Mercury-Impacted Scrap Metal: Source and Nature of the Mercury

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Abstract

The reuse and recycling of industrial solid wastes such as scrap metal is supported and encouraged both internationally and domestically, especially when such wastes can be used as substitutes for raw material. However, scrap metal processing facilities, such as mini-mills, have been identified as a source of mercury (Hg) emissions in the United States. This research aims to better define some of the key issues related to the source and nature of mercury in the scrap metal waste stream. Overall, it is difficult to pinpoint the key mercury sources feeding into scrap metal recycling facilities, quantify their associated mercury concentrations, or determine which chemical forms are most significant. Potential sources of mercury in scrap metal include mercury switches from discarded vehicles, electronic-based scrap from household appliances and related industrial systems, and Hg-impacted scrap metal from the oil and gas industry. The form of mercury associated with scrap metal varies and depends on the source type. The specific amount of mercury that can be adsorbed and retained by steel appears to be a function of both metallurgical and environmental factors. In general, the longer the steel is in contact with a fluid or condensate that contains measurable concentrations of elemental mercury, the greater the potential for mercury accumulation in that steel. Most mercury compounds are thermally unstable at elevated temperatures (i.e., above 350°C). As such, the mercury associated with impacted scrap is expected to be volatilized out of the metal when it is heated during processing (e.g., shredding or torch cutting) or melted in a furnace. This release of fugitive gas (Hg vapor) and particulates, as well as Hg-impacted bag-house dust and control filters, could potentially pose an occupational exposure risk to workers at a scrap metal processing facility. Thus, identifying and characterizing the key sources of Hg-impacted scrap, and understanding the nature and extent of associated releases, represent a practical research need that is essential for improving the environmental management of Hg-impacted scrap and assessing measures to protect workers from potential health and safety hazards that might be posed by mercury and Hg-impacted scrap.

Keywords: mercury; scrap metal; metal recycling; mercury-impacted metal; mercury switches; oil and gas scrap

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1. Introduction

Mercury (Hg) is widely known as a persistent toxic substance in the environment that has been distributed throughout the globe via atmospheric transport (ATSDR 1999; UNEP 2008). This extremely volatile metallic element is present in air, water, soil, and sediment, as well as in manufactured materials and their wastes. The human contribution to mercury in the environment has been steadily increasing. It is estimated that approximately half of the current atmospheric mercury levels can be attributed to anthropogenic sources (UNEP 2008), with levels increasing approximately 300 to 500% in the past 250 years (Sunderland and Mason 2007). Although combustion facilities, such as coal-fired power plants and waste incinerators, have been identified as the main source of mercury emissions in the United States, other sources include secondary steel production, such as scrap metal processing facilities (AMAP/UNEP 2008).

Currently, the reuse and recycling of industrial solid wastes, such as scrap metal, is supported and encouraged both internationally and domestically, especially when such wastes can be used as substitutes for raw material (UNEP 2014; EPA 2013; EC 2011). Mini-mills that melt and refine steel, typically in an electric arc furnace (EAF), are the largest recyclers of scrap metal in the United States (EPA 2009). The EAF can produce steel from 100% scrap metal feedstock in an energy-efficient and flexible manner.

In the United States, under the Resource Conservation and Recovery Act (RCRA) 40 CFR 261.4(a)(13) (EPA 1980), recycled scrap metal is not considered solid waste; it is excluded from regulation as a hazardous waste and does not require waste characterization as long as it meets the definition of scrap metal as defined by the statute. With the exception of a few states (e.g., California and Massachusetts) that impose stricter regulations on the use of scrap metal, most states follow the RCRA regulations. As a result, the scrap metal stream is not commonly tracked or screened for contaminants (other than radioactivity) or required to comply with U.S. Environmental Protection Agency (EPA) and Occupational Safety and Health Administration (OSHA) requirements related to hazardous waste. The research presented in this paper aims to better define some of the key issues related to the source and nature of mercury in the scrap metal waste stream in order to improve the environmental management of that material.

