

The Release of Cadmium to the Environment Due to Corrosion from Anthropogenic Sources

Hugh Morrow
International Cadmium Association

Introduction

Cadmium is a naturally occurring metallic element, one of the components of the earth's crust and present everywhere in our environment. However, in certain forms and in high concentrations, cadmium may produce adverse human health and environmental effects, and therefore it has been the subject of considerable research and regulation over the past 50 years. Cadmium emissions to the environment may arise from both natural and anthropogenic or man-made sources. Estimates of the proportion of total cadmium emissions due to natural sources have ranged from as low as 10% to as high as 50% (Van Assche 1998, Garrett 1995, Cook and Morrow 1995). Some of these natural emission sources include weathering and erosion of parent rocks, volcanic activity and forest fires (WHO 1992, OECD 1994, Nriagu 1980, Niargu 1989).

Anthropogenic sources of cadmium releases include products and materials to which cadmium has deliberately been added to impart a specific chemical, mechanical or physical property and products or materials in which cadmium is naturally present as a residual or impurity element. In general, the cadmium content of the products or materials in which cadmium is present as a deliberate additive are greater than the levels in the products or materials in which cadmium is present as a residual or unintentional impurity. However, the amounts of the intentional uses of cadmium are far less than the amounts of the unintentional uses of cadmium. For example, even though the cadmium level in NiCd batteries is higher than the cadmium levels in phosphate fertilizers, the amounts of fertilizers utilized are many times higher than the amounts of NiCd batteries produced.

Anthropogenic Sources of Cadmium

Intentional Uses of Cadmium - Because of its unique chemical, mechanical and physical properties, cadmium is utilized deliberately in a number of important industrial and consumer applications. These include nickel-cadmium (NiCd) batteries for both industrial and consumer uses; cadmium corrosion-resistant coatings on iron, steel, aluminum and titanium; cadmium pigments in plastics, glasses, ceramics, enamels and artists' colors; weathering and ultraviolet light stabilizers for polyvinyl chloride (PVC); as an alloying element in soldering, brazing, electrical contact, high electrical conductivity and high thermal conductivity alloys; and in electronic compounds such as cadmium telluride and cadmium sulfide employed in solar cells, detectors, electronic gates, switches, sensors and relays. The approximate cadmium levels or ranges of cadmium contained in these various products are summarized in Table I.

Of these deliberate additions, most are cadmium compounds. In nickel-cadmium batteries, the cadmium-containing materials are cadmium metal and cadmium oxide in the charged state and cadmium hydroxide in the discharged state. In cadmium pigments, the compounds utilized are based on cadmium sulfide with additions of either zinc or selenium to produce a broad spectrum of colors from orange to yellow to red. Cadmium carboxylates, such as cadmium laurate and cadmium stearate, are normally used, in conjunction with barium sulfate, as stabilizers in PVC. Electronic cadmium compounds are usually based on the sulfide or the telluride. Only in the two

cases of cadmium coatings and cadmium alloys are the materials utilized truly metallic materials. Thus, the bulk of the discussions on corrosion of intentional cadmium uses will focus on coatings and alloys.

Unintentional Presence of Cadmium in Other Products – Cadmium is a naturally-occurring constituent in a wide range of materials which are utilized for industrial and consumer purposes. It is sometimes found in high concentrations in phosphate fertilizers since some marine phosphates and phosphorites have been reported to contain cadmium levels as high as 500 ppm (WHO 1992, Cook and Morrow 1995). It has also been reported at fairly low levels in iron and steel because all of the raw materials (iron ore, limestone, coke and scrap steel) utilized to make iron and steel usually contain residual amounts of cadmium. Virtually all fossil fuels contain various levels of cadmium depending on their specific type, and are a significant contributor to cadmium air pollution since they all must be combusted to be utilized. The raw materials employed to produce cement and concrete also contain residual amounts of cadmium and are a contributor to total cadmium environmental emissions. Cadmium is present as a residual element in zinc, lead and copper ores. However, cadmium is normally removed as a by-product during the smelting and refining processes for these metals and converted directly to cadmium metal for sale into the market. It is not generally left in zinc, lead or copper metals and alloys unless it is deliberately done to achieve an improvement in some mechanical, chemical or physical property. The tendency, in fact, in recent years has been to produce these nonferrous metals, their alloys and their compounds with lower and lower residual cadmium levels. Furthermore, deliberate cadmium additions to zinc, lead or copper alloys are generally quite low (<1%), and even in these cases they are almost never utilized for atmospheric exposure applications. Residual or unintentional cadmium levels in various products or materials are summarized in Table II.

