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The future of automotive lithium-ion battery recycling: Charting a sustainable course

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ABSTRACT

This paper looks ahead, beyond the projected large-scale market penetration of vehicles containing advanced batteries, to the time when the spent batteries will be ready for final disposition. It describes a working system for recycling, using lead–acid battery recycling as a model. Recycling of automotive lithium-ion (Li-ion) batteries is more complicated and not yet established because few end-of-life batteries will need recycling for another decade. There is thus the opportunity now to obviate some of the technical, economic, and institutional roadblocks that might arise. The paper considers what actions can be started now to avoid the impediments to recycling and ensure that economical and sustainable options are available at the end of the batteries' useful life.

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1. Introduction

Recycling, per se, is not inherently good or bad [1]. For some materials such as glass [2], the benefits are dubious and depend on factors like the shipping distance. There has been some debate about the benefits from recycling primary alkaline batteries over simple disposal [3] because the materials are abundant and non-toxic, now that the batteries no longer contain mercury. For automotive batteries, however, the environmental benefits are clear, although they vary with battery type and recycling method. There are potential economic benefits as well. If usable materials can be recovered from used batteries, less raw material needs to be extracted from the limited supplies in the ground. If the raw materials come from abroad, recycling domestically reduces the quantities that need to be imported, improving the balance of payments. In addition, significant negative environmental impacts can occur from mining and processing ores (e.g., SO_x emissions from smelting of sulfide ores, such as those that yield copper, nickel, and cobalt), and these are avoided if the materials can be recycled [4]. Recycling has its own environmental effects, but these are generally smaller than those from primary production. There are, of course, exceptions, such as recovering lithium from pyrometallurgical process slag. Recycling of materials avoids processing costs for waste treatment. In addition, some spent batteries are classified as hazardous wastes, increasing transportation, treatment, and disposal costs, as well as the effort needed to achieve regulatory compliance. Lithium-ion batteries are classified as Class 9 miscellaneous hazardous materials, and lead–acid batteries are listed as Class 8 corrosive hazardous materials under United States regulations (40 CFR 173.21(c)) [5].

Lithium-ion batteries are starting to be used in significant quantities for automotive propulsion. Because these batteries are expected to last the life of the vehicle, they will not be ending their useful lives in large numbers for about 10 years. They may subsequently be used for utility energy storage, but eventually their useful lives will end. The question is, what steps can be taken to ensure that these spent Li-ion batteries are recycled. In an ideal system, these batteries would be sent for responsible recycling and not be exported to developing countries with less stringent environmental, health, and safety regulations. Methods are needed for the safe and economical transport and processing of the spent batteries, as well as environmentally sound recycling. In addition, the recycled product needs to be of high enough quality to find a market for its original purpose, or it must find an alternative market. Fortunately, a battery recycling system is in place that already works well, and many lessons can be learned from it.

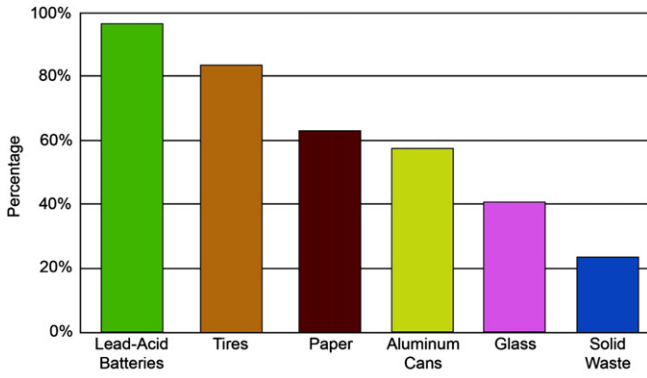
1.1. Lead–acid battery example

Lead–acid batteries are recycled more than any other major consumer product (see Fig. 1). Even with advertising programs, education, and convenient curbside pickup, recycling of common consumer products in the United States has not been a resounding success. However, lead–acid batteries (and to a lesser extent discarded tires) have achieved exemplary recycling rates.

