

The Leaching Behavior of Zinc Oxide in Medical Waste of Dental Clinics

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ABSTRACT

Zinc Oxide is an indispensable substance in the field of dental treatment. It is used daily and intensively in all governmental and private dental clinics, leading to the disposal of very high concentrations of zinc with waste and eventually in landfill sites as a final destination for solid waste removal. This indicates the urgent need to investigate its behavior upon disposal due to the surrounding conditions. Approximately 4195 g of mixed dental waste samples were collected from (17) healthcare centers in Baghdad Al-Karkh. The leaching behavior of ZnO powder was investigated through batch reactors using makeup dental solid waste samples. The ZnO leaching was tested with 3 conditions; acidic, alkaline, and Ionic Strength (IS). The acidic condition was considered the most hazardous condition compared with basic and salinity due to the increasing tendency of ZnO to release Zn ion within waste leachate. In solutions with low pH, the dissolution of ZnO tends to produce zinc ions due to the attack of the ZnO surface by proton. In solutions of pH more than 9, the degradation of zinc oxide produces hydroxide complexes. Increasing (IS) decreased zinc concentration in leachate samples with time by promoting solids aggregation, decreasing the repulsive forces of ZnO particles, and accumulating in the bottom of reactors.

Keywords: ZnO, Dental waste, Acidic effect, Metal oxide.

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سلوك راسح أكسيد الزنك في النفايات الطبية لعيادات الأسنان

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الخلاصة

أكسيد الزنك مادة لا غنى عنها في مجال علاج الأسنان، تستعمل بشكل يومي ومكثف في جميع عيادات الاسنان الحكومية والخاصة مما يؤدي لطرح تراكيز عالية جداً من الزنك مع النفايات وبالنهاية تصل الى مواقع الطمر كمصير نهائي لها مما يجعل التحقيق في سلوكها بعد التخلص منها حاجة ملحة. تم جمع ما يقارب من 4195 غم عينة من نفايات عيادات الاسنان الطبية من خلال زيارة (17) مركز رعاية صحية في جانب الكرخ في بغداد. تم فحص سلوك راسح مسحوق ZnO من خلال المفاعلات الدفعية باستخدام عينات حقيقية من النفايات الصلبة لعيادات الأسنان التي تم جمعها وخلطها وتصنيفها وتقطيعها. تم فحص راسح ZnO عن طريق أخذ عينات من الراسح في الظروف الحامضية والقاعدية والمرتفعة القوة الأيونية (IS). تعد الحالة الحامضية أكثر الظروف خطورة مقارنة بالقاعدة والملوحة بسبب زيادة ميل أكسيد الزنك لإطلاق أيون الزنك مع راسح النفايات في المحاليل شديدة الحموضة، وبسبب ذوبان أكسيد الزنك في الظروف الحامضية يزداد تركيز الزنك في الراسح مع مرور الزمن نتيجة تحلل ZnO إلى Zn^{+2} من بسبب هجوم البروتون المباشر على سطح ZnO. في المحاليل المائية ذات الرقم الهيدروجيني الأكبر من 9، يرتبط انحلال ZnO بهيدروكسيده الذي ينتج معقدات مثل $Zn(OH)_2$ و $Zn(OH)_3^{-1}$ و $Zn(OH)_4^{-2}$ في الظروف القاعدية. عند مضاعفة القوة الأيونية للمحلول وعدم تغيير الاس الهيدروجيني يقل تركيز الزنك مع الزمن في الراسح نتيجة تعزيز عملية تجميع المواد الصلبة بتقليل قوى التناثر بينها وتراكمها في قاع المفاعلات.

الكلمات الرئيسية: أكسيد الزنك، نفايات عيادات الأسنان، التأثير الحامضي ، اكاسيد المعادن.

