

Impact of modern battery design and the implications for primary and secondary lead production

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Abstract

The emerging change in the automobile industry with the advent of the 42 V electrical operating system will impose a revolutionary change not only on the car industry, but also on the battery industry overall. The implications of this change will be felt by the battery producers, most of whom will require new or advanced production techniques for 36 V batteries, and subsequently by their suppliers of raw material. The demand for batteries of higher quality—in particular, the valve-regulated lead-acid (VRLA) battery, which is the battery of choice for the new automotive system—will place much higher demands upon the quality of the raw materials used in battery manufacture. It has been well documented that high-quality raw materials, such as lead, acid and separators, are a requirement in order to guarantee battery performance. The presence of impurities (antimony, arsenic, tellurium, etc.) in the enclosed system of the VRLA battery will impart problems such as dry-out, self-discharge and negative-plate capacity loss which will result in premature failure of the battery. One major problem for both primary and secondary lead producers is the presence of these impurities in their metal streams. Of particular interest to the smelters are the levels of antimony and silver. The latter element is increasing to alarming levels. With changing battery technology, both elements will pose serious problems to the lead producers in maintaining high-quality lead under the present cost structure. Some of the challenges that face the lead industry in meeting the demands of VRLA battery producers for product of higher quality are examined in this paper. © 2002 Elsevier Science B.V. All rights reserved.

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1. Criteria for the modern VRLA battery

The highly successful valve-regulated lead-acid (VRLA) battery requires raw materials (lead, separators, acid) of high-quality in order to prevent gassing, self-discharge, poor cycle-life and negative-plate capacity loss. Hence, lead used for the active material must not carry into the battery elements ('impurities') that will contribute to the gassing rate. Non-antimonial alloys, such as those in the calcium family, lead-tin and even pure-lead, are used to ameliorate the gassing problem associated with the use of antimonial alloys. Thus, growth in the application of VRLA batteries will result in lower usage of antimony.

The VRLA battery is expected to be the preferred battery product when the automotive industry converts to the 42 V system. With the increased voltage comes a demand to reduce weight and this will result in the production of thinner grids made from corrosion-resistant alloys, particularly

those which incorporate silver. Therefore, there will be less dependence on book-mould casting and more focus on other grid-production techniques such as expanded metal and the more recent extruded metal and (Conroll) processes. All such techniques necessitate the use of non-antimonial alloys. Effectively, this means less antimony will be required for alloys and more silver will be employed. On the other hand, soft lead for the production of active material will specify low maximum levels for both antimony and silver. This raises major issues for lead supply.

2. Problems facing the lead industry

2.1. Antimony

Antimonial alloys have been the mainstay of the battery industry for many years and have been used for the grids, straps and terminals of batteries. Although the level of antimony in alloys has been gradually reduced via alloy improvements and modifications in battery technology, the

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alloys are still widely used, although to varying extents in different countries. For example, in USA, the calcium-type alloy is most dominant, whilst in countries in the South East Asian region, the antimonial alloy is the most popular choice grid for production.

Many replacement alloys have been suggested over the years and have ranged from calcium to strontium to barium alloys. Thus, the demise of the antimonial alloy has long been predicted. Nevertheless, antimony has remained in widespread usage. On one hand, this is due to its availability, ease of manufacture and price, and on the other hand to its proven robust performance in batteries, especially under deep-cycling service. The antimony content has changed from initial levels of around 12 wt.%, through 6 to 4 wt.%, to 2.5 and 1.6 wt.% in the present selenium-refined alloys, in line with the demand for better battery performance. Now, with the development of the 42 V VRLA battery system for automobiles, producers will be required to convert to calcium and ‘antimony-free’ alloys, which will effectively eliminate the major use of antimonial alloys in the original equipment market.

Antimony alloys have been a benefit to both primary and secondary lead producers, as antimony is a natural arising in lead streams, albeit from differing sources. In the primary system, antimony reports in concentrate inputs from mines where it occurs naturally with lead ores such as galena and the zinc ore sphalerite (see Table 1). Levels vary according to the ore body, and range from 0.005 wt.% in Ireland to 1.0 wt.% in Peru. It is uncommon to find an ore body without antimony present. The source of antimony for the secondary lead industry is from spent automotive and industrial batteries—the principal feedstock—that have grids made from antimonial alloys.