In the United States, about 99% of lead–acid batteries are recycled [6]. Lead–acid (Pb–acid) battery recycling is also working well in Europe and Japan. In disadvantaged areas, backyard operations have exploited children to disassemble batteries and electronics for treatment in smelters without emission controls, and dumped lead-contaminated acid into the water supply, but such practices are now being eliminated [7].

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Sources:
 Smith Bucklin Market Research and Statistics Group (2011 National Recycling Rate Study)
 The Rubber Manufacturer's Association (2009 tire recycling rates) 10/2011
 The American Forest & Paper Association (2010 paper recycling rates) 04/2012
 The Aluminum Association (2010 aluminum recycling rates) 06/2011
 Glass Packaging Institute (2010 glass recycling rates) 04/2012
 Environmental Protection Agency (2010 solid waste recycling rates) 12/2011

Fig. 1. Batteries are the most recycled consumer product. (Courtesy of Battery Council International)

To date, the model shown in Fig. 2 has worked admirably for lead-acid battery recycling. We consider whether some variation of this model would work well for other battery types.

1.2. Comparison of automotive battery types

Before considering that topic, it is useful to first compare the physical and chemical structures of different types of automotive batteries: namely, the lead-acid batteries used for starting-lighting-ignition (SLI) and commonly found under the hood of most cars, nickel-metal-hydride (Ni-MH) batteries used in hybrid vehicles, and Li-ion batteries used in plug-in vehicles and some hybrids. The latter two battery types are used primarily for propulsion and heating, ventilation, and air conditioning (HVAC), although some designs are also available for use in conventional SLI applications. These will be discussed later.

The three battery types are all conceptually and structurally similar but chemically quite different. Each consists of electrode (cathode and anode) active materials on grids or foils that serve as the current collectors, with an electrolyte that carries charge between the electrodes.

