

Characterization of Lead Leachability from Cathode Ray Tubes Using the Toxicity Characteristic Leaching Procedure

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Cathode ray tubes (CRTs) in television and computer monitors are one of the most common components of discarded electronics in the solid waste stream. CRTs present a disposal problem because of their growing magnitude in municipal solid waste (MSW) and their role as a major source of lead in MSW. Using the EPA Toxicity Characteristic Leaching Procedure (TCLP), lead leachability from CRTs was studied. Lead leached from the CRT samples at an average concentration of 18.5 mg/L. This exceeded the regulatory limit of 5.0 mg/L. Several factors affected the lead concentrations of each CRT sample. These included the sample fraction of the CRTs, the particle size used in the tests, and the CRT type. The most significant quantities of lead were obtained from the funnel portion of the CRTs at an average lead concentration of 75.3 mg/L. The major source of lead in the funnel is the frit seal of color CRTs. Samples containing the frit seal had lead leaching levels nearly 50 times those without. Samples comprised of smaller particle sizes exposed a greater surface area resulting in higher lead leaching levels. While 21 of 30 color CRTs exceeded regulatory lead limits, none of the six monochrome CRTs did. Age of the CRTs was not a significant factor for lead leaching. These results provide useful information to the regulatory and waste management community for developing policies for managing discarded CRTs.

Introduction

The management of discarded electronics is an issue of concern to solid waste management professionals. In 1996, the computer and electronics industry comprised 11% of the gross domestic product and was growing at an annual rate of 4%, with computer sales growing 15% annually (1, 2). Cathode ray tubes (CRTs) in televisions and computer monitors are one example of discarded electronics now recognized as a disposal problem. In 1996, there were over 300 million existing CRTs (TVs and monitors) in North America. Meanwhile, in that same year, 42 million new CRTs were sold in the U.S., and 79 million computers were retired (3).

The rapid development of computer technology has resulted in frequent consumer replacement of computer monitors. It is estimated that for every three new computers

TABLE 1. Lead Content in Various CRT Glass Components by Mass (4, 6)

glass	color CRT (%)	monochrome CRT (%)
panel	0-3	0-3
funnel	24	4
neck	30	30
frit	70	N/A

purchased, two currently used units will become obsolete. That ratio is expected to increase to 2:1 by 2005 (2). The future transition from analogue to digital high-definition televisions will also result in increased disposal of television CRTs.

CRTs are the technology used in most televisions and computer display screens. A CRT uses high voltages to accelerate electrons toward a luminescent material called a phosphor. The phosphor is deposited on the facepanel and emits light upon excitation from the electrons. The electron guns require a high vacuum to achieve long life; thus the envelope must have sound mechanical integrity to resist the force of atmospheric pressure. The high voltages used to accelerate the electrons must be insulated from the external surfaces. Therefore the envelope must also have excellent electrical insulating properties. The decelerating electrons emit X-rays and the envelope must be a good X-ray absorber. To achieve all of these requirements a lead-impregnated glass is used for the construction of the tube. The lead, added in the form of lead oxide, provides the shielding necessary for the X-rays produced (4, 5).

The internal composition of a color CRT requires an envelope that can be opened for deposition of the phosphor screen and other components. The two halves of the envelope are mated with a high-lead solder glass called the frit. Monochrome tubes for direct view or projection can be made from one-piece bulbs without using the frit glass seal. The lead content of the CRT is predominantly confined to the neck and funnel of the CRT, and the frit seal if used. The industry uses both a lead free and a 2% to 3% lead facepanel composition with a trend toward increasing the use of the no-lead composition. The approximate lead content, by mass, for color and monochrome CRTs is shown in Table 1 (4, 6).

Television and computer CRTs present a disposal problem because of their growing magnitude in the waste stream and their role as a major source of lead in municipal solid waste (MSW). Lead's toxic effects are known, specifically its effects upon the development of children. The leading source of lead in MSW is lead-acid batteries, comprising 138,000 tons and 65% of lead discards in 1986. Without recycling, batteries would contribute up to 700,000 additional tons of lead. Consumer electronics accounted for 27% of lead discards in MSW in 1986 and are projected to make 30% of lead discards by 2000 (7). CRTs are the main source of lead in electronics. By 2000, CRTs are projected to contribute 29.8% of all lead in MSW (98.7% of lead from electronics) (7).