Corrosion of cadmium from unintentional impurities in metallic materials such as irons and steels and nonferrous metals is expected to be extremely low and not to contribute significantly to overall cadmium releases due to corrosion. The presence of cadmium in and its emission into the environment from fossil fuels, cement manufacture or wear, and fertilizers do not arise from corrosion processes but represents chemical dissolution or wear/erosion processes rather than electrochemical corrosion processes.

Corrosion of Cadmium

General Considerations – Cadmium is one of several metallic coating materials which are electrochemically active and which are therefore used as sacrificial coatings to protect iron and steel from corrosion. Sacrificial coatings for iron and steel include cadmium, zinc and aluminum. Cadmium has been utilized as a coating material on aluminum and titanium alloys as well. In addition to providing good corrosion resistance, cadmium coatings possess excellent lubricity for frictional applications, good solderability, superior electrical and thermal conductivity and compatibility with aluminum alloys. Metals with strong electropositive potentials will dissolve or be sacrificed when coupled with a metal with a more electronegative electrode potential in the presence of a conductive medium.

When both cadmium and iron are exposed to corrosive media, cadmium will preferentially dissolve to form cadmium ions which then react with oxygen in the corrosive media or atmosphere to form an adherent thin cadmium hydroxide layer. This layer is soft and malleable, does not readily spall, and thus diminishes further corrosion unless it is disturbed by mechanical factors. If abrasion or disruption of the cadmium hydroxide film occurs, then the process begins all over again. Thus, cadmium is protective of iron and steel by both a sacrificial corrosion mechanism and a cadmium hydroxide film barrier corrosion protection mechanism.

In general, cadmium coatings are utilized where a combination of good corrosion resistance is required along with some other property such as a low coefficient of friction (fasteners), low electrical conductivity (electronic connectors and housings) or compatibility with aluminum (aircraft). They also have good corrosion fatigue properties and resistance to stress corrosion cracking, making them valuable in high strength steel fasteners utilized in the aircraft industry. Cadmium coatings are often used where good corrosion resistance to marine or salt atmospheres is required. For example, it has been used in the truck and automotive industry on steering columns and disk brakes for safety applications to ensure freedom from seizure due to corrosion products from road salt. Cadmium coatings are employed in some shipbuilding applications because of their high resistance to sea salt, and are employed in railroad and ordinance applications because of their excellent frictional characteristics combined with good corrosion resistance. An estimated breakdown of the USA cadmium coatings market by application from 1989 through 1998 is shown in Figure 1.

Corrosion Rates of Cadmium - The corrosion rate of cadmium varies considerably with the pH of the corrosive medium and with the absence or presence of certain other chemical species. In the acidic ranges (pH <5.0), the corrosion rate of cadmium increases rapidly and therefore cadmium coatings are normally used for applications where the intended service is to be in the neutral to alkaline regime. Corrosion rates also depend to a great extent upon the presence of other chemical species, such as chlorides or carbonates, oxygen concentration, the presence of carbon dioxide, and other factors. Corrosion rates of cadmium increase rapidly as pH value decreases and acidity increases. The protective cadmium oxide/hydroxide film is continually dissolved at low pH values and accelerated corrosion occurs. Corrosion rates for cadmium do not markedly increase, however, in the very high pH or alkaline ranges.

The corrosion behavior of cadmium coatings is normally expressed in several different ways. The corrosion rate may be expressed directly as a thickness loss or penetration per unit time. Cadmium coating thicknesses are normally 5 μm (0.2 mils), 8 μm (0.3 mils), 12 μm (0.5 mils) or 25 μm (1.0 mils) thick. Corresponding corrosion rates would therefore be micrometers per year ($\mu\text{m}/\text{year}$), mils per year or inches per year (ipy). Corrosion rates may also be expressed as coating weight losses per unit area per unit time for a standard thickness coating. An example of this unit often used is milligrams per square decimeter per day (mdd). Tables are readily available for converting these units (Durney 1984). The corrosion behavior of cadmium coatings is also expressed as the time to the development of various percentages of "red rust" (iron oxide) on cadmium coated steel substrates in either standardized or atmospheric tests. The corrosion rate values give a quantitative rate at which material is lost, while the time to red rust values yield a more practical service life estimate of the cadmium-coated component. It should be kept in mind that the corrosion rate or the appearance of red rust may not always be linear functions of time. If protective oxide films are formed, then corrosion kinetics may slow down to less than linear rates. If other phenomena such as abrasion are part of the service environment, then corrosion kinetics may accelerate due to the destruction of protective films.