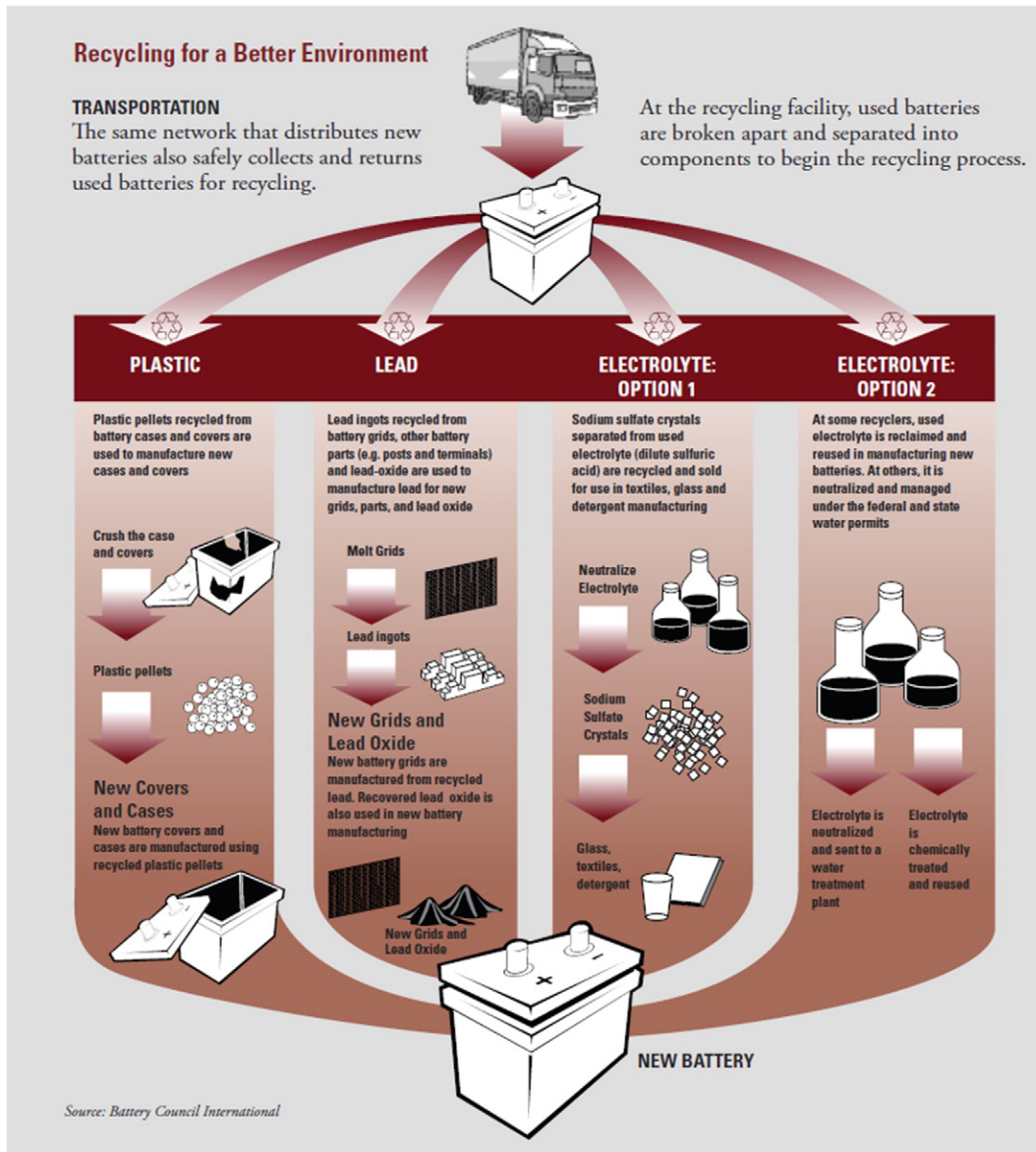


Fig. 2. Simple processes are used to recycle lead-acid batteries. (Courtesy of Battery Council International)

Table 1
Comparison of cell materials.

Cell component/battery type	Pb–acid	Ni–MH	Li-ion
Cathode	PbO ₂	Ni(OH) ₂	LiMO ₂
Cathode plate/foil	Pb	Ni foam	Al
Anode	Pb	MH (AB ₅)	Graphite
Anode plate/foil	Pb	Ni-plated steel	Cu
Electrolyte	H ₂ SO ₄	KOH	Organic solvent + LiPF ₆
Separator	PE or PVC w/silica	Polyolefin	PE/PP
Cell case	PP	Stainless steel	Varies (metal or laminate)

PE = polyethylene; PVC = polyvinyl chloride; PP = polypropylene.

These components are housed in an enclosure. As shown in Table 1, the compositions of these components differ greatly among the battery types.

It is apparent that the number of distinct materials within a battery type increases across the table. The more diverse the battery materials, the more complex the recycling. For Pb–acid batteries, all of the internal components contain lead, which makes up about 60% of the battery mass. Except for electrical connectors, which can be removed when the cells are opened, no other metal is present; as a result, no separation processes are required. Because nickel dominates the Ni–MH battery, it can be the focus of recycling. In addition, the nickel and steel alloy that results from current recycling processes is a valuable input to stainless-steel manufacturing, making recycling of these batteries economical today. The many different materials in a typical Li-ion battery complicate recycling.

2.1. Lead–acid battery recycling

Disposal of Pb–acid batteries is illegal in most states, and many states require a monetary deposit as an incentive for consumers to return their batteries. Most Pb–acid batteries are collected when new ones are purchased (the dealers are required to accept them and are paid for their trouble). In some cases, spent batteries can be returned to the manufacturer via back-haul (in the United States, not Europe), minimizing transportation costs. Additionally, as required by law, batteries are stripped from vehicles that have gone out of service and are about to be shredded. Regulations concerning transportation and processing of batteries are in place and widely known.