Consumer electronics are not recycled to the same large extent as lead-acid batteries. Instead, management of discarded electronics, including CRTs, takes place through the traditional methods of municipal solid waste (MSW) management: landfilling and incineration. When disposed in landfills, increased concentrations of heavy metals in landfill leachate may occur. When discarded electronics are disposed at waste-to-energy facilities, the heavy metals become concentrated in the ash, limiting disposal and reuse options. Thus CRTs are now being targeted for removal from

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the MSW stream and for subsequent recycling (8). On April 1, 2000 Massachusetts banned all CRTs from landfills.

The management options and requirements for solid wastes in the U.S. depend largely on whether the solid waste is characterized as hazardous. The Toxicity Characteristic Leaching Procedure (TCLP) is the regulatory method required when determining whether a solid waste is hazardous from leaching of hazardous pollutants (9). CRTs have been anecdotally referred to as failing the TCLP for lead, but the results of TCLP analysis are not available in the scientific literature (3, 10–12). While a number of problems have been cited with the TCLP in regard to its true representation of environmental conditions (13), the test has been found in recent work to leach many heavy metals (including lead) in a manner similar to domestic landfill leachate (14), the intended result of the test.

This paper reports the results of a study examining lead leachability from CRTs using the TCLP. The objectives of the research were to determine if CRTs exceed the 5 mg/L toxicity characteristic concentration for lead and to examine several factors that impacted lead leaching (particle size, sample mass, sample location). The objectives did not include any attempt to characterize the actual environmental impact under different disposal scenarios. Regardless of whether the TCLP truly reflects environmental conditions encountered by CRTs upon disposal, the classification as hazardous does have an impact on how CRTs may be managed in the current U.S. regulatory system. If CRTs are truly a hazardous waste as often anecdotally cited, regulators would have additional options to require removal from the waste stream. Since the cost of hazardous waste management is much greater than MSW management, recycling becomes a more cost-effective alternative. Regulations to encourage their reuse and recycling, such as the universal waste rules, could be applied (15).

Methods and Materials

Experimental methods included preparing the CRT samples, conducting the TCLP, and leachate analysis. Two separate leaching experiments were performed: Experiment 1 and Experiment 2. Experiment 1 was the initial investigation of lead leaching from CRT glass samples using the TCLP. Experiment 2 examined the effect of particle size (large fraction vs small fraction), sample heterogeneity, sample mass, and the frit on the lead concentration in the TCLP leachate.

Experiment 1. Sample Preparation. Over 10 weeks, televisions and computer monitors were collected from individual donations, electronics repair facilities, an electronics manufacturer, and institutional electronics disposal. To observe any changes in TCLP leachable lead levels with age, collected monitors and televisions were grouped into three categories by date of manufacture: 1988 and earlier, 1989 to 1993, and 1994 to 1998. Eleven to thirteen CRTs were collected from each group, utilizing televisions and computer monitors. The brand of each computer monitor or television was recorded. Following disassembly, the CRT manufacturer was also recorded.

Each CRT was divided into three sections to compare lead leachate concentrations of each CRT section. The sections consisted of the neck, the funnel, and the facepanel (Figure 1). After carefully breaking the glass seal at the cathode connection point to release the tube vacuum, the sections were scored using a diamond tipped rotary cutting tool. The neck was scored two to three centimeters below the point it flared. The funnel was scored between the frit seal (color monitors) or support frame (monochrome) and the facepanel. The score was tapped with a screwdriver and hammer to cause the CRT to break along the scored lines. The mass of the complete CRT, the neck, and the funnel were recorded.

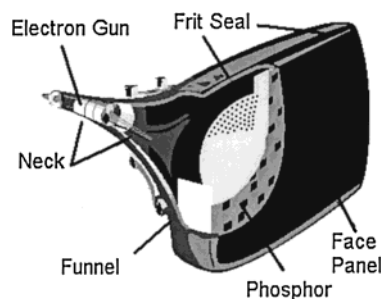


FIGURE 1. Sample locations of CRTs.

The mass of the facepanel was computed by subtracting the funnel and neck mass from the total mass.