Secondary smelting operations benefit from contained antimony because, due to the pyrolytic smelting regimes employed in the industry, the bullion lead from the furnace often contains up to 2 wt.% contained antimony. With simple chemical dressing operations to ‘clean’ the lead by lowering the level of impurities, the lead/antimony

bullion is available for alloying almost immediately [1]. In primary smelting, the operations are somewhat different since antimony is considered to be an impurity at the ‘refinery floor’. The element must be totally removed from the bullion lead as part of the refining process to gain access to contained valuable metals such as silver, gold and copper. At a later stage, antimony is recovered in the metallic form and re-alloyed to pure-lead to produce antimonial alloys [2].

The problem facing all smelters is that there is a continuous arising of antimony in their lead streams, against a quickly declining demand for the element. Declining sales of the alloy will result in antimony effectively becoming a major contaminant and, therefore, it must be removed. The decline in demand is not new, as the change to hybrid and maintenance-free batteries and developments in grid-casting technology has caused a growth in non-antimonial alloys world-wide and the replacement of a large tonnage of antimonial alloy over a period of time. With increased production of ‘non-antimony’ VRLA batteries for automotive 42 V systems there will be an accelerated and massive decline in demand for the alloy.

The change in the alloy matrix poses several problems to the smelters, particularly in the refining of the metal and the by-products produced. To understand better the impact to smelters, the following is a brief outline of the removal of antimony from lead.

1. Generally, the antimony content in bullion lead in a smelting stream is between 0.5 and 2 wt.% as determined by the feed-source of the material.
2. The major removal method for antimony from either a primary or a secondary lead stream is through the injection of oxygen via a lance into a pot of molten lead held at 500 °C [2].
3. The other method for the removal of antimony from lead is by the addition of chemicals such as sodium hydroxide and sodium nitrate to the molten metal. This procedure is much more costly and less efficient than oxygen lancing.
4. After a short period, a high antimonial slag is formed on the surface of the molten lead and is removed periodically. This is termed ‘softener slag’.
5. The softener slag removed from the process contains approximately 6–8 wt.% antimony and minor amounts of arsenic, tin, and selenium.

Whilst the process is efficient in the removal of antimony and other elements from the molten lead, the problem is that the softener slag is basically around 84 wt.% lead. Further processing, i.e. re-smelting and refining of the softener slag, can upgrade the slag to around 21 wt.% contained antimony, but is costly and the dominant element is still lead. To date, this is the only ‘economical’ method for removal of antimony, but the downside is that it ‘ties up’ large amounts of lead units in the softener slag. This gives rise to two issues.

Table 1
Antimony content of major world lead concentrate mines in 2000 [3]

Mine	Concentrate (10 ³ × t)	Sb content (wt.%)	Sb arising (t)
Broken Hill, Australia	120	0.15	262
Cannington, Australia	300	0.28	840
Red Dog, USA	145	0.1	145
Black Mountain, South Africa	90	0.05–0.1	90
Lisheen, Ireland	35	0.005	2
Milpo, Peru	35	0.4	140
Huaron, Peru	15	1.0	150
San Cristobal, Peru	100	0.2–0.4	300
Sub-total	840		1929
Total concentrate	2200		4700

Note: ‘total concentrate’ antimony arisings are estimated from known world supply.

Table 2
Antimony arisings from secondary smelters in 2000 [4]

Region	Secondary Pb ($10^3 \times t$)	Sb content (wt.%)	Sb arising (t)
Australia/New Zealand	42	1.3	570
Asia	470	1.5	7050
America	1400	0.7	9800
UK/Europe	1000	0.9	9000
Total	2912		26500

Note: the 'Sb content' column reflects the change in alloy usage across the regions.

2.1.1. Issue 1: what is the extent of the antimony problem and what effect would it have on the lead market?

At present, there is no commercially viable method for the extraction of antimony from high-antimonial softener slag. As the total world market finds antimony to be in excess, it may become necessary to stockpile the antimony-rich material as there is no other use for the product. This process means that significant lead units are 'tied up' in the softening slag. The extent of this problem can be seen by examining present world antimony arisings from lead smelting (Tables 1 and 2). In year 2000, there were around 31 000 t of antimonial arisings. Assuming a 6 wt.% antimony softener slag, this equates to the 'tying up', theoretically, of over 500 000 t of lead units per annum. Of course, this is based upon total antimony units and is the extreme case. The most likely scenario is that, due to their processing methods, primary smelters will be the first to stockpile their softener slag, which would equate to approximately 80 000 t of lead units per annum in lead availability.