As shown in Fig. 2, the lead–acid battery components are recycled by a simple process. First, the battery case is broken open, and the sulfuric acid electrolyte is drained out and collected. The plates and connectors can be removed from the case at this point and recovered whole.



Fig. 3. Lead–acid battery cases and plates after separation.

(Fig. 3 shows recovered plates and cases.) Alternatively, the drained battery can be sent to a hammer-mill for size reduction, and the plastic and lead can be separated by a simple sink–float device. The recovered lead (a low-melting metal) is remelted and purified to make new battery components. Lead and sulfur emissions from secondary smelting are tightly regulated by the Environmental Protection Agency [8]. The plastic is melted and molded into new cases. The acid can be neutralized or processed to sulfate salts for various uses, such as the manufacture of soap.

The recycling operation is profitable because recycled lead (taken back to its elemental form and purified) is known to be of high quality, so there is little incentive to export to places with less-stringent regulations, although some batteries do find their way to Mexico [9]. Some battery manufacturers prefer new over recycled lead.

A key reason for the success of lead–acid battery recycling is that essentially all of the manufacturers use the same raw materials: lead, lead oxide, and sulfuric acid in a polypropylene case. Because the battery design is similar for the manufacturers, automated technology can be used for battery disassembly. In summary, lead–acid recycling works well because it is profitable, it is illegal to dispose the batteries without recycling, the battery disassembly is simple because of the standard design used, the battery chemistry does not require segregation, and the recycling process is simple.

2.2. Nickel–metal–hydride battery recycling

Large-format Ni–MH batteries have been used in hybrid vehicles for long enough that some now require disposition by either reuse or recycling. A recycling system is already in place because consumer batteries from smaller devices such as power tools, which have much shorter useful lives, have been recycled commercially for many years (see Fig. 4). However, not all of the battery materials are being recovered as high-value products. The nickel and iron are recovered by rotary hearth and electric-arc furnaces as ferronickel to feed stainless-steel production. Because this is a high-value product with a huge market, there is no need to separate the nickel from the iron. However, using this technique loses the rare earths in the metal hydride to the slag, which is used as roadbed aggregate in place of gravel [10].

Recently, the increasing demand for rare earths in batteries, motors, and other components of vehicles and wind turbines, coupled with China's policies to restrict exports so that they can satisfy their own demands, has provided a significant economic incentive to recover these metals during recycling.

Nickel–metal–hydride batteries all have very similar chemistry (AB₅), although the exact mix of rare earth elements in the hydride may vary slightly, and pack configurations differ. For that reason, differentiation among Ni–MH batteries is not needed. Several companies have announced programs to recover the rare earths from Ni–MH batteries. Umicore, in Belgium, recovers nickel and has an agreement with Solvay (formerly Rhodia) to process slag and recover the rare earths [11]. Retrieval Technologies (formerly Toxco) has a plant under construction in Ohio, partially funded by American Recovery and Reinvestment Act funds received in 2009, and plans to recover rare earths when its first processing line is completed. Honda has an agreement with Japan



Fig. 4. Hand-sorting of Ni–MH consumer batteries. (Courtesy of Kinsbursky Brothers, Inc.)

Metals and Chemicals [12] to recycle its Ni–MH batteries. In Australia, Toyota offers a \$100 rebate when a Prius battery pack is returned, and a discount on a replacement [13]. In sum, Ni–MH batteries seem to be on track for successful recycling.