2. Sources Of Mercury In Scrap Metal

Various studies have indicated that scrap metal processing and recycling facilities could emit measurable amounts of mercury (OSHA 2008; EPA 2008; Sastry et al. 2004; Morgan et al. 2003). However, limitations in the current literature make it difficult to pinpoint key sources of mercury in the scrap metal used as feedstock by these facilities. Researchers who have attempted to correlate mercury emissions with a specific raw material have indicated that neither the entry of mercury into an EAF nor the chemical forms that are most significant in determining potential hazards are well understood (Rosenborough et al. 2006b; Orrling 2010). Some possible sources of mercury in scrap metal include mercury switches, other electronic-based scrap, including household appliances and related industrial systems, and Hg-impacted scrap metal generated by the chlor-alkali and oil and gas industries. These potential sources are discussed below.

2.1. Vehicles

Switches are standard components of automobiles, trucks, and other vehicles, serving the essential function of opening and closing electrical circuits. They can be activated by a change in temperature (e.g., temperature switch), pressure (e.g., pressure switch), or position/motion (e.g., tilt switch, float switch) (MPCA 2001). Because of the inherent properties of liquid mercury, including high electrical conductivity, it has been widely used in hermetically sealed switching devices. Mercury switches are preferred over other switching devices for several reasons, including their reliability (consistent operation throughout their electrical life), high load capability, and moderate cost (Sastry et al. 2004).

Nearly all (99%) of the mercury in vehicles is contained in switches (Woodruff 2004). Most vehicles manufactured from 1960 into the 1990s contain mercury switches. In the early 1990s, European automobile manufacturers stopped using mercury switches in all models. However, U.S. companies did not discontinue this use across all models for another several years, e.g., not until the late 1990s for Chrysler and the early 2000s for Ford and General Motors (Griffith et al. 2001). It has been estimated that as many as 250 million mercury switches were present in vehicles on the road in North America in 2001 (Griffith et al. 2001). Mercury switches in cars were used in various applications, such as convenience lights in trunks and on hoods, four-wheel drive antilock braking systems (ABS), antitheft systems, and headlamps (i.e., high-intensity discharge lamps). A vehicular mercury switch contains around 0.7 to 1.5 grams of mercury (IEPA 2005; Sastry et al. 2004; Woodruff 2004; MPCA 2001; Griffith et al. 2001).

Vehicles that reach the end of their operating life and are discarded are commonly sent to recycling yards, where they are shredded and packed into bundles that are eventually melted at recycling facilities such as steel mini-mills. Thus, most of the mercury from unremoved switches eventually ends up in scrap processing facilities. This mercury can then be released into the environment, presumably as metallic mercury or elemental mercury [Hg(0)], during processing and recycling. Two studies using data from the late 1990s provide some context for the scale of the mercury issue associated with discarded vehicles. The first study from the Steel Recycling Institute estimated that the steel industry recovered and recycled enough steel from old cars to produce more than 13 million new automobiles in 1999, for a recycling rate of 91% (Sastry et al. 2004). The 2012 automotive recycling rate was estimated to be approximately 93% (SRI 2012). The second study used 1997 data from the U.S. Environmental Protection Agency (EPA) to estimate that about 11 tons of metallic mercury waste was generated from car switches in the United States (IEPA 2005). It is noted, that the mercury contribution to scrap metal resulting from vehicle switches should decrease with time as the number of older vehicles containing mercury switches (i.e., pre-2003) declines.

2.2. Household and Industrial Devices

Mercury switches are available in different shapes, sizes, and electrical ratings designed to assure their reliable performance in applications that range from thermostats to gas pressure regulators to sensitive instruments and system controls used in heavy industry (EPA 2011; Tibbetts 2011;

NEWMOA 2010; Sastry et al. 2004). Mercury switches are used as measurement and control devices in a variety of household and industrial appliances, including furnaces, refrigerators, freezers, and sump pumps. In addition, mercury switches are used under the lids of washing machines, where they are responsible for turning on a light or stopping a spin cycle, and they can also be found in safety switches of smaller appliances such as clothes irons and space heaters.

