Substance flow analysis of rare earth elements and precious metals from end-of-life vehicles including next-generation vehicles

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Acronyms and Abbreviations

ABS	Antilock brake system
ANS	Audio and navigation system
ASR	Automobile shredder residue
CD	Compact disc
CV	Conventional vehicle
ELV	End-of-life vehicle
ETC	Electronic toll collection
EV	Electric vehicle
HEV	Hybrid electric vehicle
ICP-MS	Inductively coupled plasma mass spectrometry
LCD	Liquid crystal display
LED (light)	Light-emitting diode (light)
LIB	Lithium ion battery
MCV	Mini size conventional vehicle
NdFeB (alloy)	Neodymium-iron-boron (alloy)
NGVSP	Next-generation vehicle specific part
NiMH (battery)	Nickel metal hydride (battery)
PGM	Platinum group metal
PM	Precious metal
PWB	Printed wiring board
REE	Rare earth element
RPR	Recycling potential ratio
SFA	Substance flow analysis
SmCV	Small size conventional vehicle
StCV	Standard size conventional vehicle

List of tables and figures

List of publications

For Chapter 2 Resource characterization in end-of-life vehicles through dismantling survey:

Yano, J.; Xu, G.; Liu, H.; Sakai, S. Identifying Resource and Toxic Potentials in End-of-Life Vehicles through Dismantling Survey. *Journal of Material Cycles and Waste Management* (major revision, re-review not required as of 27th June, 2019)

For Chapter 3 Scenario analysis for recycling rare earth elements considering recovery technologies:

Xu, G.; Yano, J.; Sakai, S. Scenario Analysis for Recovery of Rare Earth Elements from End-of-life Vehicles. *Journal of Material Cycles and Waste Management*. 2016, 18 (3), 469–482.

For Chapter 4 Recycling potentials of precious metals by selective dismantling:

Xu, G.; Yano, J.; Sakai, S. Recycling Potentials of Precious Metals from End-of-Life Vehicle Parts by Selective Dismantling. *Environmental Science and Technology*. 2019, 53 (2), 733–742.

Chapter 1 Introduction

1. Background

Continuous developing of the world economy and infrastructures has created chances for people in more and more counters and regions to live their lives in a modern lifestyle. As automobiles are widely being considered as one of the indispensable elements for this lifestyle, the global automobiles ownership has increased rapidly in recent decades and reached approximately 1.3 billion units in 2015.[1] Consequently, the global end-of-life vehicles (ELVs) generation has also raised. This trend is particular notable in Asia, Central and South America, and parts of Africa. Gradually, the management of ELVs has become a global concern nowadays.

ELVs generally consist of over 70 % (in mass) of Fe. Thus, they are traditionally treated as a secondary resource pool for Fe, and their recycling is conducted autonomously by dismantlers based on market mechanisms.[2] However, fluctuations in the price of steel scraps and rise of the treatment cost regarding automobile shredder residue (ASR) at times pull down the economic value of ELVs. In addition to Fe and other base metals such as Cu and Zn, some scarce metals are also used in automobiles in minor amounts, but essential for their performances, such as rare earth elements (REEs) and precious metals (PMs). Because of their rareness, those metals are often with high economic value. There is therefore a desire to collect scarce metals as secondary resources from ELVs.

The REEs are a group of 17 chemically similar metallic elements: 15 lanthanides, plus Sc and Y. Among them, La, Ce, Pr, Nd, and Dy are important for automotive industry. For instance, misch-metal, which is a mixture of La, Ce, Pr, and Nd, is a material used for hydrogen storage in nickel metal hydride (NiMH) battery cells; Nd is also a fundamental material used in motor magnets in the form of neodymium-iron-boron (NdFeB) alloys; Dy is added to motor magnets to enhance their stability against temperature induced demagnetization.[3,4] Meanwhile, some PMs which usually refer to platinum group metals (PGMs), Au, and Ag, are also important constituents for many automotive components. For instance, Pt, Pd, and Rh are fundamental materials of catalysts due to their catalytic properties; Au and Ag are widely used in printed wiring boards (PWBs) of electrics of automobiles because of their excellent conductivity and resistance to corrosion.[5]

Nevertheless, recycling ratios of REEs and PMs from ELVs are still low in general,[6] mainly because of issues: product design and end-of-life collection, evolving recycling technologies, and economic unsustainability of secondary production.[7] As the global automotive industry moves toward "greener" (e.g. preference for next-generation vehicles) and "smarter" practices,[8,9] increasing amounts of some REEs and PMs are be used in automobiles. For example, as Figure 1 shows that in Japan the newly registered next-generation vehicles grew 55% to 1.4 million units from 2012 to 2017; while the ownership of next-generation vehicles reached to 8.4 million units in 2017, about 10% of all.[10,11] If the recycling system for ELVs does not react positively to these issues, many of the REEs and PMs in ELVs may not be returned to their material streams for reutilization in the future.