2.3. Lithium-ion battery recycling

Several factors contribute to making Li-ion battery recycling more complicated than Pb–acid or Ni–MH recycling. First, as shown in Table 1, Li-ion batteries have a wider variety of materials in each cell. The active materials are in the form of powder, coated onto metal foil, and these different materials must be separated from each other during recycling. Lead–acid batteries have a relatively small number of large lead plates packed together in a single plastic case, while a Li-ion pack is likely to have 100 or more individual cells (~5000 for a Tesla electric vehicle), which are connected into modules and, in turn, are assembled into a pack with control circuitry attached to each cell (see Fig. 5). A thermal management system may also be included. These components could

possibly be recovered intact or may contain valuable materials that would provide some economic incentive for recycling the battery pack.

Within the cells, the chemical compositions of the active materials – especially the cathode – vary with manufacturer and battery function, are changing, and may never standardize. The most common cathode material for the batteries now prevalent in consumer electronics is LiCoO_2 (LCO), but various combinations of Ni, Mn, and Al can be used to replace some or all of the cobalt to optimize performance while lowering raw material cost, which is the key for automotive batteries. Another promising cathode material that uses very low cost inputs is LiFePO_4 (LFP). Most manufacturers use some form of graphite for the anode, but silicon is also being used, and other materials and mixtures are being studied.

Lead–acid batteries are small and easily removed from their location under the hood, while the larger, more complex Li-ion packs vary in shape and location in the vehicle. As a result, removal may be more difficult. However, if removal takes a professional, fewer people need to be trained on proper handling and separation. If Li-ion batteries last the entire vehicle life, they will all end up in scrap yards or at auto dealers. These are both sufficiently large enterprises to facilitate collection. Vehicle batteries subsequently used for stationary energy storage could be collected from utilities after this second use; collection from homes or smaller commercial installations would be more difficult.

At present, there are no regulations regarding recycling of large-format Li-ion batteries. This condition might be thought to be good for recyclers, who would face no restrictions in process design. However, they would face the significant possibility that restrictive regulations could be imposed after the fact. Therefore, processes must be designed to be compliant with anticipated regulations. In addition, battery technology is still evolving. Recycling processes designed for a specific design or chemistry could become irrelevant quickly.

Automotive Li-ion batteries have only been in commercial use for about 5 years, and it will take some time until they are used in large volumes. And with their long product life (ideally, the life of the car), not nearly enough batteries have reached the end of their lives to support large-scale recycling plants. However, lithium batteries from consumer electronics, as well as processing scrap, are available and could supply feedstock for the fledgling recycling industry for automotive Li-ion batteries. Several recycling methods have been proposed, each with its advantages and disadvantages, as discussed below (process descriptions from Ref. [14]).

2.3.1. Pyrometallurgical recycling (Smelting)

Lithium-ion batteries, after having been dismantled to the module level, are fed to a high-temperature shaft furnace along with a slag-forming agent that typically includes limestone, sand, and slag. The electrolyte and plastics burn to supply some of the energy for the smelting, and the valuable metals are reduced to an alloy of copper, cobalt, nickel,



Fig. 5. Lead–acid battery and Li-ion pack.

and iron. These metals are recovered from the alloy by leaching. The slag contains lithium, aluminum, silicon, calcium, iron, and any manganese that was present in the cathode material. Recycling of aluminum or lithium from the slag is neither economical nor energy efficient. Gas cleanup steps are necessary to avoid release of potentially toxic by-products. This process is operating commercially now, and it is economical for batteries with cathode materials containing cobalt and nickel, but not for newer designs with manganese spinel or LFP cathodes (see Table 2 for approximate cathode and elemental constituent values).

2.3.2. Intermediate recycling process

In this process, commercially used in Canada, batteries undergo size reduction in a hammer-mill and a shaker table separates mixed plastics and metals. Filtering of the aqueous stream leaving the hammer-mill yields mixed metal oxides and carbon, and a liquid stream, which is then dewatered to some extent. The liquid stream can be mixed with soda ash to precipitate Li_2CO_3 , which is subsequently filtered from the solution and sold. The metals (including the Al) can be separated and sent for recycling. However, as with pyrometallurgical recycling, the process is only economical if cobalt and/or nickel are contained in the cathodes of the feed batteries.