Projecting forward, the antimonial arisings will decline as antimony units decrease in the secondary lead stream, although there will be at least a 5–6-year lag due to the natural battery recycling rate (see Fig. 1). Importantly, primary smelters will always have a problem with antimony, as the level from the concentrates (as shown in Tables 1 and 2) will remain relatively constant.

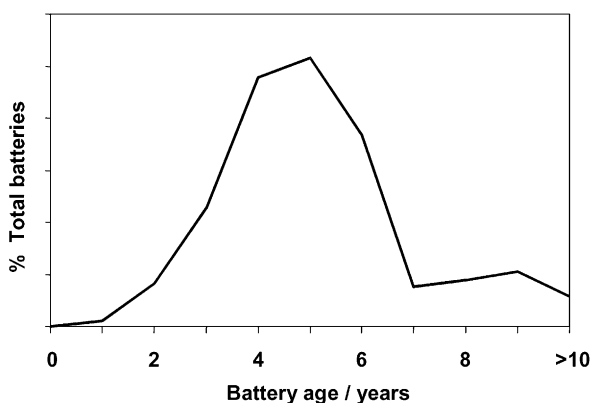


Fig. 1. Age of batteries upon recycling in Australian/New Zealand market. Zero represents current year.

2.1.2. Issue 2: when does antimony become detrimental in the lead stream for lead producers?

In fact, antimony has already become an impurity in some markets, e.g. North America, which predominantly supply calcium alloys to battery producers.

In the present production of antimonial alloys at a smelter, pure antimony or antimony-containing alloys such as the softener slag are required to top up the antimony level in the alloy which is being produced (since the recovered antimony from the furnace is generally insufficient on a batch-by-batch basis). Overall, the producer will be in either excess or deficit in antimony units. Those who have excess antimony remove the high-antimony bullion from the softening process and sell this material to other markets who still require the element, such as the southern and west Asian regions which are in deficit.

This overall change in the antimony balance has been less noticeable to the lead industry, as the high bullion lead has replaced the use of metallic antimony metal used in blending in the alloy mainly due to a price differential. Whilst not visible to the lead market, the effect on the reduction in antimony usage can be seen in the antimony metal market. There are regular market reports on the continued decrease in recent times of exports from China—the largest producer of antimony—in particular, in the number of antimony export licences granted. In the last few years, Chinese export tonnage has dropped from 180 000 to 60 000 t per annum [5]. Metal traders often blame smugglers for the decrease in export, but the decreasing price and demand does not reflect these rumoured reports.

More lead producers will, over time, find that the antimony contained in their metal stream turns from a benefit to a deficit as more battery makers adopt non-antimonial alloys. The point at which the world antimony market will be in excess, and the stockpiling of softener slag commences, will not be far off as outlined below in Section 3. Of course, smelters will find that not all of the lead stream will require softening, because there will be a requirement for antimonial alloys of some type and amount. In most cases, however, the lead stream will have to be softened and this will add costs and time to current processing operations, particularly for the secondary lead sector.

2.2. Silver

In the smelting of primary lead, silver is considered to be a valuable by-product and is recovered during the refining process. In fact, most of the world's silver is produced as a by-product of lead and copper smelting. The initial content of silver in the bullion lead can be up to 0.5 wt.%, and after subsequent refining the levels are below 0.001 wt.% in the final product. This is not the case in secondary smelting where the element is considered to be an impurity. Historically, silver levels in secondary lead have ranged from 0.001 to 0.005 wt.% for most smelters world-wide, and these levels have been stable for many years.

In fact, silver is not taken to be a ‘true’ impurity, such as nickel, cadmium and zinc, due to its stable level. Although silver-containing lead alloys have been made for many years, their overall tonnage has been small and has had little impact on recycling. Now, with the introduction of the various calcium–tin–silver alloys [6–9] for use in automotive batteries, the level of silver is rapidly increasing in recycled lead streams throughout the world. The level is encroaching on the upper limits of many specifications. For example, as the silver level increases, it starts to inhibit the oxidation of lead, particularly in the Barton–pot process [10]. It has been reported that minor levels of silver can reduce the rate of production of lead oxide by up to 10%. The problem now facing the secondary smelters is that whilst there is a method for removing silver, it is a very costly exercise due to the low level of silver present in relation to primary smelting. To understand the extent of the problem, the method for silver removal is outlined, as follows.