Leaching Tests and Analysis. Once divided, each section was reduced in size as required by EPA SW 846 Method 1311, the Toxicity Characteristic Leaching Procedure (16). Each section was tested separately (i.e. the neck, funnel, and facepanel were analyzed individually). A portion of each section (two or three pieces between 200 and 500 g) was placed in a stainless steel bowl. The glass was covered by a cloth for protection and manually crushed with a standard hammer. Intermittently, the crushed glass was separated through a 9.5-mm sieve and the remaining large fraction returned to the bowl for further crushing. Unused glass portions were retained for later testing.

One hundred grams of each size-reduced CRT sample was then loaded into an extractor bottle (high-density polyethylene (HDPE)). To determine the appropriate extraction fluid for the TCLP test, a preliminary test was performed to measure the pH of the CRTs samples (5 g of CRT sample:96.5 mL of reagent water) (16). Since the pH of all samples was less than 5, TCLP extraction fluid #1 was selected. Two thousand grams of extraction fluid (5.7 mL of glacial acetic acid in 500 mL reagent water per 64.3 mL of 1 N sodium hydroxide solution, diluted to a volume of 1 L) with a pH of 4.93 ± 05 was added to the extraction vessel. The sample was rotated at 30 rpm for 18 ± 2 h in a 12 vessel rotary extractor (Analytical Testing Corporation). The extract was filtered through a glass fiber filter of 0.8- μ m pore size and the sample preserved using 2 mL of nitric acid per 500 mL of sample. The extracts were stored at 4 °C until digestion. EPA method 3010A (Acid Digestion of Aqueous Samples and Extracts for Total Metals for Analysis by FLAA or ICP Spectroscopy) was used to digest the samples (16). The digested samples were analyzed to determine lead concentration using Flame Atomic Absorption Spectrophotometry (Perkin-Elmer Model 5100 AAS). EPA method 7420 was used to analyze the digested samples (16).

Experiment 2. Experiment 2 was conducted to examine the variability of lead concentrations observed during Experiment 1. First, the effect of sample composition including particle size and sample heterogeneity on lead leaching from the CRT samples was investigated. Second, three different sample masses were used to explore the effect of sample mass on lead leaching. Sample preparation, the TCLP leaching test, and the analytical methods in Experiment 2 were the same as those in the Experiment 1 (unless otherwise noted).

Examination of Particle Size Effect. To measure the impact of particle size and CRT funnel heterogeneity on lead leaching levels, the funnel fraction of three CRTs from Experiment 1 was selected for additional testing. In Experiment 1, the three CRTs chosen possessed different funnel lead concentrations (high, moderate, and low levels). All remaining portions of the funnels not crushed in Experiment 1 were crushed and sieved into two size fractions, 4.75 mm to 9.5 mm and 4.75 mm and smaller. By including the entire funnel a more representative sample was achieved