2.3.3. Direct recycling

This bench-scale physical process, which has been demonstrated for several cathode types, recovers battery materials for reinsertion into the battery supply chain with little or no additional processing. Breached discharged cells are placed in a container to which CO_2 is added, and the temperature and pressure are raised to bring CO_2 above its critical point. The supercritical carbon dioxide extracts the electrolyte (ethyl methyl carbonate, diethyl carbonate, and LiPF_6) from the cells, and is removed. The electrolyte separates from the gaseous CO_2 , and after further processing, can be recycled for use in batteries if it is determined to be economic. The cells, devoid of electrolyte, undergo pulverization or other size-reduction steps, possibly in the absence of water or oxygen to avoid contamination of materials. Subsequently, the cell components are separated through techniques that exploit differences in electronic conductivity, density, or other properties. Cathode materials may need to undergo re-lithiation prior to reuse in batteries.

This process has the advantage that almost all battery components, including aluminum but excluding separators, are recovered and can be reused after further processing. Most important, cathode materials constitute a potentially valuable product from direct recycling, regardless of cathode type. There is some question, however, about whether the recovered material will perform as well as virgin material, which could have implications for battery power and lifetime, so manufacturers may be reluctant to purchase recycled compounds. Manufacturers of products with tight quality standards have historically been reluctant to purchase recycled materials because of performance concerns [15]. Recovered materials could possibly be used in applications with less-stringent requirements. Product quality will depend on having a known, uniform input stream; mixing cathode materials is likely to reduce the recycled product value.

2.3.4. Discussion

Several factors could help promote Li-ion battery recycling. For example, the large, recognizable packs will be removed from end-of-life vehicles if there is an economic incentive or a regulatory imperative to

do so. They will be labeled to enable identification for proper routing. The recovered batteries could be returned to the original manufacturers (in Europe this may be a requirement [16]), which could enable subsequent recycling. However, while these manufacturers would know what the batteries are composed of, they might not want to be in the recycling business, and they would be required to process recycled compounds that could be obsolete 10 years or more after initial production.

Several developments would facilitate making Li-ion battery recycling economically viable:

- Separation technology for recovered cells that enables processing different chemistries, recycling processes for each cell chemistry, or technology that produces valuable products from a mixed stream;
- Methods for separation of cathode materials after initial processing;
- Greater recycling process flexibility or standardization of battery materials and designs; and
- Assurance that regulations will not impede recycling.

Initial battery manufacturers can promote eventual recycling using design for recycling, including the following steps: inclusion of labels or other distinguishing features, use of a minimum number of different materials, standardization of formats and materials, avoidance of toxic materials (cadmium, arsenic, mercury, halogens, etc.), and designs that allow easy separation of parts. Examples of the last item include a separable cooling system, reversible joining (nuts and bolts instead of welds), and avoidance of potting or adhesive compounds to hold cells in place. Of course, these design changes cannot be made at the cost of any reduction in performance or safety during the battery's useful life. A committee of the United States Advanced Battery Consortium (USABC) is working on design-for-recycling guidelines for United States auto manufacturers [17].

Battery manufacturers and recyclers are working to address the roadblocks to Li-ion battery recycling within the 10 years or so before large numbers of large-format automotive batteries are ready for final disposition.