The removal of silver is performed in two stages [11]. First, zinc is added to lead bullion held at 500 °C. (The bullion is considered ‘clean’ as it has already passed through the copper removal and softener slag stages.) The molten bath is then allowed to cool to around 350 °C at which a silver–zinc–lead crust forms on top of the molten lead. The crust, which is subsequently removed from the process, is highly valued as it contains approximately 6 wt.% silver. This material is then fire-refined to a final 99.999 wt.% silver product. The difficulty for secondary plants is that whilst the process could be used for the removal of low levels of silver (i.e. up to 0.01 wt.%) in lead bullion, the same amount of zinc must be added as for high-silver primary bullion (up to 0.5 wt.%). The resultant crust would be very low in silver and, therefore, low in value. Thus, the process is very costly and does not give the benefit of silver credits.

With increasing silver levels in the lead stream, it will become even more difficult, or even impossible, for secondaries to meet present lead specifications. This includes specifications such as the London Metals Exchange (LME) registered standard lead which, based on the EN standards, requires a maximum of 0.0050 wt.% silver.

2.3. Other elements

Another element that is now increasing in some secondary lead streams is bismuth. As with silver, bismuth has been a ‘stable impurity’ for many years with levels that range from less than 0.001 to 0.025 wt.% in both primary and secondary lead.

In recent years, bismuth has been added to high-purity lead at levels of between 0.05 and 0.60 wt.% for oxide production. Unlike antimony and silver, however, bismuth has been found to be beneficial in a number of aspects of battery production. For example, work carried out by CSIRO [12–17] and Pasminco [16,17] has demonstrated that this element imparts many advantages to VRLA batteries. Other research from China has reported no adverse

effect of bismuth on grid alloys when added at levels below 0.1 wt.%.

Due to the relative ratios of current levels versus the projected level, bismuth will not increase to the same extent as silver and threaten specification limits. The absolute maximum level bismuth can achieve is 0.05 wt.% which, as noted above, has been proven beneficial in the production and use of lead oxide.

3. A case study of antimony and silver levels

To understand the effect and extent of change of the two elements into the future, a study of the Australian/New Zealand region has been conducted to examine in the levels of antimony and silver in the recycled lead stream. The region could be termed a ‘controlled’ or ‘closed loop’ market as each criteria of the market is well understood, i.e. lead supply, tonnage of product, recycle rates, etc. Another important advantage in studying this market is its relative isolation from the other major markets, whilst maintaining world trends in batteries and alloys types. Any changes in production, alloy composition or battery technology over the past 10 years is well known.

3.1. Background to the data

The lead supply and battery market of the region consists of:

1. one primary smelter (Pasminco Port Pirie);
2. three secondary smelters (Australian Refined Alloy plants in Sydney and Melbourne, Exide plant in New Zealand);
3. two major battery producers who supply around 70% of the market (Century Yuasa, Exide Technologies);
4. the remaining 30% of battery market is imported predominantly from the South East Asian region.

The production of secondary lead is around 42 000 t per annum and consumes almost 100% of the battery arisings, whilst there is little importation of spent batteries for recycling. The battery market itself has undergone several changes over the past 15 years. These include:

1. introduction of a calcium–tin–silver alloy in 1999 following its usage in USA;
2. the gradual introduction of calcium alloys;
3. reduction of the antimony content in alloys, i.e. from 4–6 wt.% antimonial to 1.6 wt.% antimonial selenium-refined alloys for hybrid batteries;
4. closure of two battery plants (Besco Batteries in 1987, Apollo in 1994).