Data on thermostats found in residences, businesses, and industrial settings, estimate approximately 3 grams of elemental mercury per switch, noting that some larger units can contain multiple switches with up to 12 grams of mercury total (Tibbetts 2011; NEWMOA 2010). According to 2007 figures, mercury use for thermostats sold in the United States has decreased approximately 73% since 2001 (i.e., from approximately 14.6 tons [13,000 kg] in 2001 to around 3.9 tons [3,500 kg] in 2007). This downward trend is likely a result of more states enacting legislation that restricts the sale and/or distribution of mercury-containing thermostats, leading to an increase in the use of non-mercury programmable thermostats (NEWMOA 2010). In addition, numerous collection and recycling programs exist for mercury thermostats at both the mandatory state (e.g., California, Connecticut, Illinois, Iowa, Maine, Massachusetts, Minnesota, Montana, New Hampshire, New York, Oregon, Pennsylvania, Rhode Island, and Vermont) and voluntary industry-sponsored (e.g., Thermostat Recycling Corporation) level (TRC 2014; NEWMOA 2010).

As with discarded vehicles, metallic mercury can be released from these devices and appliances after they are scrapped and sent to a recycling yard. No recent, reliable estimates exist for the amount of mercury actually scrapped each year from all household and commercial products combined. As context for the scale of this issue, the Steel Recycling Institute estimated a 90% recycling rate for U.S. steel appliances in 2011 (SRI 2014), but provided no information on the potential mercury content of these appliances. However, a 1997 EPA report suggested thermostats alone would account for approximately 10.3 tons (9,300 kg) of mercury waste generated in the United States in 2000 (EPA 1997); while a study from 2008 estimated mercury releases to land resulting from thermostat disposal to be on the order of 12.7 tons (11,500 kg) (Cain et al. 2008). As previously noted for vehicle switches, the mercury contribution to scrap metal resulting from household and industrial devices should decrease with time as the use of non-mercury devices increases and fewer older mercury containing devices remain in existence.

2.3. Chlor-Alkali Industry

Chlor-alkali plants utilize mercury as a catalyst in the chlorine production process. First developed in the 19th century, the process involves running an electrical current through a vat of salt water with a layer of mercury acting as a conductor on the bottom of the cell. The electrochemical reaction separates the salt ions, and the mercury binds with the sodium (in metallic form), leaving chlorine as the desired product. Based on data compiled in 2013, about 75 facilities (down from 96 in 2010) around the world still have some industrial mercury-cell chlorine capacity (UNEP 2013). Most U.S. facilities using this mercury-cell technology have already been shut down or are committed to phasing out their mercury use by the year 2020 (ZMWG 2011). Because the steel structures and equipment used in this process are known to be highly impacted by mercury, the potential exists for Hg-impacted steel to enter the recycling

stream as a result of plant decommissioning and dismantlement until there are no more chlor-alkali plants in existence.

Most mercury at these plants is in metallic form on the surface of the steel; however, it may also penetrate beneath the surface to a limited extent. Limited published industry data, (Euro Chlor 2004, 2009), indicate that this surface and near-surface mercury appears to be inhomogenous, with a reported typical range of 0.001 to 1% mercury by weight for impacted steel. From a recent European Commission report (EC 2014), guidelines for decommissioning mercury chlor-alkali plants recommend that the waste steel be cleaned until the level is acceptable for recycling by smelting, identified as less than 5 to 15 parts per million (ppm). Earlier reports targeted 25 to 100 ppm with no visible mercury present, i.e., no surface “sweating” (Euro Chlor 2004, 2009). In contrast, no information has been found that identifies a target concentration or quantifies the amount of Hg-impacted metal sent for recycling as scrap from the chlor-alkali industry in the United States.

2.4. Oil and Gas Industry

Mercury occurs naturally in the earth’s crust and is present in many of the world’s crude oils. As a result, petroleum products have the potential to carry mercury from subsurface reservoirs and distribute it throughout the production and refining system. The amount of mercury in crude oil and gas depends on the geologic characteristics of the region from which the crude oil originated. Concentrations have been reported to span several orders of magnitude, from parts per trillion (ppt) to as high as ppm in rare cases. A recent global survey of mercury content in nearly 450 crude oils identified a range of 0.1 to 1,000 parts per billion (ppb), with an average of 7.5 ppb and a median value of 1.3 ppb (Doll et al. 2012; IPIECA 2013). Reported concentrations in U.S. crude oil range from approximately 5 to 50 ppb (Wilhelm 2001; Wilhelm et al. 2007), while those in Canadian crude are somewhat lower at 0.3 to 12 ppb (Hollebone and Yang 2007; Wilhelm 2001). For comparison, the mean concentration of mercury in conterminous U.S. coal has been estimated at approximately 200 ppb (Toole-O’Neil et al. 1999).