1. INTRODUCTION

The ZnO is an important material used in dental therapy for the temporary filling, implantology, periodontal, and orthodontics fields (**Moradpoor et al., 2021**). ZnO as a powder has good features, including high selectivity, solid antimicrobial activity, biocompatibility, enhanced cytotoxicity, and easy synthesis (**Jabbar, 2020**). As a result of extensive uses of zinc oxide in the field of dentistry as an indispensable material, the fate and transport of zinc upon release to the environment need to be investigated, as one of the most commonly known toxic heavy metals that can affect public health and environment (**AL-Saedi and Ibrahim, 2019**). Although heavy metals are usually discharged to the surrounding environment in low concentration, they may be highly elevated through bio-magnification to levels that begin to exhibit toxic characteristics (**Ebrahim and Alhares, 2015**).

Many researchers were accentual that the ZnO hazardous could be imputed to the dissolution process, which is considered more toxic than other metallic oxides due to the ability of ion-shedding (**Vimercati et al., 2020**). A series of processes happened to ZnO, such as biological degradation, sedimentation, and dissolution, that modify its fate and might form potentially toxic compounds and metal ions (**Poynton et al., 2019**). Many factors have



effects on zinc oxide behavior in waste; the two most important factors are pH and ionic strength (IS) (Alameen and Majeed, 2020), which are needed to study the stability and aggregation behavior of ZnO (Wang et al., 2020).

(Bian et al., 2011) concluded that ZnO dissolution in solutions with highly acidic values tends to produce Zn⁺² from attacking the ZnO surface by direct proton. In solutions of pH more than 9, ZnO dissolution produces many hydroxides (OH⁻¹) complexes forms.

(Bolyard, 2012) studied in his thesis the effect of Zinc Oxide (ZnO) on biological processes at the landfill by examining the interactions between landfill leachate components and ZnO. He observed that the relatively high dissolved solids concentration and ionic strength in leachate decreased the propensity of ZnO to dissociate and promoted the potential to aggregate solids. (Sakallioglu et al., 2016) investigated the leaching behavior of municipal solid waste by determining whether ZnO leaching or remaining within the waste by studying samples grabbed out of a landfill site. They found no significant effect on ZnO leaching in acidic, alkaline, and SI condition and the concentration tend to reduce due to retained ZnO within the solid waste. This was attributed to the municipal solid waste MSW capacity to buffer pH and the complex nature of fresh MSW leachates. (Vimercati et al., 2020) found that ZnO Nanoparticles are insoluble in neutral solutions. On the contrary, it is pretty soluble in an acid environment.

To the author's knowledge, no specific studies can be found in the literature that studied the ZnO's leaching potential and final fate within the medical and dental waste. Thus, this study investigates the leaching behavior of ZnO presented in the dental waste stream through an acid, base, and ionic strength (IS) environment.

2. EXPERIMENTAL WORK

2.1 Materials

All tests are held in the Central Environmental Laboratory–Iraq Ministry of Environment. Approximately (4195) gm of mixed medical and dental waste were collected from dental rooms of (17) different healthcare centers visited in Baghdad Al-Karkh, including 10 health centers and 7 specialized dental centers. Commercial powder ZnO and NaCl were purchased from local chemical stores. NaOH and H₂SO₄ were used as well in pH-adjusting processes. All tests were held in the Central Environmental Laboratory–Iraq Ministry of Environment.

2.2 Preparation of Samples

2.2.1 Percentage of waste compositions

Samples were divided manually into six groups, and each was weighed. The weight percentage of each component was calculated as presented in Fig.1, and waste classifications depend on the Basel convention (Basel convention, 2017)

2.2.2 Bulk Density Calculation

The bulk density of waste was measured by putting the solid waste sample into known volume container V1 until it overflowed. Weights before and after filling were recorded. Density was calculated to be 60.5 kg/m³ as shown in Eq. (1) (Tchobanoglous and Kreith, 2002):

$$\text{Bulk density} = \frac{W_2 - W_1}{V_1} \quad (1)$$

where:

V_1 : volume of empty container, $\text{cm}^3 = 15 * 15 * 16 = 3600 \text{ cm}^3$

