.01	0.009	0.007
Cr	0.627	0.285	0.418	0.011	< 0.005
As	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Pb	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Cd	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Cu	0.31	0.22	0.29	0.11	0.09
Hg	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Ni	0.12	0.18	0.17	0.04	0.07
Zn	0.27	0.19	0.18	0.2	0.11
pH	11.2	11.5	11	11.4	11.7

Table 7: Leaching limit values according to Council Decision 2003/33/EC (Gomes and Pinto, 2006) and the Egyptian law (No. 48/1982) limits (Radwan, 2015)

Element (ppm)	Leaching Limit Values			Egyptian Law limit		
	Inert	Non hazardous	Hazardous	Background	Fresh water	Non fresh water
V	—	—	—	—	—	—
Cr	0.5	10	70	< 0.05	< 0.5	< 0.1
As	0.5	2	25	< 0.01	< 0.1	< 0.1
Pb	0.5	10	50	< 0.01	< 0.1	< 0.1
Cd	0.04	1	5	< 0.001	< 0.03	< 0.003
Cu	2	50	100	< 0.001	< 1	< 0.5
Hg	0.01	0.2	2	< 0.001	< 0.001	< 0.01
Ni	0.4	10	40	< 0.02	< 0.2	< 0.5
Zn	4	50	50	< 0.01	< 1	2
pH	—	—	—	6.5-8.5	6-9	6-9

—: Data not available

CONCLUSION

- Egyptian EAF slag is a basic slag and its chemical composition is closer to the cement clinker.
- The XRD of Egyptian EAF slag reveals the presence of different kinds of calcium silicate in all samples.
- The mineralogical composition of the Egyptian EAF slag was complex for the studied EAF slag samples.
- From the boiling test results and XRD patterns the percent of free CaO and/or MgO were very low , therefor the Egyptian EAF slag under the study have a volume stability and hence it could be utilized as a construction material after studying its environmental impacts.
- The leaching test results have shown that all heavy metals except vanadium and chromium was found to be very low and insignificant in term of environmental impacts.
- Other factors rather than metal concentrations have to be considered when accessing the environmental impact of slags, such as cooling rate.
- From the leaching test the reuse or the uncontrolled dumping of the air cooled Egyptian EAF slag without a previous treatment can have negative effects on the environment in the long term. Thus, the pre-treatment of slag or its stabilization before reuse is recommended, even for low value applications.
- The cooling rate affects significantly the leaching of Cr, modifying the phases in the slags and their solubility, without a significant relation of the original slag Cr content.

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الأثار البيئية لبعض عينات خبث فرن القوس الكهربائي المصري مختلفة معدلات التبريد

[٢]

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المستخلص

يعتبر خبث فرن القوس الكهربائي مخلف صناعي يتشابه في تركيبة الكيمياء مخلفات الأسمت ويتكون أساسيا من معادن الكالسيوم سيليكات. يزداد معدل إنتاج الحديد والصلب في مصر سنوياً مما ينتج عنه زيادة ملحوظة في المخلفات الصناعية والتي يجب أن يتم دراسته أثارها البيئية حتي ينتهي إعادة استخدامها بشكل آمن. يتناول متن البحث دراسة جزئين رئيسيين:

١. دراسة التركيب الكيمياء والمعدني لخبث فرن القوس الكهربائي المصري في حالتها الطبيعية.
٢. دراسة بعض الخواص البيئية للخبث المستخدم مثل:
 - درجة الحموضة
 - درجة ثبات الحجم
 - إمكانية الإرتشاح

وفيما يلي بعض الاختبارات المعملية المختلفة التي تمت علي العينات الممثلة لأفران القوس الكهربائي المصرية:

خواص الخبث:

أ- تحديد الخواص الكيميائية والمعدنية لعينات مختلفة من الخبث مقسمة لمجموعتين (مجموعة تم تبريدها ببطيء في الهواء والأخرى تم تبريدها بسرعة وبشكل مفاجئ باستخدام الماء) وشملت التركيب الكيميائي باستخدام الشعبة السينية الطيفية والتركيب المعدني باستخدام حيود الأشعة السينية الحيودية والميكروسكوب الإلكتروني الماسح.

ب- دراسة الخواص البيئية لبعض أنواع خبث أفران القوس الكهربائي المصرية بإجراء الاختبارات التالية:

- درجة الحموضة.
- اختبار الغليان (لمعرفة ثبات الحجم).
- اختبار الإرتشاح.

وخلصت الدراسة باستخدام مجموعتين من خبث أفران القوس الكهربائي انها مخلف صناعي قلوي. ويمكن استخدامه في الأعمال الإنشائية نظراً لثبات حجمه. كما لوحظ بأن معدل التبريد يآثر في إمكانية الإرتشاح لعنصر الكروم. وعند تعرض خبث أفران القوس الكهربائي المصرية للتبريد السريع فإن معدل رشح الكروم يقل بدرجة ملحوظة و يكون في المعدل المسموح به في القانون المصري رقم ٤٨ لسنة ١٩٨٢.