The nature of mercury compounds in crude oil has not been well characterized because speciation is complicated by the volatility of many forms and the inherent complexity of the hydrocarbon matrix. Chemical reactivity studies indicate that crude oil and gas condensate can contain several forms of mercury that differ in their chemical and physical properties. The mercury in crude oil has been identified as mostly Hg(0), with trace amounts of organo-mercury species (e.g., monomethylmercury, monoethylmercury, dimethylmercury, and diethylmercury) and inorganic content (e.g., mercuric chloride), as well as insoluble mercury compounds such as mercury sulfide and mercury selenide (Rahman et al. 2010; Wilhelm 2001; Yan 1996).

Dissolved Hg(0), in its elemental form, is soluble to a few ppm in crude oil and other hydrocarbon liquids. This form adheres to metallic components (e.g., pipes and vessels) and to suspended solid materials in the hydrocarbon liquid. The measured concentration of dissolved Hg(0) typically decreases with distance from the wellhead as a result of adsorption, reaction with iron, conversion to other forms, and loss of the suspended fraction (Wilhelm 2001). Liquid mercury deposits have been observed on the inside bottom of oil pipelines and other transport

vessels that carry gas containing very high concentrations of elemental mercury. These deposits result from cooling and compression and may provide galvanic acceleration to the naturally occurring corrosion processes in the pipeline (Wilhelm 2009).

Organic mercury compounds typically include dialkylmercury (i.e., dimethyl and diethyl forms) and monomethylmercury-inorganic complexes with halides or other inorganic ions. In general, these compounds are highly soluble in crude oil and gas. They are similar to elemental Hg(0) in their adsorptive tendencies; however, because of differences in their boiling points and solubilities, organic mercury compounds differ from Hg(0) in the way they partition to distillation fractions. For instance, organic mercury compounds in produced gas are expected to partition to the liquid hydrocarbon condensate (as the gas is cooled), as opposed to the product fuel, pipeline deposits, or metallic components (Wilhelm 2001). Other organic mercury complexes may also be formed from reaction with ligands (such as an organic acid, porphyrin, or thiol) and sorption onto other suspended particles present in the hydrocarbon fluid (Wilhelm 2001).

Inorganic divalent mercury [Hg(II)] salts, most of which are halides (notably chlorides), are soluble in oil but preferentially partition to the water phase. Mercuric chlorides in particular have a fairly high solubility in organic liquids, about 10 times higher than Hg(0). Like the elemental form, these ionic salts can also be physically suspended in oil or they can adsorb to suspended particles. Inorganic suspended mercury compounds, the most common of which are mercuric sulfide (HgS) and mercuric selenide (HgSe), are insoluble in water and oil; they exist as suspended solids with a very small particle size.

Mercury has been found to primarily adsorb to the internal metal surfaces of oil and gas production and refining equipment (IPIECA 2014); penetration into the steel structure itself generally seems to be minimal. The surface coverage of mercury deposits appears to be highly heterogeneous, but the degree of deposition has not been generally characterized. Further details regarding the chemical forms and levels of mercury associated with scrap metal are offered in the next section.

No data have been found describing the annual volume of Hg-impacted scrap generated by the oil and gas industry. In addition, little is known about how cleaning processes, such as those used to remove scale and sludge from equipment taken out of service, might affect the amount of mercury associated with the steel. Thus, estimating the percent contribution of the scrap metal and its associated mercury content originating from the oil and gas industry is not possible at this time.