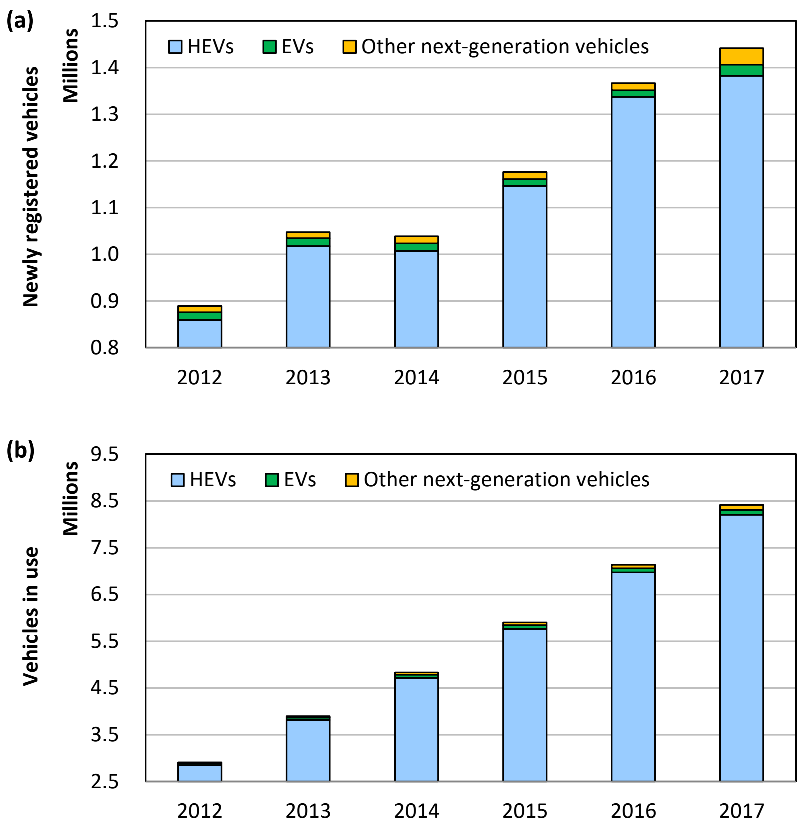

Figure 1. Number of (a) newly registered next-generation vehicles and (b) next-generation in use in Japan. Other next-generation vehicles includes plug-in HEVs and fuel cell vehicles.

According to the UN's Sustainable Development Goals on Responsible Consumption and Production (i.e., goal 12), member states are supposed to substantially reduce their waste generation and achieve sustainable management and efficient use of natural resources within the next two decades.[12] As for ELVs management, it is of great sustainable value to continue increasing the diversity of recycled materials and improving their recycling efficiencies. In Japan, the "Strategy for Ensuring Stable Supplies of Rare Metals" promotes development of new materials to substitute scarce metals in products at upstream of the automotive industry, such as developing low Dy content NdFeB magnets.[13] On the other hand, at the downstream, some ongoing pre-shredding demonstration works have been carried out following this strategy to examine the possibility of recycling scarce metals from automotive parts, such as recycling Au from various PWBs in ELVs.[14]

2. Purposes

Previously, regarding REEs, Alonso et al.[15] reported the REE contents in a typical conventional sedan, and a full hybrid electric vehicle (HEV) with NiMH battery in the US. Cullbrand and Magnusson[16] focused on Nd, Dy, and so on, and investigated their contents in a conventional large car and a hybrid midsize car in Sweden. Graedel et al.[6] estimated global average recycling ratio for REEs. Regarding PMs, Seo and Morimoto[13] investigated PM contents in automotive catalysts in Japan. Widmer et al.[17] investigated PM contents in automotive electrics and electronics in Switzerland, and Restrepo et al.[18] mapped the annual PM flows before and after dismantling and shredding in Switzerland, using a layered model. Additionally, Andersson et al.[19] identified the fates (e.g., functional, nonfunctional, and no recycling) of 25 scarce metals including REEs and PMs passing through the ELV treatment system in Sweden. Through studies such as these, the status quo of recycling REEs and PMs from ELVs can be pieced together.

However, there is still lack of sufficient quantification of future REE and PM recycling potentials in regard to the increased use of next-generation vehicles. Therefore, this study focused on REE/PM-containing parts that are both common in all vehicles and specific to only next-generation vehicles, considered recycling technologies and collection efficiency, and provided an estimation of the recycling potentials of the elements from ELVs in future two decades to inform future recycling goals and schemes.

This study addresses closely on following research questions:

- What are the element constituents per vehicle including next-generation vehicles?
- What are the mass distributions of the REEs and PMs in ELVs recycling system?
- From a mass perspective, which automotive parts/components are crucial to the recycling potentials, and what are the potentials?

3. Structure

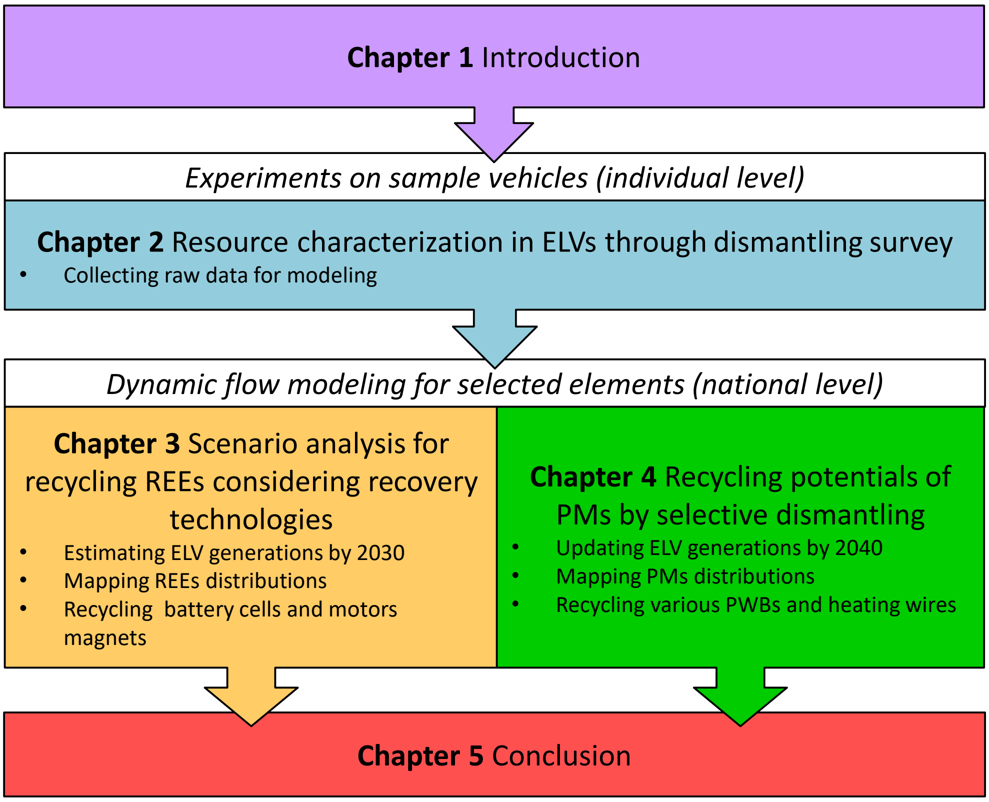

Figure 2. Structure of thesis. ELV, REE, PM, and PWB signify end-of-life vehicle, rare earth element, precious metal, and printed wiring board, respectively.