W_1 : weight of container before filling (empty container), gram = 40 g

W_2 : weight of container after filling = 258 g

$$= \frac{(258 - 40) \text{ gm}}{3600 \text{ cm}^3} = 0.060555 \text{ gm/cm}^3 = 60.5 \text{ kg/m}^3$$

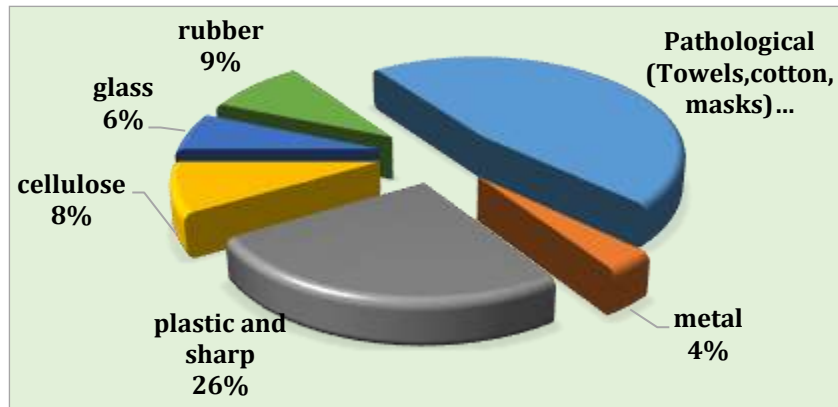


Figure 1. Main Components Percentage Distribution of Dental Solid Waste

2.2.3 Moisture Content Estimation

The moisture content of medical and dental wastes was determined to be 11.2 % by placing the sample in the furnace at a temperature of 100 °C for 5 hours (Altin et al., 2003). The sample was weighted as discarded, represented as (A), and weighed after heating to represent the dry weight as (B). The weight loss was expressed as the moisture content percentage and is determined using equation Eq. (2):

$$\text{Moisture content} = \frac{(A - B)}{A} * 100 \quad (2)$$

where:

A = 50 g, and B = 44.4 g

So, the moisture content = $\frac{(50 - 44.4)}{50} * 100 = 11.2 \%$

2.2.4 Heavy Metals Analysis

The 17 samples were homogeneously mixed and shredded manually to be one sample for laboratory tests.

Dental solid waste was analyzed for heavy metal concentration using an Atomic Absorption device (nova 300). The analysis included zinc (Zn^{+2}), Ferrous (Fe^{+2}), lead (Pb^{+2}), cadmium (Cd^{+2}), chromium (Cr^{+2}), copper (Cu^{+2}), and nickel (Ni^{+2}) ions. Preparing of sample and analysis procedures were held in real samples according to the manual of the laboratory



International Center for Agricultural Research in the Dry Areas (ICARDA) (Ryan et al., 2001).

2.3 The Batch Experiments

The leaching behavior of powder ZnO was investigated through batch reactors using dental solid waste samples. The 6 batch reactors were filled with 20-gram solid waste and 400 mL deionized water (DI) with a ratio of (20:1), according to the American Society for Testing and Materials batch test (ASTM D 1193). It recommends determining the leachability of solid inorganic ingredients using shredded waste material up to less than 10 mm with water and an L/S ratio of 20:1 at 30 rpm (Townsend et al., 2003).

As the experiments progressed, the moisture amount in the reactors decreased with time. No moisture was recommended to add instead of the collected samples to avoid the system dilution.

The experiment was held in (6) reactors (2) for each condition). The first reactor was spiked with a known concentration of ZnO stock solution. The second was the blank reactor "No ZnO concentration was spiked." The stock solution was freshly prepared by adding (25) mg of ZnO to (1000) ml of deionized water (DI) to obtain a (25) ppm solution, followed by a sonication process of 30 min (Sakallioğlu et al., 2016).

The required amounts of stock solution were spiked to the reactors. After several trials, it was found that the appropriate amount of ZnO Stock solution was 20 ml which gave an acceptable concentration of zinc ions to start the experiment. As the research was not conducted to study the removal of Zn ions but to study the stabilization of that ion, a spike of ZnO was strongly needed for all tested media to increase zinc ions detection and to give clear readings and accurate explanations.