The major data collected and used in the case study include:

1. battery age recycle rates (Fig. 1); this data was gathered from over 2000 batteries examined before recycling [18] in Australia;

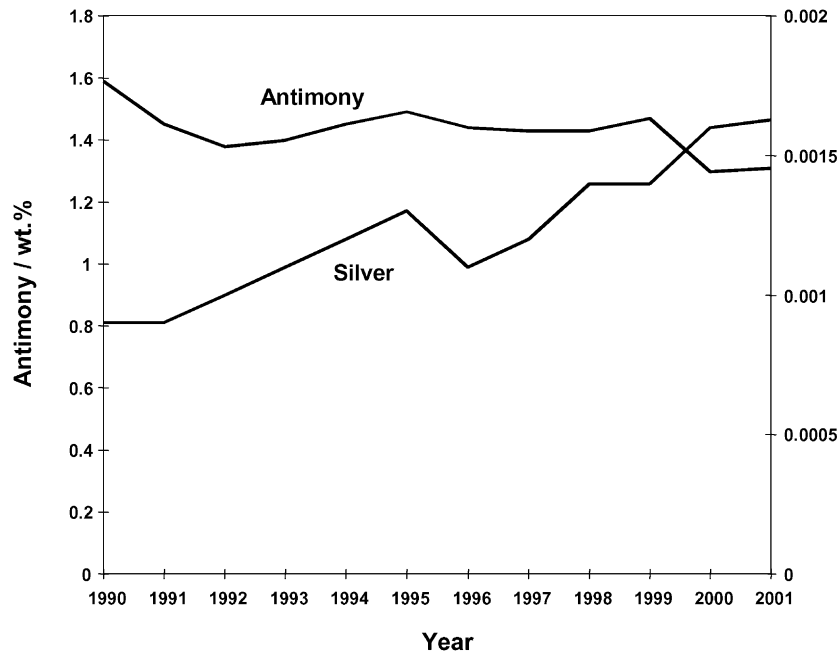


Fig. 2. Secondary bullion analysis from Melbourne plant of Australian Refined Alloys over past 12 years.

- secondary bullion lead analysis collected from over 12 years of production at Australian Refined Alloys (Fig. 2) [19];
- tonnage of the Australian/New Zealand market in terms of alloy type and volume, recycle volumes, and timeline of major events.

3.2. Interpretation of data

Using the data derived from the above information, a mathematical model was produced to project the levels of antimony and silver over the next 10–15 years, as presented in Section 3.3. The two keys to the model were, first, the time delay from the production of the battery until it is recycled and the lead recovered as given in Fig. 1 and, second, the trends in the two elements over the previous 12 years, see Fig. 2.

An earlier recycling study was used in the compilation of the data for Fig. 1, which shows there is delay of around 4–6 years from the production of a battery until it is recycled and the lead recovered. Therefore, any change in battery technology, e.g. alloy composition, will not flow through to the recycled lead stream until 3–4 years later, while the full effect of the change will appear in 6 years. (Note that the data in Fig. 1 do not indicate the life of a battery, rather the time it takes for the battery to be recycled.)

The information given in Fig. 2 show that, as expected, there has been a decrease in the antimonial content over the past 12 years. It also reveals an accelerated rate in the last 2 years, which corresponds to changes in the antimonial content of some of the alloys that had been used previously. More surprising is the increase in silver level, as this was

expected to remain relatively steady since there has been no increase in silver content from Pasminco Port Pirie and the calcium–tin–silver alloy was not introduced until 1999. From other evidence, it is believed that the increase is most likely from the 30% of batteries imported into the Australian market. This rate of increase in silver levels was factored into the model.

3.3. Results

As stated in Section 3.2, the bullion data reveal an expected decrease in the antimony content in the secondary lead stream over the past 12 years. The major changes in the graph, as shown in Fig. 3, can be related back to changes in the battery manufacturing. For instance, the decrease in antimony levels in 1992 corresponds to the closure of Besco Batteries in 1987. Besco used predominantly high-antimony alloys of around 4 wt.% in their batteries unlike that of the other major Australian battery manufacturers. The second decrease in the antimony level in 1999 reflects a change in battery technology by the two major manufacturers, in particular a move to the hybrid battery. This trend has been maintained as there is very little production of antimonial alloy that contains more than 2 wt.% antimony, other than for industrial batteries and cast-on-strap and terminal usage.