One example of corrosion rate data (Meyer 1986) on cadmium in three different atmospheres is shown in Figure 2. As previously indicated, cadmium exhibits generally higher corrosion rates in industrial atmospheres which may be mildly acidic than in neutral suburban atmospheres or marine atmospheres. In these tests, corrosion rates varied from 10.4 μm per year to 1.3 μm per year. In other tests reported in the literature (Carter 1977) a corrosion rate of 25 μm per year was reported for a severe industrial environment in the United Kingdom while a correspondingly lower rate of 5 μm per year was noted for a UK marine environment. Another reference (Shreir

1976) notes a cadmium corrosion rate of 33.3 μm per year for an industrial exposure site in Sheffield, UK, and a 5.25 μm per year corrosion rate in a marine exposure at Calshot in the UK. Thus, industrial exposures consistently appear to result in cadmium corrosion rates in the range from 10 to 30 μm per year, while marine exposures consistently result in corrosion rates of 1 to 5 μm per year.

The other technique for establishing cadmium's corrosion performance is measurement of the time of a certain degree of rust on a test specimen under certain conditions. For example, the U.S. Army Armament Research and Development Center (Farrara 1984) conducted extensive tests on coated metal fasteners for ordinance applications. They exposed various coatings to a standard 5% salt spray test and observed the time to the first appearance (1%) of red rust and the time to appearance of red rust all over the surface (100%). Corrosion tests were performed on both unused fasteners and on fasteners subjected in abrasion by threading into and out of a steel plate three times. The results of these tests are summarized in Figure 3. Red rust appeared on the new fasteners at about 2100 hours and was completed at a time in excess of 4000 hours when the tests were discontinued. Abrasion of the fasteners before testing reduced corrosion lives by about half, although the amount of life reduction would presumably be a function of the amount of abrasion imposed. Assuming that the 100% red rust time corresponds to complete loss of the 0.2 mils (8 μm) cadmium coating, the corresponding corrosion rates of cadmium in these 5% salt spray tests are 17.7 μm per year for the abraded fasteners and 10.7 μm per year for the new fasteners. These corrosion rates are higher, as expected, than those observed in the marine atmosphere tests since the 5% salt spray test is an accelerated and more severe corrosion test.

Extensive atmospheric corrosion testing on cadmium was also conducted on cadmium-coated plain carbon steels in both industrial and marine environments (Sample, Mendizza and Teel 1976). Specimens were electroplated with cadmium coatings varying from 0.05 to 2.00 mils (1.27 to 50.8 μm), exposed to the testing environments, and the times to various degrees of red rust measured. The results of these tests are summarized in Figure 4. As expected, the time to 50% red rust corrosion was observed to be approximately linear with the cadmium coating thickness. Performance of cadmium coatings in the marine atmosphere at Kure Beach, North Carolina in the United States was two to three times better than in the industrial atmosphere of New York, NY which is consistent with the corrosion data reported above. Corrosion rates in the marine atmosphere ranged from 1.23 μm per year to 1.89 μm per year, while those in the industrial atmosphere ranged from 4.33 μm per year to 5.30 μm per year.

Cadmium Emission Levels From Corrosion

Emission Levels Based on Corrosion Data – Corrosion rate data presented above is summarized in Table III along with conversions of the corrosion rate data into cadmium weight losses per unit area of coating based on conversion factors available in the literature (Durney 1984) which specify that a one-mil (25.4 μm) coating of cadmium corresponds to a coating weight of 22.0 milligrams per square centimeter (mg/sq cm). Corrosion rate data in industrial, suburban, and marine atmospheres are shown in units of both micrometers of cadmium coating loss per year and weight per unit area of cadmium coating loss per year. To convert these corrosion weight loss measurements to total cadmium emissions data, one would need to estimate the total surface area of cadmium coatings applied. The problem here is that it is extremely difficult to estimate the total surface area of cadmium coatings in service since such a wide variety of applications are utilized, many of them on thousands of small fasteners, as previously shown in Figure 1.