The behavior of ZnO leaching was tested under three different conditions: (i) acidic condition at pH=5.5, (ii) basic conditions at pH=9.5 (iii) increased ionic strength (IS), as shown in **Table 1**. In two IS reactors, the samples were doubled from their natural value of 7.15 mM to 14.5 mM using a stock solution of NaCl (1 N). pH indication was undertaken by (HACH) laboratory instrument.

Table 1. Experimental Conditions of the Leaching Behavior of ZnO.

No.	Conditions	Spiked Zn concentration (mg/l)	Adjusted pH	Average pH	Initial IS (mM)	Doubled IS (mM)
1	Acidic	Zero	5.5	6.9	None	
		25	5.5	6.7	None	
2	Basic	Zero	9.5	8.3	None	
		25	9.5	8.1	None	
3	IS	Zero	none	7.6	7.15	14.5
		25	none	7.9	4.8	9.6

At the beginning of each batch test, the pH was adjusted using (1 N) H₂SO₄ and (1 N) NaOH solutions. The acidic condition was at (5.5), and the basicity was at (9.5). Since pH values were normally changed from acid to alkaline during waste stabilization in landfills, the pH or IS was only initially adjusted because the batch tests were closed system operation, and there was no adjustment during the test. As the reactors were prepared, a laboratory shaker

was used to agitate the reactors for 72 h (Sakallioğlu et al., 2016). 10 mL of leachate samples were drawn out of every reactor to be analyzed for Zn^{+2} concentration, pH, and conductivity. In total, 11 leachate samples at 0, 1, 2, 3, 4, 12, 24, 36, 48, 60, and 72 hr., totaling 66 representative leachate samples plus ten samples as blanks, were analyzed throughout the whole experiment. The experimental reactors are illustrated in Fig. 2.



Figure 2. The experiment batch of six reactors and samples

3. RESULTS AND DISCUSSION

Zn ions had the highest concentration dramatically compared with the rest metals concentrations. It's worth mentioning that (Pb^{+2}) and (Cr^{+2}) had nil concentration, as shown in Fig. 3.