TABLE 2. Summary of TCLP Leachable Lead Concentrations for All Samples

maker	TV/MON	color/ mono	year man.	tube maker	leachable lead concentration (mg/L)			
					neck	funnel	face	weighted av
Acer	MON	C	93	Panasonic	9.5	347.3	<1.0	57.2
Digital	MON	M	90	Clinton	4.2	<1.0	<1.0	<1.0
Elite	MON	C	92	Chunghwa	9.7	81.2	<1.0	19.3
Emerson	TV	C	84	Goldstar	6.5	6.6	<1.0	1.5
Gateway	MON	C	93	Toshiba	9.0	9.2	<1.0	3.2
Gateway	MON	C	92	Toshiba	12.8	174.5	<1.0	54.1
Hp	MON	M	84	Matsushita	<1.0	<1.0	<1.0	<1.0
Hp	MON	M	85	Matsushita	1.7	<1.0	<1.0	<1.0
IBM	MON	C	87	Matsushita	9.5	38.4	<1.0	9.4
IBM	MON	C	89	Panasonic	9.5	142.9	<1.0	41.5
IBM	MON	M	92	Phillips	1.1	<1.0	<1.0	<1.0
Imtec	MON	C	89	Samsung	8.2	200.6	<1.0	60.8
Imtec	MON	C	89	Hitachi	13.6	403.6	<1.0	85.6
Memorex	MON	C	97	Toshiba	10.1	103.0	<1.0	21.3
Memorex	MON	C	97	Kch	12.7	49.4	<1.0	15.4
Memorex	MON	C	98	Samsung	7.0	25.7	<1.0	6.1
Memorex	MON	C	98	Chunghwa	10.9	7.8	<1.0	2.3
Memorex	MON	C	97	Toshiba	8.4	34.9	<1.0	9.1
Memorex	MON	C	98	Samsung	7.1	7.1	<1.0	2.2
Memorex	MON	C	97	Chunghwa	8.3	35.3	<1.0	10.6
NEC	MON	C	87	NEC	11.3	50.3	<1.0	10.7
Orion	TV	C	96	Orion	9.1	132.5	<1.0	33.1
Panasonic	TV	C	84	Matsushita	22.4	11.8	<1.0	3.5
Quasar	TV	C	84	Quasar	13.6	182.4	<1.0	43.5
Samsung	MON	M	89	Samsung	<1.0	<1.0	<1.0	<1.0
Seiko	MON	C	87	NEC	9.1	100.0	8.0	26.6
Sharp	TV	C	94	Sharp	8.7	16.4	<1.0	4.4
Sharp	TV	C	84	Sharp	7.9	6.0	<1.0	1.5
Tandy	MON	C	85	Sharp	17.6	116.1	<1.0	35.2
Techmedia	MON	C	95	Samsung	<1.0	20.1	<1.0	6.9
Teknika	TV	M	86	Phillips	1.6	<1.0	<1.0	<1.0
Ttx	MON	C	91	Chunghwa	7.5	10.0	<1.0	2.8
Zenith	TV	C	94	Zenith	18.3	198.8	<1.0	54.5
Zenith	TV	C	94	Zenith	15.8	7.1	<1.0	1.6
Zenith	TV	C	77	Zenith	<1.0	97.7	<1.0	21.9
Zenith	MON	C	85	Toshiba	7.5	92.1	<1.0	21.5
av					8.6	75.3	<1.0	18.5

^a Weighted average was calculated based on the total mass of each fraction of the crushed CRT samples in Experiment 1.

than those taken in Experiment 1 that had included only a randomly chosen portion. The two size fractions allowed an examination of particle size while ensuring that the samples continued to meet the requirements of the leaching procedure. For each of the three CRT funnels evaluated, three samples of the large fraction and three samples of the small fraction were extracted and analyzed to check repeatability. This produced six lead leachate measurements per CRT funnel.

Examination of Sample Size. One CRT funnel from Experiment 1 was selected to further investigate the effect of sample size on the variability of lead leaching. This step also examined if the minimum of 100 g mass per sample required by TCLP was appropriate to represent lead leaching in CRTs. All remaining portions of the funnel were crushed and sieved into the two particle size groups. The samples were carefully mixed with a stainless steel scoop and bowl for 10 min. Three different masses of the sample were chosen, 40 g, 70 g, and 100 g. Masses greater than 100 g were not possible due to the volume limitations of the extractor bottles. Three samples of each mass were extracted and analyzed using the same solid-to-liquid ratio (1:20 by mass) for a total of nine samples.

Results and Discussion

Lead Leaching of CRTs in Experiment 1. A total of 36 CRTs were processed and analyzed. CRT screen size ranged from 18 cm (8 in.) to 63 cm (27 in.). As shown in Figure 1 each tube was divided into three sections: the neck, the funnel, and

the face. The average glass composition of the CRTs by mass was 4.9% neck, 25.2% funnel, and 69.9% face.

The pH of the leaching solution, an important controlling factor in the leaching of heavy metals from wastes, was measured for each TCLP performed. The leaching behavior of lead is typically characterized by the greatest amount of leaching at low pH values, a minimum leachability observed at pH values in the range of 9 to 10, and an increased degree of leachability at pH values above 11 (14). The change in pH during the TCLP was minor. The initial pH of all TCLP extraction solutions was 4.93 ± 0.05 , and the final pH ranged from 4.80 to 5.20.