Emission Levels Based on Measurements – Many direct measurements have been made on cadmium emissions to the environment, but very few have attempted to specifically identify the source as originating with the application or use of cadmium coatings. One study (Elgersma 1992) attempted to specifically identify cadmium emission sources to the Rhine River basin from 1970 through 1988. These results are shown in Figure 5. Between 1970 and 1980, cadmium aqueous emission factors for electroplating were relatively high since cadmium coating was the predominant use for cadmium at that time and control of aqueous effluent discharges from electroplating shops was not well regulated. As the usage of cadmium for coatings decreased and regulatory and technological pollution control advances reduced cadmium electroplating emissions to the environment, the relative amounts of cadmium emitted to the environment from plating sources decreased significantly.

Another study (Nguyen et al. 1994) examined the reduction in heavy metal content to the Greater Montreal, Quebec, Canada sewer system from 1980 through 1993. In the case of cadmium, much of the aqueous emissions were felt to arise from the electroplating industry although the contributions from other sources were not quantified. These results are shown in Figure 6, which again indicate a significant decrease in cadmium emissions during the 1980s. Both of these sets of data appear to indicate that by the 1988-1993 time frame, aqueous emissions from cadmium coatings sources in these two specific areas (Rhine River Basin and the Greater Montreal sewer system) had decreased to levels of less than 1,000 kg per year. The problem here, of course, is that different time frames, different areas, and incomplete separation of the various cadmium sources make it very difficult to accurately gauge the input from cadmium coatings alone.

Emission Levels Based on Consumption and Applications – At least three analyses (Morrow 1996, WS Atkins International 1998, and Corden & Floyd 2001) have attempted to examine cadmium emission levels into the environment from cadmium coatings based on assumptions of the amounts of cadmium consumed for various applications, their service lives in those applications, and the probable corrosion and wear mechanisms in those applications. At the OECD Conference on *Sources of Cadmium in the Environment*, the International Cadmium Association (Morrow 1996) stated that “Depending on the coating thickness, the absence or presence of a supplementary coating, and the service conditions, a cadmium coating normally last from 10 to 25 years. During that time, the amount of cadmium which is lost to the environment through the corrosion process is probably 50 per cent of the original thickness.”

This estimate was further refined (WS Atkins International Ltd 1998) in their report on cadmium pigments, stabilizers and coatings performed for the European Commission. The WS Atkins analysis differentiated between abrasive and non-abrasive conditions for cadmium coatings and concluded that, over the lifetime of the product, about 25 per cent of the cadmium coating would be lost. The WS Atkins study further assumed 195 mt of cadmium consumed in plating applications in the EU in 1996, and thus extrapolated cadmium losses to the environment of 49 mt from plated products which it distributed between the marine and non-marine environments based on their relative areas in Europe. The net result was 23 mt of cadmium distributed to the non-marine environment in the EU according to the WS Atkins analysis.

A further conclusion was made (Corden & Floyd 2001) in the Risk & Policy Analysts Limited (RPA) study on pigments, coatings and stabilizers which was also performed for the European Commission. RPA assumed the same rate of cadmium emissions to the environment as WS Atkins but assumed that a significant portion of cadmium coatings were used on plated fasteners and landing gear in the aircraft industry and that therefore cadmium concentrations should be “relatively greater” around major airports. However, discussions, for example, with authorities at Heathrow Airport near London, UK have not indicated higher than normal cadmium

concentrations which might be associated with corrosion or wear from cadmium aircraft applications.

A more detailed analysis of cadmium coatings applications than those discussed above is now in order to more accurately estimate the amount of cadmium emissions to the environment from coatings applications. First, it must be recognized that use of cadmium for coatings applications is far different in Europe than it is in the United States and Canada. Cadmium coatings usage in these three areas, the only ones for which decent estimates are available, is shown in Figure 7. It is clear that the cadmium product restrictions on coatings under 91/338/EEC have lowered cadmium coatings usage in Europe compared to that in the USA. The relatively low percentage of cadmium consumption in plating shown in Japan is due to the fact that such a high percentage of the world's nickel-cadmium batteries are made in Japan that the amount consumed for plating applications appears insignificant, and that cadmium plating usage in aircraft applications is somewhat lower in Japan than in the United States and Europe.

Furthermore, if the market applications for cadmium coatings in the USA are examined in more detail, it appears as if only a few of them can be considered applications which are continually under abrasive conditions and which therefore would lose significant portions of their coatings during service life as previously discussed by Morrow (1996), WS Atkins International Ltd (1998) and Corden & Floyd (2001). Automotive applications such as steering columns and disk brake parts would be subject to abrasive conditions. Some of the fastener applications would be subject to abrasion, but only on a limited basis. Fasteners might be torqued to a certain applied load level, but during their useful service life only disassembled and reassembled a few times. In the aircraft applications area, landing gear are certainly frequently subjected to abrasion conditions and a heavy cadmium coating is utilized for this application to ensure that landing gear always may be safely raised and lowered during aircraft operation. Ordnance and ammunition applications also result in abrasion and wear as do railroad applications.