Figure 2 presents the structure of this thesis, which covers five main chapters as briefly described as follows:

- Chapter 1 outlines the background of this study and clarifies the research purposes.
- Chapter 2 presents a resource characterization in ELVs through dismantling survey, and its role is the basis of the study as an experimental work.
- Chapter 3 addresses on REEs, and it presents the potential recycling scenarios considering recovery technologies.

- Chapter 4 addresses on PMs, and it presents the recycling potentials by selective dismantling.
- Chapter 5 draws the conclusions of this study and recommendations for ELVs management and future researches.

Chapter 2 Resource characterization in end-of-life vehicles through dismantling survey

1. Introduction

Base ferrous metals and non-ferrous metals respectively account for about 70–80% and 5–10% of an automobile.[20] The remaining weight consists of other materials such as plastics, textiles, and rubbers. Ferrous alloys contain critical metals such as Ni, Cr, Mn, and Mo,[21] while automotive electrical devices contain various metals such as Co, Ta, In, REEs, and PMs.[15,16,18,22–26] As Al and Cu are economically valuable, and they tend to be contained in ASR during the recycling. Therefore, some studies tried to identify the distribution during the shredding procedure to improve collection efficiency and reduce environmental burdens.[27,28] In general, although valuable elements are used in various products including vehicles, it is still difficult to recycle many of them.[6] When contained elements are not recycled, either they result in a sink at the landfilled site or become contaminated fractions in the recycled products.[19,29] Such behavior of constituent elements depends on the overall recycling and treatment flows employed.

A dismantling survey has advantages: (1) providing detailed information about the composition of an ELV on weight and/or material basis, and parts with location; (2) providing samples for chemical analysis; (3) highlighting the challenges and their countermeasures during the on-site dismantling process and throughout the ELV management system. Dismantling time can also be measured,[23,30] as this is an important economic consideration for vehicle dismantlers. On the other hand, the disadvantages of the dismantling survey are that not many vehicles can be dismantled and therefore the findings are necessarily based on a small sample of representative vehicles. It is also difficult for researchers along to conduct a dismantling survey, therefore, cooperation with vehicle dismantlers is also essential.

To improve the recycling efficiency of constituent elements within an ELV management system, the content of valuable elements per vehicle or per component should be identified. The purpose of this chapter is to obtain useful information for dynamic flow analyses. A dismantling survey was conducted from 2012 to 2015. Some collected parts underwent chemical analysis to identify and quantify their constituent elements. Combining the findings with data from previous studies, the elemental contents per ELV were estimated. Previous studies clarified the elements contained in each vehicle using measured data by means of chemical analysis including sampling collection and statistical data from part suppliers and automobile manufacturers.[16,17,22,25,31] This chapter employed such former approaches.

2. Materials and methods

2.1. Vehicle samples

According to the Road Vehicle Act, conventional vehicles (CVs) in Japan (Appendix 1),[32] vehicles are categorized into three classes: (1) mini size vehicle less than 660 cc displacement; (2) small size vehicle between 660 and 2,000 cc; (3) standard size vehicle of more than 2,000 cc. During the survey, in total six ELVs were dismantled as listed in Table 1. Among them, four were CVs and two were next-generation vehicles. Of the CVs, a typical vehicle and a luxury vehicle were selected and dismantled to represent the small size conventional vehicle (SmCV) and standard size conventional vehicle (StCV), respectively. The next-generation vehicles dismantled were an HEV and an electric vehicle (EV). Considering the about 13 years average lifespan of passenger vehicles in Japan,[33] the four dismantled vehicles represent the types of vehicles that are increasingly reaching the end-of-life stage. In order to clarify the differences between vehicles in terms of technical innovations such as electricalization during this decade, two vehicles, a mini size conventional vehicle (MCV) and an EV produced in approximately 2010 were also dismantled. The MCV produced in 1999 and 2009 belong to the same vehicle family.

Table 1. Vehicle samples in dismantling survey

Type and size	**Conventional vehicles**				**Next-generation vehicles**	
	Mini size		**Small size**	**Standard size**	**–**	**–**
Production year	1999	2009	1997	1997	1998	2011
Abbreviation	MCV99	MCV09	SmCV	StCV	HEV	EV
Displacement (cc)	658	658	1498	2997	1496	–
Weight (kg)	760	810	1100	1490	1240	1520
Survey date	2015	2015	2013	2012	2012	2014

Weight information presented here was obtained from brochures.

HEVs in particular have undergone rapid technical innovation since they were first released in 1997. Model changes every 6 years have resulted in three generations of dismantled HEVs: 1st generation in 1997–2003, 2nd generation in 2003–2009, and 3rd generation in 2009–2015. To clarify the difference between generations, some components in the 2nd and 3rd generations (Appendix 2) were additionally collected. The 1st generation components were collected from an HEV listed in Table 1. Hybrid transmission, electric power steering, and an air compressor were considered because that motor magnets in these components contain REEs such as Nd and Dy.[34] A hybrid transmission comprises two motors: a driving motor and a generator.

2.2. Dismantling procedure

The procedures followed during the dismantling survey are (1) brief dismantling, (2) detailed dismantling, and (3) chemical analysis, more details are described in Appendix 3. At first, each part (engine, seat, door, etc.) was briefly dismantled by hand or with tools (electric tools and gas-powered cutting torch). The weights of each part were then recorded. This allows the locations of the parts to be identified and taken into consideration for their separate collection during the dismantling process, for efficient material recycling and recovery of elements. Parts were categorized into nine locations (Appendix 4): engine room, four interior points (front, seat area, rear, and trunk), two exterior points (suspension and others), the remaining body, and others.

Following a brief dismantling process, a vehicle was also hierarchically categorized into subparts (if necessary) and components during the detailed dismantling process. Subparts refer to assemblies such as motors that are more detailed than parts but still consist of some components. A component is the smallest category, such as a magnet, PWB, resin and textile. Although PWBs and compound textiles are still composed of several materials, they were categorized as a single component. The definition of parts, subparts, and components in this thesis are illustrated in Figure 3. Based on this categorization, subparts and components were further separated and weighed after brief dismantling. Finally, some components or parts were collected as samples for chemical analysis, as explained in next section.