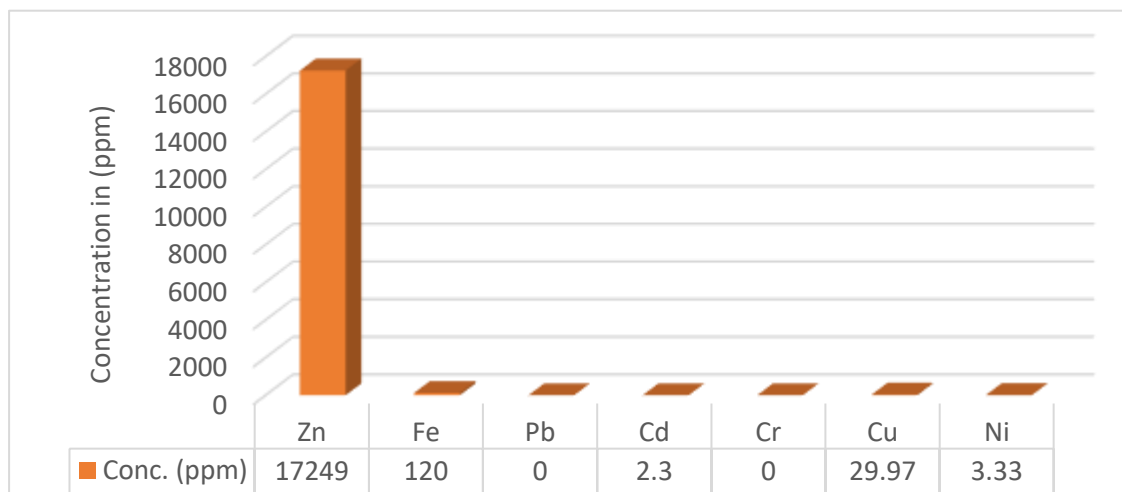
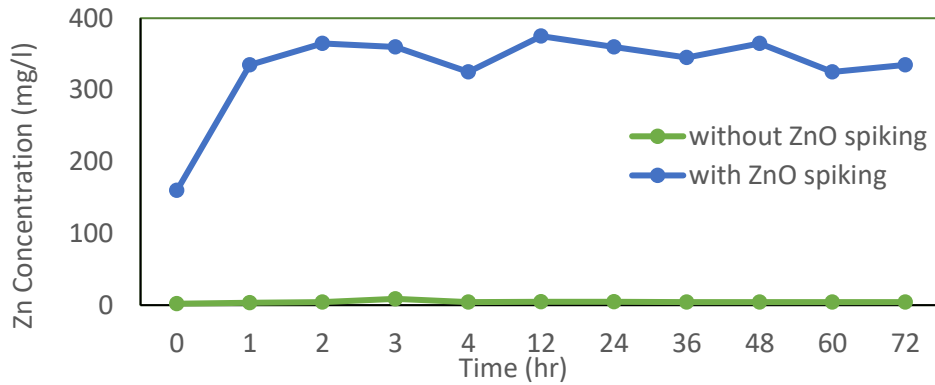
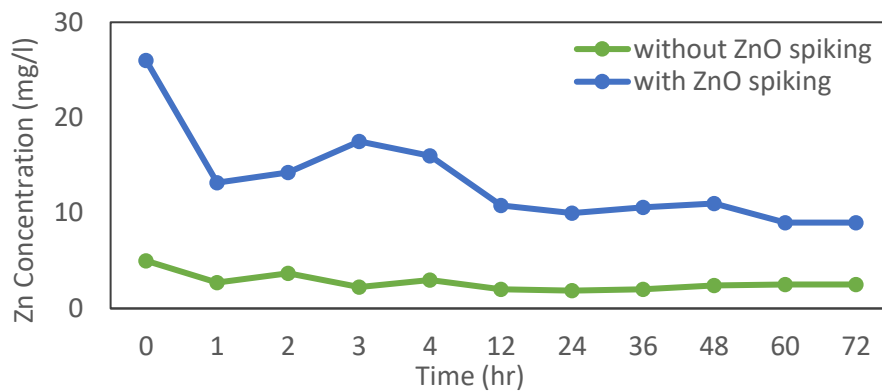


Figure 3. Heavy Metals Concentration in Dental Waste.

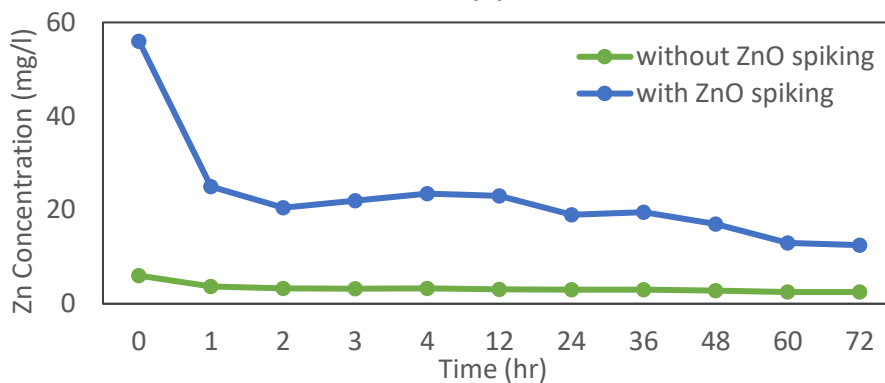
Zn ion concentration results were presented in Fig. 4 A, B, and C as a function of time during the 6 batch reactors in acidic, alkaline, and IS conditions, respectively. The concentration in three conditions had been stable after 24 hr., but the sampling processes continued for 72 hr. for more certainty.