From the model, and factoring the demand and type of alloys being used by the major battery producers, Fig. 4 gives the projection of antimony level in the secondary lead stream. With the greater usage of the calcium–tin–silver alloy by one manufacturer and a shift to expanded metal by the other, antimony will continue to decline in the bullion lead stream. It is predicted that, even with current demands,

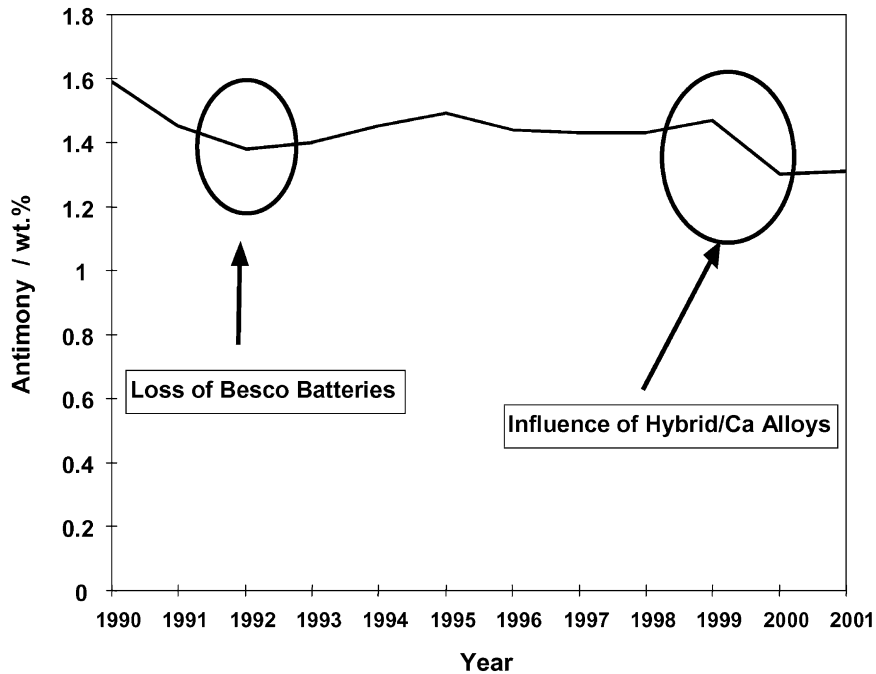


Fig. 3. Antimony values from secondary bullion analysis at Melbourne plant of Australian Refined Alloys.

the Australian market for antimony will go into oversupply between 2004 and 2005 where the antimonial lead units will have to be removed from the recycled lead stream.

By contrast, silver presents a different picture. Modelling the silver values, with consideration of the tonnage of calcium–tin–silver alloy being used currently and since 1999 and its imminent return to the scrap market, the data indicate a rapid increase in silver levels over the next 3–4 years, see Fig. 5. The model demonstrates that once the initial increase occurs, the rate of increase in silver slows to a rate which is similar to that experienced before 1999. The broken line on the graph attempts to show that should

another producer or importer introduces a silver-containing alloy to the market, then the silver level in the recycled lead steam will increase significantly. This assumption is based on the battery taking 5% of the Australian/New Zealand market, which would equate to half the present input of silver from the Australian battery manufacturer.

At the projected 2005 levels and not including any overseas influence, a number of current alloy and soft-lead specifications will not be met because of the increasing level of silver. Considering further increases in the levels, specification limits may need to be adjusted to accommodate secondary lead alloys. Importantly, even with levels exceeding

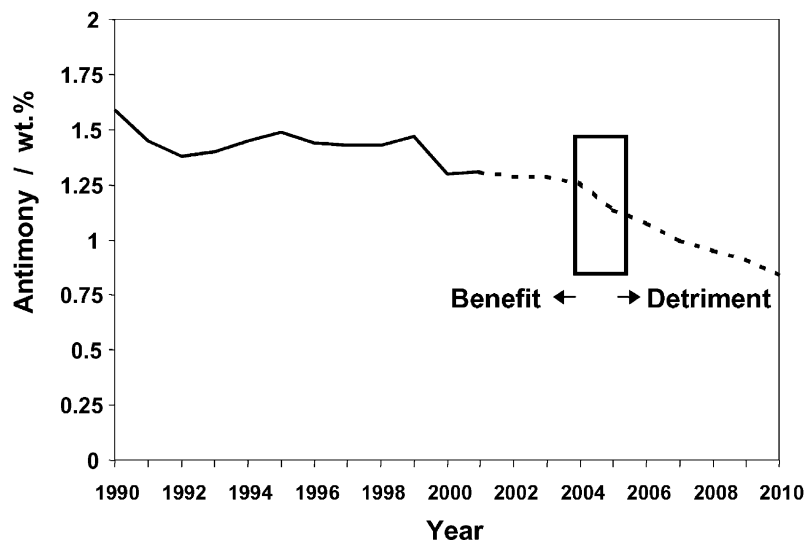


Fig. 4. Projected levels of antimony in secondary lead stream in Australian/New Zealand market.