3. Controls for Eliminating Mercury In Scrap Metal

In general, controls do not currently exist at scrap metal recycling plants to screen or monitor for mercury in their incoming scrap feedstock. In the United States, these facilities generally rely on end-of-life vehicle switch removal programs that target removal of Hg-containing switches (e.g., from automobiles and household and industrial devices) before they are shredded for recycling. At present, no federal law requires scrap recyclers to remove mercury switches before the

shredding operation; however, several states regulate this activity (e.g., California, Illinois, Indiana, Maine, New York, and North Carolina) (IDEM 2009; IEPA 2008; CalEPA 2007; NCDENR 2007; NYSDEC 2006; ME DEP 2005). In addition, a voluntary switch-removal program exists at the national level (i.e., National Vehicle Mercury Switch Recovery Program, NVMSRP), which other states agreed to join (e.g., Michigan, Pennsylvania, and Wisconsin) (PA DEP 2013; ECAR 2010; MDEQ 2006). However, neither monetary incentives nor compensation are required or consistently offered as part of the various switch removal programs. As a result, mercury switches often are not removed before vehicles are scrapped. No additional regulations, guidelines, or controls for managing mercury associated with scrap metal, particularly as it relates to the other suspected sources (e.g., chlor-alkali and oil and gas industries), were identified.

4. Nature Of Mercury In Scrap Metal

4.1. Chemical Forms of Mercury Associated with Scrap Metal

Metal and steel surfaces have been shown to react with mercury in produced fluids (e.g., crude oil) and accumulate it in potentially considerable quantities. Mercury in oil and gas production and process streams can accumulate in equipment as a result of adsorption, chemical reaction, dissolution and deposition from sludges, and condensation. Condensation from the liquid phase occurs when the vapor pressure of mercury exceeds the limiting partial pressure for condensation because of changes in temperature and pressure in the given system. Mercury can precipitate from the liquid phase whenever its limiting solubility is reached, notably as the temperature decreases (Radford 2013). Chemisorption generally dominates for carbon steel surfaces, whereas adsorption dominates for stainless steel surfaces (Radford 2013).

Mercury has been reported to exist predominantly in the elemental form, $\text{Hg}(0)$, in the surface corrosion or rust layer of steel, at a penetration depth of 50 to 200 μm ; mercuric chloride (HgCl_2) and mercuric sulfide (HgS) have also been reported (Zettlitzer and Kleinitz 1997). The specific mechanisms of mercury sorption by steel from oil or gas are not completely understood; however, this process is thought to involve either diffusion of $\text{Hg}(0)$ into steel grain boundaries or reaction with grain boundary compounds (e.g., corrosion products). It is also believed that the adsorption is reversible, such that the surface-adhered $\text{Hg}(0)$ can re-evaporate at ambient temperatures into gas or redissolve into liquids that come into contact with the impacted surface (Wilhelm 2009). Thus, Hg-impacted scrap metal from the oil and gas industry has the potential to desorb mercury even after the equipment has been purged and cleaned of hydrocarbons (Radford 2013). Temperature has been reported to play a critical role in mercury adsorption/desorption processes (Rosenborough et al. 2006b). It is also interesting to note that storage experiments measuring the stability of mercury in liquid hydrocarbons indicated organic mercury species were stable in solution (i.e. did not absorb to the container walls) for at least 30 days in stainless steel containers, whereas $\text{Hg}(0)$ was not (Bloom 2000).

The mercury found in scrap metal resulting from the destruction of mercury switches in vehicles, household and industrial devices, and chlor-alkali industry-impacted steel initially occurs in metallic form. As a result of the mechanical agitation, cutting, and other physical disturbance

associated with the scrap processing, the mercury that does not volatilize can be distributed as fine droplets throughout the bulk scrap metal. Because bulk scrap normally has a high percentage of rusted surfaces, once in contact with the scrap, the mercury could react with and affect the steel in a manner similar to that described for oil and gas industry equipment. Alternately, the mercury droplets could become coated with fine particulates during the scrap processing, which would leave them in a liquid metallic state but with a dramatically reduced vaporization rate until redisturbed (and separated from that coating).

Most mercury compounds are thermally unstable above temperatures of 700°C to 800°C (about 1,290°F to 1,470°F) and will decompose to form Hg(0) (Capri 1996). Thus, when the scrap is heated during processing (e.g., torch cutting or shredding) or melted in a furnace, Hg(0) is expected to be volatilized out of the metal. Therefore, the recycled scrap metal product is not expected to contain any mercury.