Table 2 presents the lead concentrations of the TCLP leachate for all samples tested during Experiment 1. Generally, the highest concentrations of lead were obtained from the funnel fractions. Leachate from these fractions had an average lead concentration of 75.3 mg/L. The average concentration of lead obtained from the neck fractions was 8.6 mg/L. No lead was detected from the face of the CRTs excluding one sample at a concentration of 8.0 mg/L, resulting in an average TCLP lead concentration for all facepanel glass of 0.22 mg/L. Based upon the percentage of glass by weight in each section, a weighted average for each complete CRT was computed. The weighted average TCLP lead concentration of the complete CRTs was 18.5 mg/L. The 99% confidence interval for all CRTs was 9.1 mg/L to 28.0 mg/L. This concentration exceeds the regulatory limit of 5.0 mg/L for TCLP lead (9).

Table 3 provides a summary of results by CRT characteristic. Twenty-one of 30 color CRTs exceeded 5.0 mg/L

TABLE 3. Summary of Results by CRT Characteristic

category	no. of samples	no. of exceeding regulatory limits	av leachable lead concns (mg/L)
all CRTs tested	36	21	18.5
televisions	10	4	16.5
computer monitors	26	17	19.3
CRTs — 1988 and before (color CRTs)	13 (10)	7 (7)	13.5 (17.5)
CRTs — 1989 to 1993 (color CRTs)	11 (8)	6 (6)	29.5 (40.6)
CRTs — 1994 to 1998 (color CRTs)	12 (12)	8 (8)	13.9 (13.9)
color CRTs	30	21	22.2
monochrome CRTs	6	0	<1.0

^a As measured by EPA Method 1311, Toxicity Characteristic Leaching Procedure.

with an average leachate lead concentration of 22.2 mg/L. For color CRTs, the 99% confidence interval was 12.6 to 31.9 mg/L. However, monochrome CRTs did not exceed the regulatory limits; with an average leachate lead concentration below detectable limits.

The TCLP lead concentrations were more variable than originally expected. This variability was especially noted for some CRTs of the same manufacture of the same year. For example, for two CRTs of identical manufacturer, model, and year of manufacture, the funnel section of one leached 7 mg/L, while the other leached nearly 200 mg/L. Due to this variability, the tests of Experiment 2 were necessary to determine potential causes.

ANOVA analysis of the three age groups (1988 and before, 1989–1993, 1993–1998) yielded an *F* value of 3.23 and a *p*-value of 0.0385. Based on the statistical analysis, there was a significant difference between the CRTs from 1989 to 1993 and the other two age groups. However, no significant changes in CRT construction were found during these years. Instead, the difference is more likely due to sample heterogeneity or variability in the sample composition.

Sample Heterogeneity. A substantial cause of variability identified by Experiment 2 was sample heterogeneity resulting from the frit seal of color CRTs. During Experiment 2, two samples from a CRT funnel containing the frit seal were compared with two samples containing the glass solely. The leachate lead concentration of the funnel samples containing the frit (492 mg/L, 575 mg/L) were nearly 44 times more than the samples containing the glass only (10.8 mg/L, 13.3 mg/L). Thus, when sampling a CRT, the amount of the frit contained in the sample makes a large difference in the measured lead level.

The effect of the frit on leachate lead levels was observed in several aspects of Experiment 1. The funnel, which is comprised of 24% lead for color CRTs and only 4% lead for monochrome CRTs (Table 1), would be expected to leach lower amounts of lead than the neck which is comprised of 30% lead for both types. In Experiment 1 this was true for all monochrome CRTs; however, for color CRTs it was true for only 4 out of 30.

The frit seal of color CRTs results in higher funnel lead leachate values, causing the color CRTs, unlike monochrome, to exceed 5 mg/L. The frit seal contains a large amount of lead. Color CRT funnels in Experiment 1 that contained a portion of the frit seal would result in lead concentrations higher than the neck samples. Monochrome CRTs, lacking the frit seal, had leachate levels from the neck higher than those from the funnel. During Experiment 1, the effect of the frit on leachate values was unknown. Therefore, no effort was made to standardize the amount of frit in the funnel

samples. Thus variations in the amount of frit in a sample would cause large variations in measured lead leachate concentrations. Inclusion of a portion of frit is theorized as the cause of the single face panel sample with a measurable lead level (8.0 mg/L).