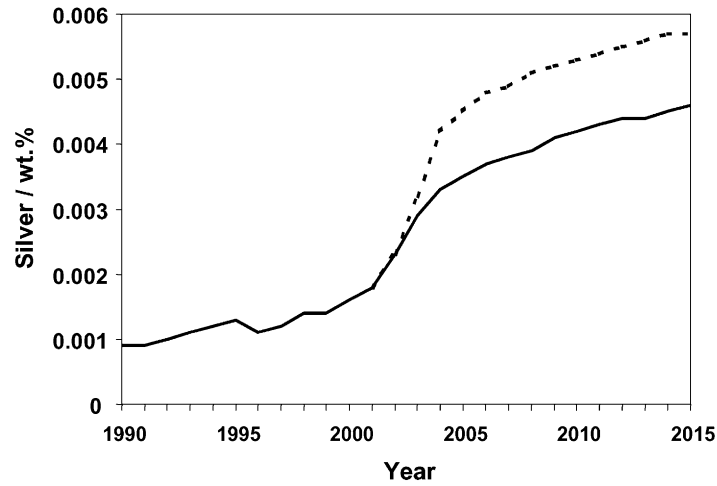


Fig. 5. Predicted silver level increase to 2015 based on Ca–Sn–Ag production levels in Australia. Broken plot reflects a possible further increase if imported batteries also contain silver-bearing alloy.

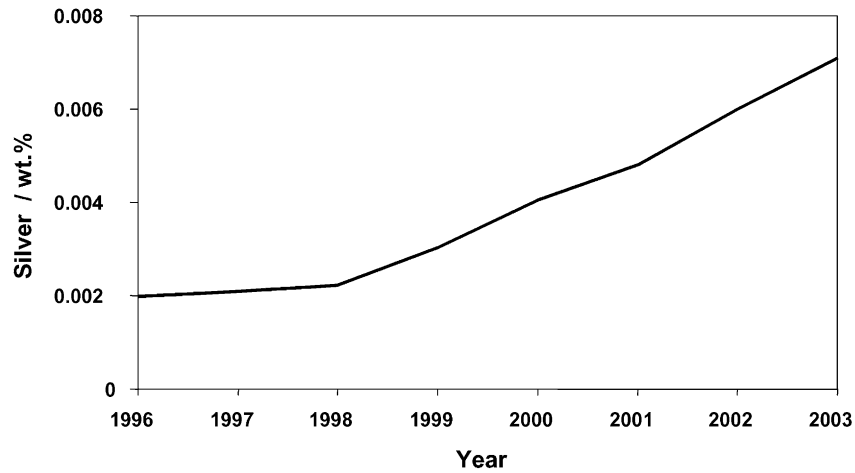


Fig. 6. Actual and projected data of silver content of pure-lead from RSR plants, USA [10,20].

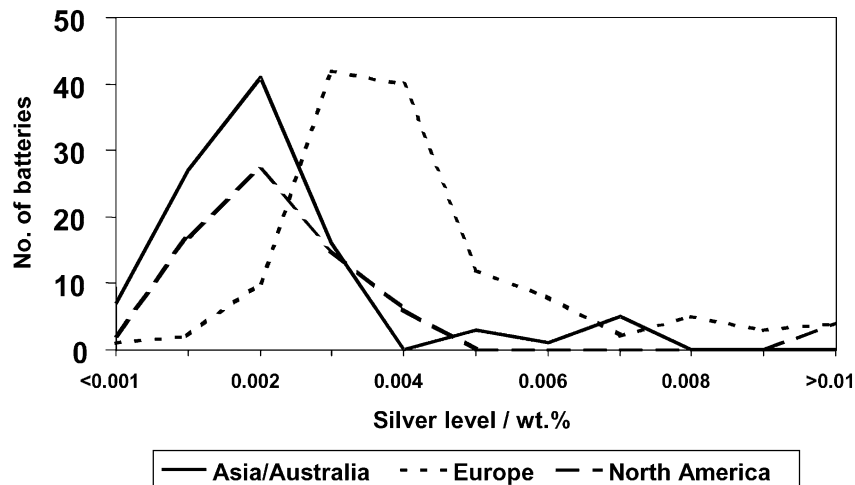


Fig. 7. World-wide silver levels in lead and lead alloys in a 1994 study.