4.2. Mercury Levels in Scrap Metal

The specific amount of mercury that steel can adsorb and retain appears to be a function of both metallurgical and environmental factors. In general, the longer the steel equipment is in contact with fluids or condensates that contain measurable concentrations of elemental mercury, the greater the potential for accumulation within that equipment. Measurements of mercury on steel from oil and gas production indicate surface concentrations on the order of 1 to 10 grams/square meter (g/m^2) (Wilhelm 2009; Wilhelm and Nelson 2010). Assuming a representative thickness of about 0.625 to 1.27 cm (one-quarter to one-half inch) for scrap steel and a density of 7.85 g/cm^3 , this translates to a bulk concentration of approximately 10 to 200 ppm by weight. Weight percent measurements of mercury in steel tubings and pipelines have been reported to range from less than 1 ppm to more than 80 ppm, with most equipment exhibiting concentrations below 5 ppm (Zettlitzer and Kleinitz 1997).

Because mercury deposits on equipment used for oil and gas production and refining are primarily associated with the corrosion layer and exist as nonvolatile species (e.g., HgS), under normal ambient conditions, it does not appear that Hg-impacted metal would pose any significant exposure risk to workers. However, if the Hg-impacted metal were to be subjected to high-temperature processes during scrap metal recycling, such as shredding, torch cutting, or welding, the mercury could volatilize and potentially pose an occupational exposure risk.

4.3. Effect of Scrap Metal Composition

Substantial partitioning of mercury between Hg(0) and Hg(II) forms, along with oxidation to chloro-mercury compounds and surface complexes, has been shown to occur in the presence of various chlorine species, including hydrochloric acid (HCl) and chlorine gas (Cl_2) (Capri 1996; Wilcox and Blower 2004; Rosenborough et al. 2006b). Thus, galvanized scrap metal exposed to chlorine sources or stored near the coastline of a saline water body will likely react, even though mercury adsorption (at room temperature) does not typically occur on galvanized surfaces (i.e., coated with zinc for corrosion protection) and only weakly on oxidized (i.e., rusted) iron. . On

the surface, the mercury is expected to react with surface chlorine to create mercuric chloride (HgCl_2), rather than simply being trapped as $\text{Hg}(0)$ (Rosenborough et al. 2006a; Orrling 2010). Regardless of the mechanism, carbon steel surfaces in process equipment exposed to mercury have been reported to accumulate some mercury in the surface layer of pipelines, valves, and process vessels, e.g., as corrosion scale (Wilhelm 2009, Wilhelm and Nelson 2010).

In addition, scrap containing copper (Cu), such as electronics-based scrap, has been shown to be prone to mercury adsorption and uptake. However, the same study found that no mercury was adsorbed on crystalline zinc (Zn) at low or room temperatures (ranging from -190°C to 25°C [-310°F to 77°F]) or at the deposition conditions tested (Rosenborough et al. 2006b). With respect to iron (Fe), it was reported that scrap iron stored between -40°C and 25°C (-40°F to 77°F) was unlikely to have any mercury on its surface because of the presence of the oxide layer. It was concluded that mercury was not likely to adsorb on iron surfaces exposed to normal atmospheric conditions because the surface oxide contamination would be too high, although clean, non-oxidized iron surfaces did show capacity for mercury adsorption (Rosenborough et al. 2006a).

5. Implications For Scrap Metal Recyclers

Scrap metal recycling is a multi-step process, starting with the collection and transport of raw scrap and continuing through pretreatment (e.g., separating, shredding, cutting, compacting), melting, refining, forming, and finishing. Because of the nature of this overall process with several high-temperature operations, and because mercury is highly volatile when heated, the recycling of Hg-impacted steel at an EAF facility has the potential to release fugitive gas (Hg vapor) and particulates, as well as Hg-impacted bag-house dust and control filters. As a result, workers in EAF facilities could potentially be exposed to airborne mercury, depending on the physical locations and activity patterns of the individual workers at a given facility.

Current characterization data are insufficient to fully understand the key sources of mercury within the scrap metal industry and quantify their associated mercury concentrations. This information is important to assessing measures to protect workers from potential health and safety hazards that might be posed by mercury and Hg-impacted scrap. Thus, identifying and characterizing the key sources, and understanding the nature and extent of associated mercury levels and releases, represent a practical research need for improving the environmental management of Hg-impacted scrap metal.

Acknowledgments

The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory ("Argonne"). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357.

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Mercury-Impacted Scrap Metal: Source and Nature of the Mercury

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