Throughout Experiment 2, it was noted that CRT funnels that had displayed low lead leachate levels in Experiment 1 produced higher concentrations in Experiment 2. In Experiment 1, samples were derived from a random portion of the CRT funnel. This sampling method produces heterogeneity between funnel samples. Some samples may contain larger portions of the high lead frit than others, thus causing a difference in lead leachate levels. Experiment 2 samples were derived from the entire funnel and thus were more likely to contain similar amounts of the frit. The lack of inclusion of the frit in color CRT samples in Experiment 1 is hypothesized as the reason that 9 of the 30 color CRTs did not exceed the 5 mg/L toxicity limit. Lead leachate tests of Experiment 2 show that well-mixed representative samples of all color CRTs surpass the toxicity limit when the frit seal is included.

Particle Size Effect. Another contributing factor to the variability in lead leaching levels observed in Experiment 1 is particle size. The results of the particle size study (large size vs small size) conducted in Experiment 2 are displayed in Figure 2. All three CRT funnels tested displayed higher lead leachate levels for smaller particle sizes than for larger particle sizes. When more surface area was exposed due to the smaller particle size of the samples, more lead leached from the samples. This demonstrates an inability of the leaching solution to penetrate the CRT glass.

The variability in measured lead leachate concentration was greater for large particle size samples than for small particle size samples. The relative standard deviations for the small particle samples were 53.4%, 20.9%, and 35.9%. For large particle sizes of the same CRTs, the relative standard deviations rose to 57.2%, 40.1%, and 73.8% respectively. Thus small particle sizes promote a more homogeneous sampling method and provide greater precision in measurement.

Sample Mass Effect. The results of the particle size testing continued to show variability even among triplicate measurements. Despite sieving to more uniform particle size and inclusion of the entire funnel to develop a more representative sample, measurements continued to display noteworthy variability. The sample mass was tested as a factor in obtaining a representative sample.

TCLP requires a minimum of 100 g of sample. A 100 g sample is placed in 2000 g of extraction fluid in a 2-L extraction vessel (1:20 ratio by weight). In Experiment 2, three different sample masses (40 g, 70 g, and 100 g) were used to test the effect of sample mass and particle size on lead concentrations in TCLP leachate. The same solid-to-liquid ratio of 1:20 was maintained for all samples. The results reconfirm the effect of sample heterogeneity on lead leachability. The larger the sample mass chosen, the greater precision between samples was obtained. This is demonstrated in Figure 3. As the sample mass was increased, the relative standard deviations of the results decreased. It is expected that sample masses greater than 100 g will provide more homogeneous samples, lower standard deviations, and more repeatable results. Ideally, samples would contain a large percentage of the total mass of the CRT crushed to a uniform size.

The results also support the previous particle size testing. For all 40 g, 70 g, and 100 g samples tested, smaller particle size samples (<4.75 mm) yielded lead leachate levels two to four times higher than samples using larger particle sizes (4.75 mm to 9.5 mm). Again, a greater surface area results in greater lead leaching demonstrating that the leachate solution has limited penetrability of the CRT glass.

CRT Disposition. Conclusions beyond those stated above in regard to the implications of the lead leaching from CRTs