(A)



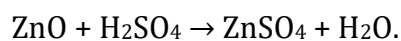
(B)



(C)

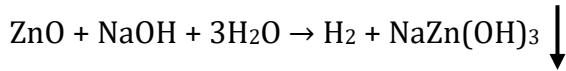
Figure 4. Zn ions concentrations in leachate samples as a function of time at (A) Acidic, (B) Alkaline, and (C) IS conditions

In acidic conditions, Zn ions concentration increased with time till the ions reached its stability, as shown in **Fig. 4A**. This behavior can be explained by the tendency of Zn ions to leach more at low pH values due to direct proton attack of the ZnO surface. Where ZnO was detached out of waste material by dissolution process and dissolved in the acid solution in addition to the added ZnO powder (**Haynes et al., 2016**):



Which corresponds with (Bian et al., 2011; Vimercati et al., 2020) but disagree with (Sakallioğlu et al., 2016).

In basic conditions, Zn ion concentration decreased with time till the concentration reached its stability, as shown in Fig. (4, B). This may be explained as most of ZnO was either retained within the waste matrix or had settled down to the bottom of the flask as a result of a heavy metal composite (Haynes et al., 2016)



which is quite close to the findings reported by (Bian et al., 2011; Sakallioğlu et al., 2016).

In IS conditions, Zn ion concentration decreased with time until the concentration had been stable, as shown in Fig. 4C. Results showed that Zn ions concentration in IS conditions rapidly decreased at the experiment beginning, followed by a slight decrease with time. This trend can be explained by the rapid sedimentation of zinc oxide with increased ionic strength. This promotes solids aggregation, potentially decreasing ZnO tendency to dissociate but accumulating in the bottom of the reactors.

The surrounding's electrical double layer (EDL) is compressed in high ionic strength, decreasing the repulsive forces of ZnO particles and leading to agglomeration, which corresponds to (Bian et al., 2011; Bolyard, 2012).

Zn ions concentration in blank samples (DI + waste) was about 1.4 mg/l and remained within this concentration level throughout the experiment with no significant changes, as shown in Fig. 5.

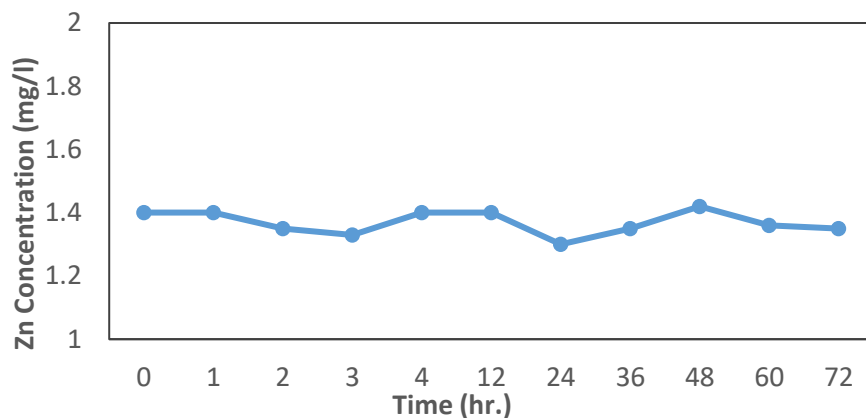


Figure 5. Zn ions concentrations in the blank sample

4. CONCLUSIONS

Dental clinics increase zinc pollution significantly by disposing of solid waste containing ZnO. The dissolution of ZnO produces zinc ions in acidic conditions (pH = 5.5) where ZnO is detached from waste material. In contrast, the zinc concentration tends to decrease in basic conditions and highly IS. The degradation of zinc oxide produces hydroxide complexes in alkaline solutions (pH = 9.9) such as $\text{Zn}(\text{OH})_{4(\text{aq})}$, $\text{Zn}(\text{OH})_{3(\text{aq})}$, and $\text{Zn}(\text{OH})_{2(\text{aq})}$. Sedimentation of zinc oxide occurs in high IS and pH between 6 and 9 due to promoting solids aggregation, which precipitate out of solution.



The acidic condition would have been considered the most hazardous condition compared with basic and salinity due to the increasing tendency of ZnO to release Zn ions within waste leachate.

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