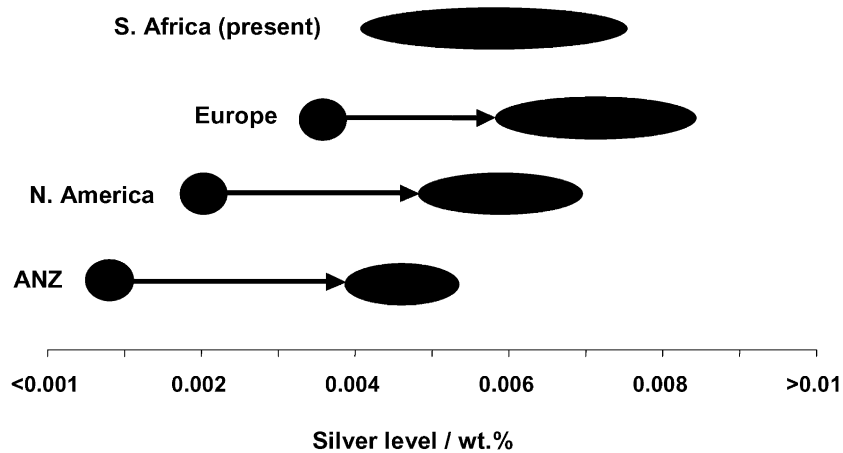


Fig. 8. Prediction of silver levels from 1994 to 2005 across the world.

0.005 wt.% contained silver, it is still not economically feasible to extract the element. The cost of zinc alone far outweighs the recovered value of the silver.

The trend shown in Fig. 5 corresponds to actual data previously reported for markets in USA, as given in Fig. 6 [10,20]. The two graphs lag in the silver increase by 3–4 years, which can be explained by the staggered introduction of the silver-containing calcium alloys to the respective markets. The alloys entered the North American markets in 1995–1996 but much later (1999) in the Australian/New Zealand region.

To estimate the level of increasing silver on a world basis, data from test-work conducted in 1994 could be used as a 'base case'. An internal Pasminco study analysed over 200 batteries sourced from around the world. Included in this work was the analysis of all lead fractions in the battery, including the silver levels, as detailed in Fig. 7. Even at the time of that report, silver levels were elevated in certain regions such as Europe. By taking the same rate of change that is estimated for the Australian/New Zealand market, we can predict the increase in silver levels world-wide in 2005, as shown in Fig. 8. It is interesting to note that recent data from South Africa indicate that current silver levels in soft-lead ingots range from 0.004 to 0.007 wt.%.

4. Conclusions

The accelerating trend towards the use of sealed lead-acid batteries (maintenance-free and VRLA types) has significant implications for the battery industry and its suppliers of lead and lead alloys. Demands for higher performance mandate the use of different and higher purity raw materials.

Some major issues facing the industry as we move forward are as follows:

1. The necessity to use calcium–tin type lead alloys in an effort to minimize water loss and consequent dry-out of

the battery means that the use of antimony-containing alloys will reduce further.

2. Excess antimony arisings from both the primary and secondary lead streams will be considered a 'contaminant'. It is metallurgically difficult to remove antimony from lead economically. The antimony that is excess to declining alloy demand will eventually lock-up lead units permanently within the lead supply chain in the form of an antimony–lead slag and, thereby, could potentially exacerbate any tightness in lead supply.
3. The growing use of silver in calcium–tin alloys will raise silver levels within the recycling chain further. Silver can be removed economically by primary smelters. Many secondary smelters may struggle to keep silver levels sufficiently low to be within many specifications, and may have to install additional processing steps to remove silver from their circuits, which will result in significantly higher processing costs.

The lead-acid battery can meet the ever-increasing demands for higher performance. Clearly, there are major challenges confronting lead producers who will be required to supply a growing market which demands lead of higher purity that can deliver the required performance criteria. These are not impossible hurdles to overcome but it will require a focused effort, particularly in the secondary lead area, to meet the stringent, but achievable, demands.

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