When weighing during the brief and detailed dismantling processes, material composition was also recorded. Material composition was categorized as metals (e.g. Fe, Cu, and Al), plastics, textiles, glass, rubbers, and others. Materials including subparts that were too complicated and/or difficult to identify the material at the dismantling site were categorized as “unidentified”.

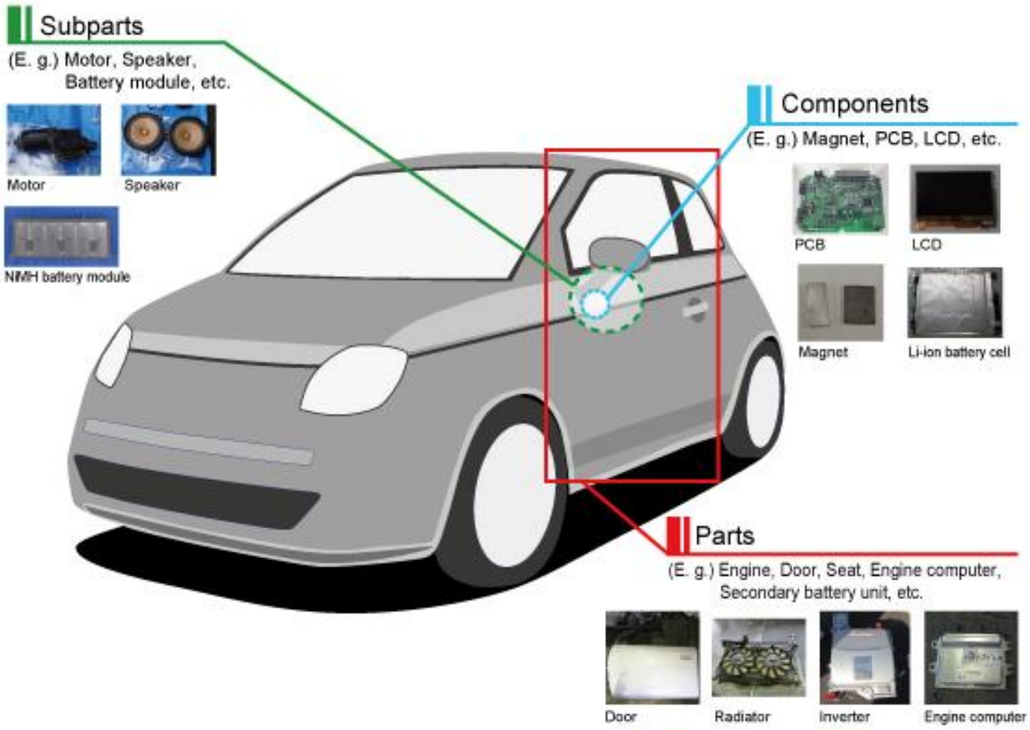

Figure 3. Definition of parts, subparts and components in vehicles

2.3. Chemical analysis

Sampled components, which would contain valuable elements, were selected by reviewing previous studies and interviews with dismantlers during the dismantling survey. As listed in Table 2, the selected components included PWBs, magnets, second battery cells, catalysts, and so on.

A total of 61 elements and one ion (i.e. Cr^{6+}) were considered, as shown in Figure 4. Pb, Cd, Hg, and Cr^{6+} are the most representative toxic elements, and were therefore subject to reduction targets in the action plan established in 2002 by Japan Automobile Manufacturers Association.[20] Br was considered because it could be used as an indicator of whether components, especially PWBs, contained brominated flame retardants. From the viewpoint of resource recovery, common metals (Fe, Cu, Al, Zn, Sn, and Mg) are considered because they are essential to the economic viability of ELV recycling. The Japanese Government defines 47 elements including 17 REEs (Sc, Y, and 15 lanthanoid) as critical metals,[35] as shown in Appendix 5. Among them, 9 elements (V, Cr, Mn, Co, Ni, Ga, Mo, In, and W) are particularly defined as critical storage metals, which are saved to meet 60 days' demand. These critical metals defined in Japan and other precious metals (Rh, Ag, and Au) are also considered in this study.

Table 2. Components in chemical analysis

Components	**Parts**	
PWBs	Engine computer	Steering computer
	ABS computer	Computer shift lock
	Speed meter	Air-bag control
	Audio and car navigation system	Switch in glove compartment
	Switch for mirror adjustment	Door switch
	Handle switch	Door lock receiver
	ETC reader	PWBs with wiring harness
	Unidentified PWBs	NiMH battery/LIB
	Hybrid computer	Inverter
	Capacitor assay for EV	
Magnets	Windscreen wiper motor	Power window motor
	Radiator fan motor	Blower fan motor
	Rear brake	Rear speaker
	Electric power steering motor	Electric air compressor motor
	Hybrid transmission: driving motor	Hybrid transmission: generator
Battery cell	NiMH battery	
	LIB	
Catalyst	Catalytic converter	
LCD monitor	Audio and navigation system	
LED Light	Light	
Ceramic	O_2 sensor	
Earth electrode	Spark plug	
Metal terminal	Rear glass	
Heating wire with glass	Rear glass	
Resins and textiles	Interior materials	

The qualitative and quantitative analyses primarily consisted of inductively coupled plasma optical emission spectrometry and inductively coupled plasma mass spectrometry (ICP-MS). The chemical analysis is resource intensive, for each collected component one time test was therefore ran. To enforce the representativeness and accuracy, the samples were manually cut into pieces of approximately 50 mm. This particle size was further reduced as small as possible and homogenized to < 1.5 mm by means of a cutting mill and an ultra-centrifugal mill in the pretreatment. After these powdering treatments, samples of 1 g were dissolved using H_2SO_4 and HNO_3. If residues remained after acid dissolution, alkali fusion was conducted using Na_2CO_3 and H_3BO_3. Motor magnets were first demagnetized at 500 °C to allow manual separation. Then wavelength dispersive X-ray fluorescence spectrometer was used for screening contained elements because several types of magnets (ferrite magnets and permanent magnets) are used in vehicles. Only the magnets from hybrid transmission, electric power steering, and air compressor, which could contain REEs according to the Japanese Automobile Recycling System,[34] were additionally analyzed through ICP-MS. Atomic absorption spectrophotometry, absorption photometry, cold vapor atomic absorption spectrophotometry, and combustion ion chromatography were also applied for some elements. More detail is provided in Appendix 6.