3. A problem in the model recycling system

Recent events have caused difficulties with the ideal system on which future Li-ion recycling could be modeled. The recycling of Li-ion batteries along with lead-acid batteries is causing trouble at secondary lead smelters. Many current Li-ion batteries are indistinguishable from lead-acid batteries on purpose, so their use is transparent for SLIs, motorcycles, and other applications. However, inclusion of Li-ion batteries in the input stream of secondary lead smelters has resulted in fires and explosions [18]. It is unclear what chemical reactions are occurring; vaporization of the volatile organic electrolytes might be causing the Li-ion cells to explode. Obviously, such events pose a serious danger and must be prevented. These batteries may end up in the wrong recycling stream through honest mistakes. However, because Pb-acid battery recyclers pay for their desired input material (and may charge to take other battery types), some entities may hide different battery types in pallets of Pb-acid batteries to avoid having to pay for their disposition. Recyclers could impose a penalty for sending contaminated loads. However, such contamination is difficult to detect. The used batteries are often delivered to recyclers in huge loads (over 3000/h—up to 70,000/day), and it is nearly impossible for plant staff to do a careful visual screening of the piles on the conveyor belts. Drivers pre-screen the bins or pallets as well, but additional safeguards may be needed. The lead-acid battery recyclers want the Li-ion batteries removed before they get to their plants. The difficulty would be reduced if there were a profitable outlet for the recycling of Li-ion batteries. At present, however, Li-ion manufacturers are moving toward less-expensive materials, which exacerbates the problem.

Similarly, for successful Li-ion recycling, it is important to prevent Pb-acid batteries from entering that input stream. The acid could react

Table 2
Recovery of cathode material maximizes product value.

Cathode	Price of constituents (\$/lb)	Price of cathode (\$/lb)
LiCoO_2	8.30	12–16
$\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$	4.90	10–13
LiMnO_2	1.70	4.50
LiFePO_4	0.70	9

with electrode substrates, and the presence of lead could force the recycler to deal with additional regulations for hazardous material. Clearly, any model system for battery recycling will need to avoid cross-contamination. This issue is being addressed in the United States by the Society of Automotive Engineers and in Europe by EUROBAT. Both groups have active working groups attempting to better define and find solutions to the problems of cross-contamination of battery types in recycling streams [19,20].

While segregation systems for large-format batteries are needed, the optimum separation point in the recycling chain is unclear, and rescreening might still be necessary before final processing. The screening could be based on density differences: Li-ion batteries are likely to be much lighter than Pb–acid batteries. Recyclers are hoping to make their suppliers responsible for providing a stream that is free of undesired type(s) of batteries, and they do not want to pay more. Careful separation is likely to increase recycling costs. Presumably, these increased costs would be borne by consumers. Separation would be facilitated if manufacturers labeled battery components by means of bar codes, RFID chips, or delegated paint color or type (e.g., visible under black light). Labeling, too, would entail costs, and the appropriate placement of labels (on cells, modules, or packs) would depend on the dismantling technology envisioned. Also helpful would be incentives for good recycling practices, and penalties for bad ones.

4. Vision of ideal future system

Ideally, the search for the best battery chemistries and designs will result in a few that satisfy everyone's requirements, and the batteries of a given type would be made as uniform as possible. At a minimum, those that could be recycled together would have at least one distinguishing feature in common, and conversely, one to differentiate them from those that need to be recycled in a different way. Mechanisms would be in place to return all batteries at the conclusion of their (first or second) useful lives. There would be an easy way to route these spent batteries to the appropriate recycling facilities in a safe and legal manner. User-friendly labeling would aid appropriate routing. Regulations would assure safe transport and handling, and discourage any sort of cross-contamination. Sorting/routing could be immediate, via a transfer station or within a unified recycling facility. Separate streams would be processed to produce valuable, high-purity materials that could be reused in batteries or in another high-value product if the recovered material had become obsolete. Recycling process development would be complete. All batteries would be designed with recycling in mind, avoiding irreversible joining and troublesome materials. An alternative to separate processing would also require process development to enable production of a valuable product from a mixed stream (or product separation into valuable streams). Strict industry standards would ensure that recycled products meet the same high quality standards as virgin materials and thereby are accepted for reuse.

Accomplishment of this future vision before large numbers of automotive propulsion batteries have reached the end of their useful lives requires research and planning to continue over the next 10 years or so. It is a daunting task, but if there is a broad commitment from industry and government, it can be done.

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