On the other hand, the major end use area, communications equipment, does not involve abrasive wear, and cadmium is used here to impart good corrosion resistance plus good electrical conductivity. Similarly, about half of the fastener and more than half of the aircraft applications do not involve abrasive conditions. Based on the market data of Figure 1, it is estimated that perhaps only 30% of the cadmium coatings applications encountered repeated abrasion conditions which would result in substantial coating loss during the component's lifetime. Thus, the cadmium coating's loss to the environment for the EU previously estimated by WS Atkins International Ltd (1998) and Corden & Floyd (2001) should probably be revised downwards from 23 mt per annum to 13.8 mt per annum.

Emission levels based on consumption and applications assume a steady state system. If an average service life for a cadmium plated component is assumed to be 10 years, then the amount of cadmium consumed in electroplating 10 years ago will gradually be released to the environment over those 10 years as will cadmium from components plated before and after that initial date. If cadmium consumption in coatings is decreasing rapidly as it has been in Europe (Figure 7), then cadmium emission or deposition amounts will also decrease but at a time displaced approximately by the service life of the coating.

Comparison of Cadmium Corrosion Emissions with Total Cadmium Emissions

There have been several studies on cadmium emissions to the environment which attempt to allocate the relative contributions from various sources (ERL 1990, Van Assche 1998, Belgium Rapporteur 2002). Many other studies have analyzed cadmium emission levels to certain

compartments and their sources (Jones, Lapp & Wallace 1993), and two risk analyses performed for the European Commission have examined cadmium emissions from electroplating as part of a study on cadmium pigments, stabilizers and coatings (WS Atkins International Ltd. 1998, Corden & Floyd 2001). The Van Assche (1998) analysis indicates that all products to which cadmium has intentionally been added account for only about 2% of total human exposure to cadmium. The draft risk assessment on cadmium and cadmium oxide for use in batteries (2002) indicates that emissions of cadmium to the EU environment from cadmium plating account for only 0.05% of total emissions. In both analyses, phosphate fertilizers, fossil fuel combustion, and iron and steel production and use account for far higher contributions. Similarly, a U.S. EPA study (Jones, Lapp & Wallace 1993) indicates that most cadmium air emissions arise from fossil fuel combustion and iron and steel production, and that only 1.1% of all cadmium air emissions can be allocated to all cadmium products. The ERL Report (1990) similarly indicates that total emissions of cadmium to the European Union environment from the production and use of cadmium coatings is on the order of 1% to 2% of total cadmium emissions although there is no data listed for several applications and compartments and an acknowledgment that double counting is probably involved in totaling emissions to land. Thus, it appears fairly clear that cadmium coatings contribute only a tiny fraction to total cadmium emissions to the environment and to human exposure to cadmium.

Cadmium Corrosion Emissions from Sources Other Than Cadmium Coatings

It is also possible that a limited amount of cadmium emissions might arise from the corrosion of other metals in which cadmium is intentionally or unintentionally present. These would include cadmium-containing alloys such as the brazing and soldering alloys, electrical contact alloys and copper-based electrical and thermal conductivity alloys. They could also include the presence of cadmium as an impurity rather than as an intentional alloying addition in zinc, lead or copper alloys. In the intentional addition category, the relative use of these alloys is now a very small fraction of total cadmium usage, less than 0.5%, their normal areas of application are not for atmospheric exposure, and thus the environmental emissions from these applications would be expected to be extremely small. In the unintentional addition category, the levels of residual cadmium contained in zinc, lead and copper alloys are generally so low as to preclude any significant emissions due to corrosion. Typical residual cadmium levels in copper and lead and their alloys are 0.0001%. While cadmium levels in zinc coatings may be higher, from 0.003% to 0.005%, zinc is electrochemically more active than cadmium and therefore will corrode preferentially, much as it does in the “dezincification” of Cu-Zn brass alloys or Zn-Ni coatings. Thus, it is not felt that cadmium corrosion emissions from sources other than cadmium coatings are significant, and that cadmium corrosion emissions from cadmium coatings are quite small, less than 2% of total cadmium emissions, compared to emissions from all sources.

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