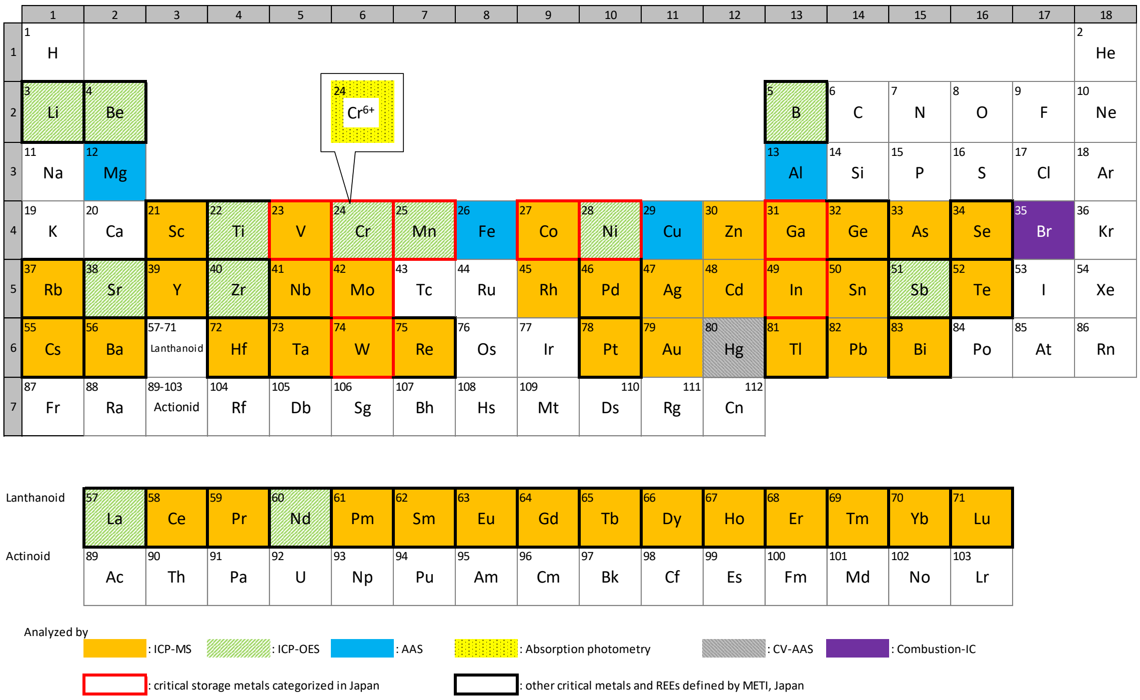

Figure 4. Elements considered in chemical analysis. AAS, CV-AAS, and Combustion-IC signify atomic absorption spectrophotometry, cold vapor atomic absorption spectrophotometry, and combustion ion chromatography, respectively.

2.4. Estimation of element contents per vehicle sample

The content of elements per ELV was estimated using the weight of components and material composition, and the element content of each component, as described in Eq. 1. The former two were obtained via the dismantling survey and chemical analysis. For element contents or concentrations, secondary data from previous studies within Japan[22,36–38] were also used because not all parts were analyzed in the present study.

$$E_i = \sum_j (W_j \times C_{i,j}) \qquad \text{Eq. 1}$$

Where *i* and *j* signify the element (e.g. Fe, Cu, Al, …) and part or subpart, respectively; E_i is the content of element *i*; W_j is the weight of part/subpart *j*; $C_{i,j}$ is the concentration of element *i* in part/subpart *j*.

3. Results and discussion

3.1. Weight distribution

The results of weight and proportional distribution based on equipment points are compared in this section. Additionally, as an example of the specific components, PWBs from every vehicle and secondary battery units from next-generation vehicles are also reported. Overall, Figure 5 shows the weight and the proportional distributions based on equipment locations. In the case of the four CVs, the remaining body accounted for 21–27% and the engine room accounted for 13–26% of the total weight in every vehicle. This was because various heavy parts including the engine and transmission were concentrated within this area. Differences in engine size (displacement volume) resulted in differences in the weight of the remaining body, engine room, and exterior. Comparing the mini motor vehicles produced in 1999 and 2009, the proportional distributions were quite similar. There appear to be few inter-generational differences in equipment locations, presumably because they belong to the same vehicle family. In the case of the HEV and EV (next-generation vehicles), most of the next-generation-specific components were located in the engine room, including the hybrid transmission (38.2 kg) and invertors (25.7 kg) in an HEV. In an EV, the transmission (99.0 kg), inverter (17.0 kg), and converter (18.0 kg) were located in the engine room. On the other hand, the NiMH battery (78.2 kg) was located in the rear interior area of the HEV while the Lithium ion battery (LIB) (294 kg) was located in the other exterior area (under remaining body).