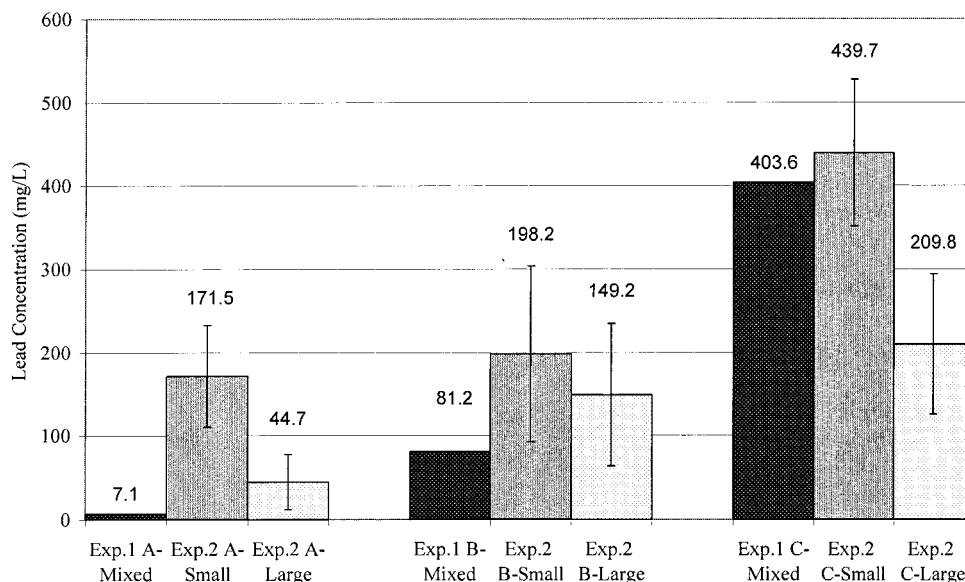


FIGURE 2. Particle size effect on lead leaching.

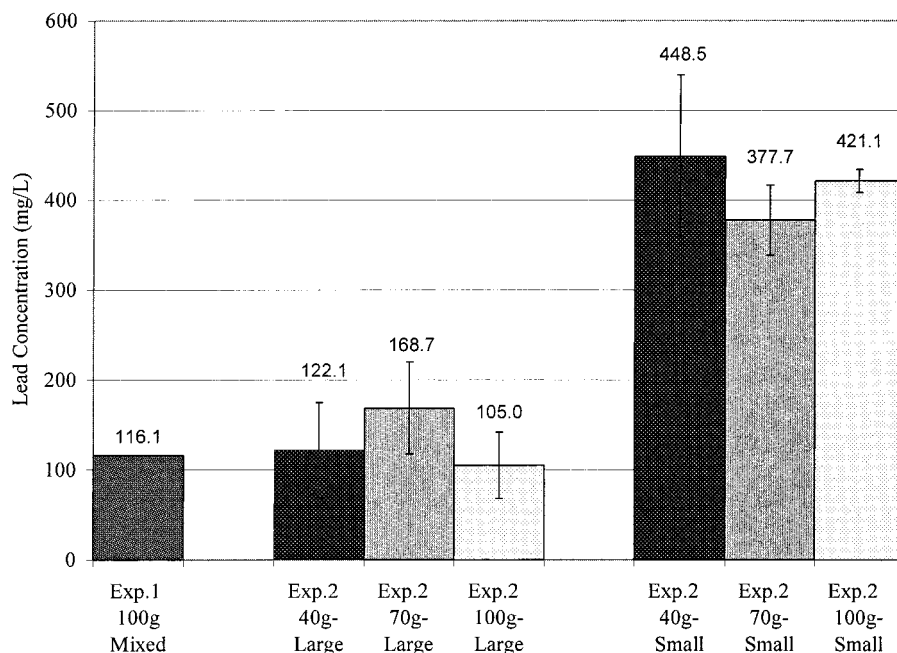


FIGURE 3. Sample size effect on lead leaching.

were not the objective of this research. The fact that the TCLP test may not represent the true condition of CRTs upon disposal was not an issue of discussion in this research. TCLP is the required regulatory test. Other leaching tests, such as the synthetic precipitation leaching procedure (SPLP), would also provide valuable information regarding the leaching of lead from CRTs because the pH of the leaching fluid may play a significant role.

Since monochrome CRTs do not fail the TCLP test, they are not considered hazardous waste; therefore, their disposal does not have to be managed as such. These CRTs are still considered solid waste. Although 9 of the 30 color CRTs were also less than regulatory lead levels, 21 of 30 samples did exceed regulatory levels. Therefore, color CRTs found in computer monitors and televisions may exceed the regulatory levels for lead given in Title 40 CFR 261.24 definition for the toxicity characteristic. These CRTs should be considered hazardous waste, and their disposal should be managed accordingly.

The magnitude of CRTs being disposed will increase in the future and appropriate management of these devices needs to be addressed. Anecdotal references to CRTs failure of TCLP are no longer necessary. The results of this study remove all doubts as to whether color CRTs exceed the hazardous waste characteristic level for lead using the TCLP. Color CRTs as a hazardous waste will now require significantly higher costs for disposal than previous simple MSW methods of incineration or landfilling. The increase in disposal costs may generate an increased demand for recycling and reuse of CRTs. Additionally, special regulatory treatment of CRTs, such as inclusion in the Universal Waste Rule, would further enhance CRT recycling by further reduction in handling costs.

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