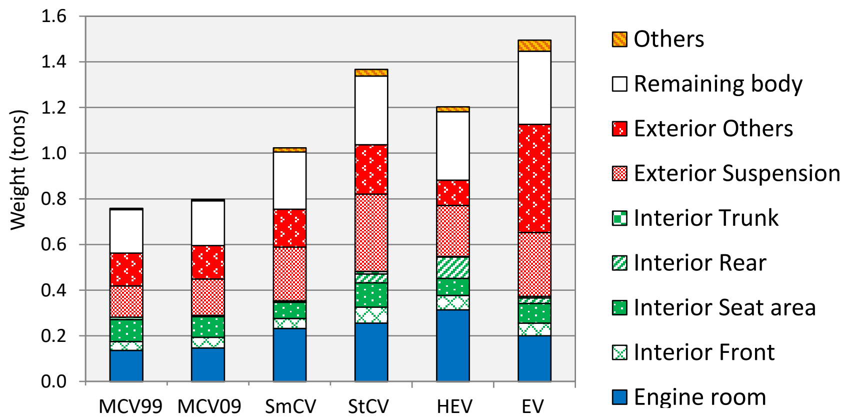

Figure 5. Weight distribution based on equipment locations in vehicles

Ferrous metals accounted for approximately 59–71% while non-ferrous metals and non-metal materials except "others" accounted for 8–12% and 14–19%, respectively. No substantial difference was observed among CVs. Next-generation vehicles contain heavy driving motors and generators, and therefore had higher copper content than CVs. The "unidentified" content in next-generation vehicles was larger than that in CVs because the former had unknown parts that could not be identified at the dismantling site. Japan Automobile Manufacturers Association reported that ferrous and non-ferrous metals account for approximately 70–80% and 5–10% of a passenger vehicle, respectively;[20] and that the ferrous, aluminum, and plastic contents of a vehicle as of 2001 accounted for 73.0%, 6.2%, and 8.2%, respectively.[39] According to the demonstration in Italy as of 2011, ferrous scrap in treated vehicles accounted for 72%. Compared with those data, the ferrous and aluminum compositions in the present study tended to be lower while the plastics composition was similar. The metal composition in our results might be underestimated, because the complex composition of some components meant they were categorized as unidentified materials. Details are provided in Appendix 7.

Regarding PWBs, Figure 6 shows the total weight and proportional distribution of PWB equipment locations. Except in StCV, the weight of PWBs in CVs was less than 1.0 kg. Of the PWBs in StCV, 54% by weight were associated with the audio player. Even so, PWBs were present in various electrical components, resulting in a total PWB weight of 4.1 kg. An HEV contained 3.8 kg of PWBs, of which approximately 39.9% was derived from HEV specific components: hybrid computer, invertor, and NiMH battery. Of the dismantled vehicles, an EV had heaviest content of PWBs, at 7.9 kg. PWBs related to next-generation vehicle specific components, which accounted for 70%, and the other PWBs were distributed in various electric components. Except for the EV, 60–90% of PWBs were located in the "Front of interior". Separate collection of PWBs from that area during dismantling will be an effective approach.

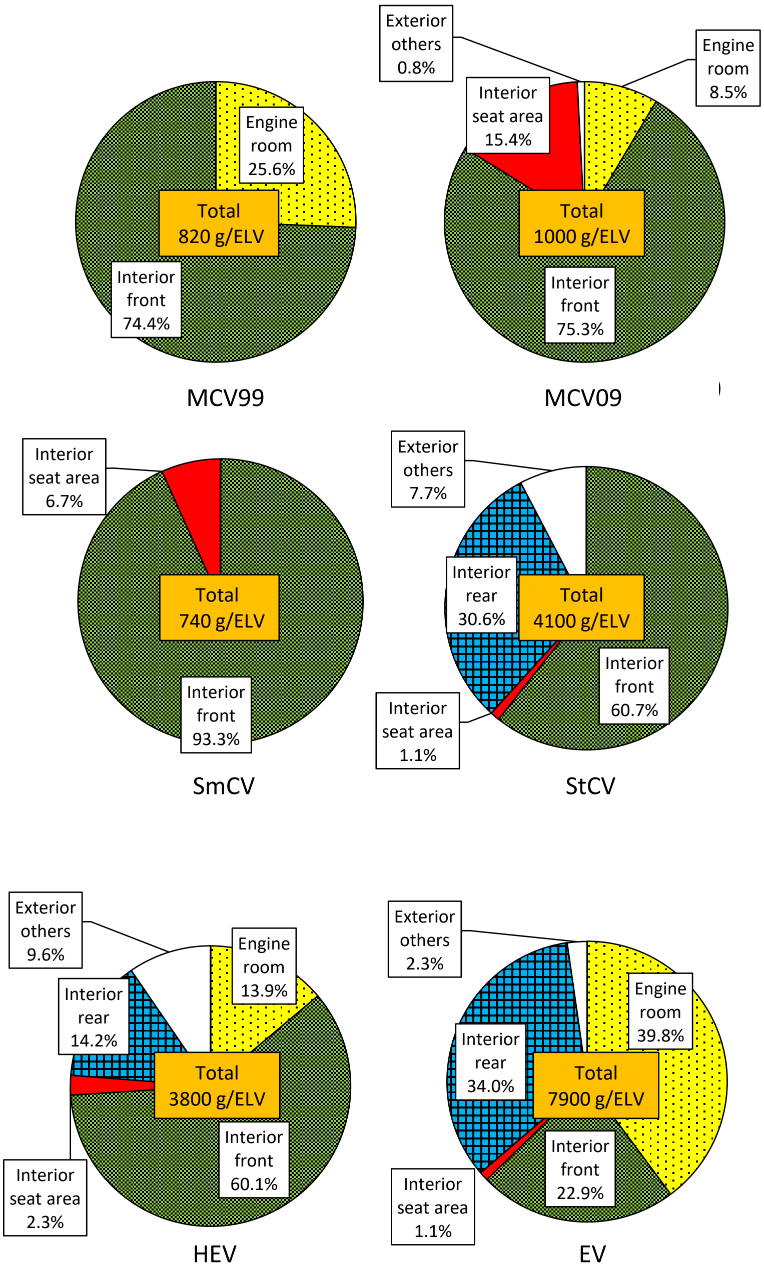

Figure 6. Weight of PWBs in vehicles according to equipment locations

A battery comprised of battery module, PWBs, cables, and covers. A module was further comprised of battery cells. As shown in Table 3, the NiMH battery cell was weighed for each generation of HEV, showing: 32.8% weight reduction of battery cells from 1st (43.3 kg) to 3rd generation (29.2 kg), achieved through reducing the number of cells. Ministry of the Environment, Japan also reported that NiMH battery cell from HEV produced in 2011 was amounted to be 16.4 kg.[40] The LIB in an EV comprised 192 battery cells weighing 181 kg. Because these battery cells were heavier than other components, the amount and variation of elements contained in the battery cells would significantly affect the resource recovery potential from next-generation vehicles, as discussed in the section on chemical analysis.

Table 3. Number and weight of battery cells in batteries

Characteristics of battery			**NiMH battery**			**LIB**
			1st HEV	**2nd HEV**	**3rd HEV**	**EV**
Electric voltage	(V/battery)	A	288	202	202	360
Discharged capacity	(Ah/battery)	B	6.5	6.5	6.5	66.7
	(kWh/battery)	C = A×B	1.9	1.3	1.3	24.0
Number of modules	(module/battery)	D	42	28	28	48
Number of cells	(cell/module]	E	6	6	6	4
	(cells/battery)	F = D×E	252	168	168	192
Weight of cell	(g/cell)	G	172	173	175	940
	(kg/battery)	H = F×G	43.3	29.1	29.2	180.5
Total weight of unit	(kg/battery)	I	78.2	39.0	41.0	294.0
Percentile of cell	(%)	J = H / I	55.4	74.4	71.2	61.4

Total weight of a battery includes module, PWB, cables, and covers.

3.2. Chemical composition

The PWBs contents of engine computers are shown in Figure 7 (a). Elements that tended to exceed 10,000 ppm were: Fe, Cu, Al, Sn, and Br; those exceeding 1000 ppm were: Zn, Ni, B, Ti, Sb, and Pb; and those exceeding 100 ppm were: Mn, In, and Sr. The results indicate that although PWBs contained various elements with a wide range of concentrations, most of them showed lower concentrations, namely less than 100 ppm. The elements at concentrations exceeding 10,000 ppm were Al, Mg for SmCV, MCV99, and MCV09, Cr for StCV, Zr for MCV99 and MCV09, La for StCV, and Ce as shown in Figure 7 (b). Related to Pt, Pd, and Rh which are generally famous elements contained in catalyst, Pt concentration differed between vehicles: 430 ppm and 510 ppm for MCV09 and StCV while not detected for SmCV. Pd was 1100–3900 ppm except for StCV which was not detected. Rh concentration was 96–490 ppm. Regarding REEs, each vehicle contained more than 1,000 ppm La and Ce (except for La for MCV99). Additionally, the Nd content of MCV09 was 9,800 ppm. As shown in Figure 7 (c), the elements at concentrations exceeding 10,000 ppm were Fe, Mn, Co, Ni, La, and Ce for NiMH batteries, and were Cu, Al, Mn, Co, Ni, and Li for LIB. Although there was not much difference between generations for NiMH battery cells, La concentration increased from 20,000 ppm for 1st HEV to 62,000 ppm for 3rd HEV. The NiMH battery cell contained REEs as misch metal in the negative electrodes. Considering that the total weights of battery cells were much heavier (43 kg for NiMH battery cells of 1st HEV, 180 kg for LIB cells of EV: see Table 3), the battery cells were expected to have higher recovery potential than the other components.

Although every vehicle contained many motor and speaker magnets, most were ferrite magnets rather than the NdFeB magnets containing REEs. Magnets used in the air compressor motor, hybrid transmission (driving motor and generator), rear brake for EV, and electric power steering motor for EV contained REEs at concentrations of approximately 300,000 ppm (30%): Of these, Nd and Dy ranged between 82,000–280,000 ppm and 4,000–99,000 ppm, respectively. Magnets in wiper motor, blower fan motor, speaker, and electric power steering motor of second-and third-generation HEVs contained Sr at 80000–120000 ppm. Although Co was also detected frequently, the concentration was few: less than approximately 30,000 ppm (3%). It should be noted that, for the magnets analyzed by ICP-MS in addition to X-ray fluorescence. Details about element concentrations of each magnet contained in ELVs are provided in Appendix 8.

In addition, The catalyst contained more than 10,000 ppm Ce with over and more than 1,000 ppm Ti, Zr, Pd, and La. Liquid crystal display (LCD) monitors in both the StCV and first generation HEV showed similar results, containing more than 10,000 ppm Sr and more than 1000 ppm B, Ti, and As. The ceramic contained in the O_2 sensor contained more than 10,000 ppm W, Y, and Zr, and more than 1000 ppm Pt. Specifically, Zr accounted for 50% (500,000 ppm). The earth electrode of the spark plug contained more than 10,000 ppm Mn and Ni, and more than 1000 ppm Cr. Specifically, Ni accounted for 54% (540,000 ppm). Except for the common metals, the elements contained in the metal terminal of the rear glass were present at concentrations of less than 1,000 ppm: 360 ppm Ti, 210 ppm As, and 780 ppm Pb. The heating wire of the rear glass contained more than 1,000 ppm Ti and Ce. This Ce is thought to be derived from additives in the glass rather than from the heating wire.[4] Furthermore, heating wire contain 260 ppm Ag while Ministry of the Environment, Japan also reported it contained 100–200 ppm.[40]

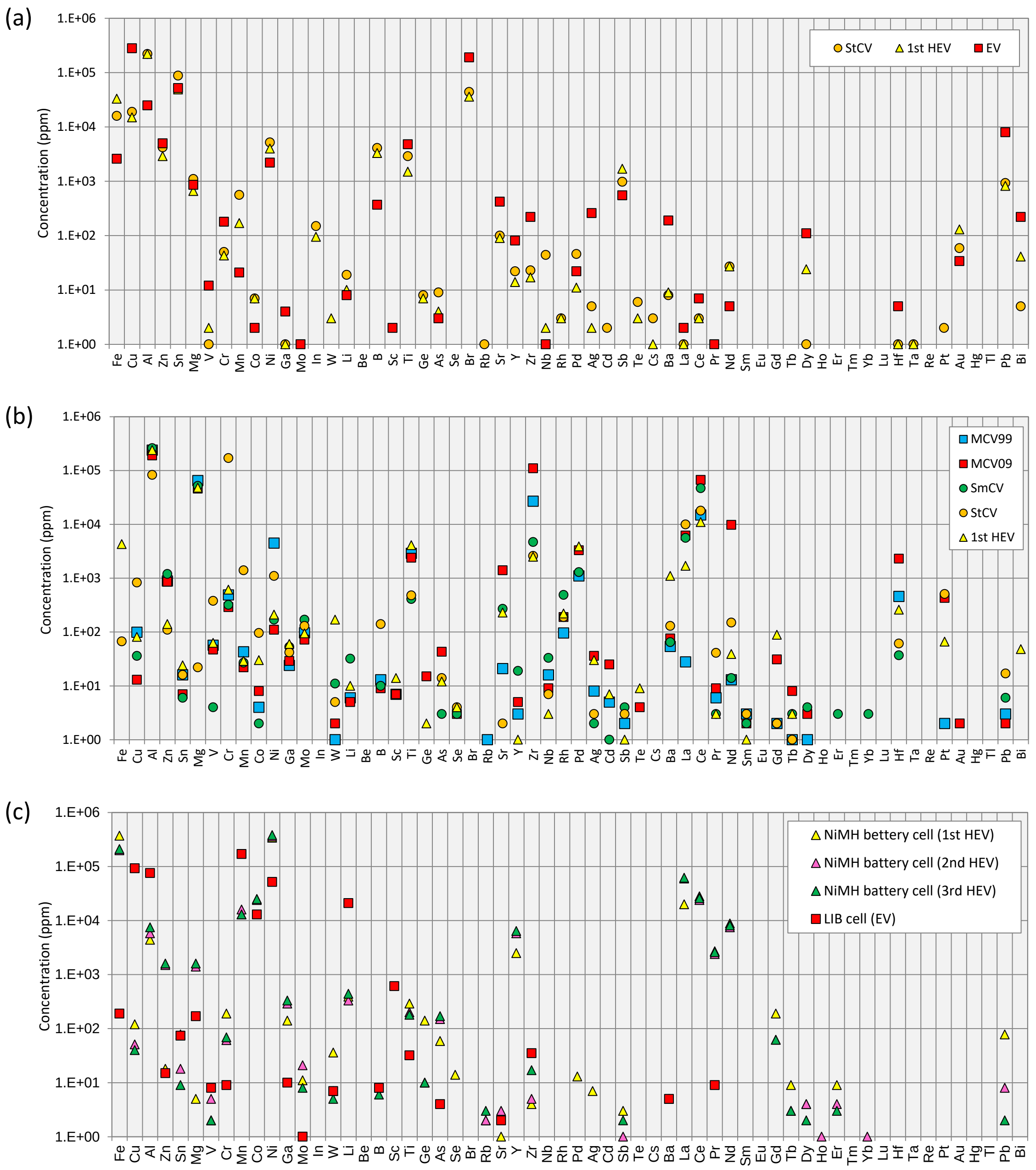

Figure 7. Elements contained in PWBs in: (a) PWBs of engine computers, (b) catalyst in catalytic converter, and (c) secondary battery cell

3.3. Element contents per vehicle sample

Figure 8 shows the estimated contents of elements per ELV. Next-generation vehicles tended to contain a greater variety of elements at higher concentrations than did CVs.

Furthermore, among the CVs, StCV tended to contain higher amounts of elements. For instance, Co content in next-generation vehicles exceeded 1,000 g because of the NiMH or LIB, while Co content in CVs was less than 5 g. REE contents were 3100 g for the first generation HEV and 710 g for EV, compared with 22 g for MCV99, 47 g for MCV09, 64 g for SmCV, and 56 g for StCV: REEs in CVs mainly derived from the catalytic converter and rear glass, whereas those in next-generation vehicles derived from the driving motor and generator magnets. The Sr content of the StCV was 230 g, because luxury-class CVs contain many automatic electronic parts that include motor magnets. PM contents (sum of Rh, Pd, Pt, Ag, and Au) were 3.1 g for MCV99, 4.5 g for MCV09, 4.7 g for SmCV, 5.6 g for StCV, 7.9 g for the first generation HEV, and 9.6 g for EV, respectively. Catalytic converters were the dominant contributors of precious metals, and PWBs were also a contributory factor in EVs. Zr, mainly contained in the O_2 sensor and catalytic converter, was within the range 6.3–42 g. It should be noted that ELVs also contained toxic elements. Pb content ranged from 4,500 to 6,900 g, which was mainly derived from lead–acid battery: additionally, NiMH battery or LIB, and next-generation vehicles still equipped with lead–acid battery.

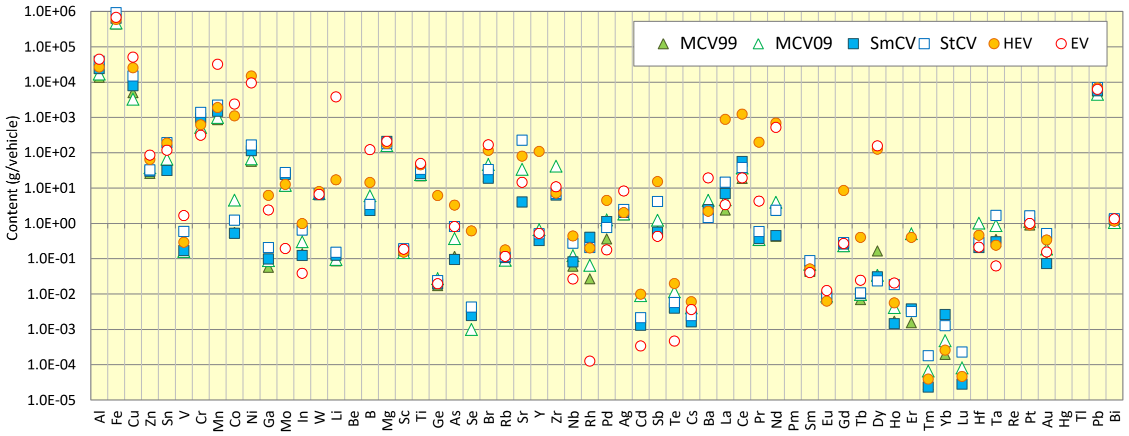

Figure 8. Element contents in end-of-life conventional and next-generation vehicle samples

Some previous studies have reported the contents of certain elements in ELVs. The collection and comparison of such primary data are significant because the elements contained in an ELV differ by various restrictions: vehicle type, automobile producer, production year, and so on. Figure 9 compares the findings of the present study with those of previous studies. The estimation methods of these previous studies are summarized in the electronic supplemental materials.

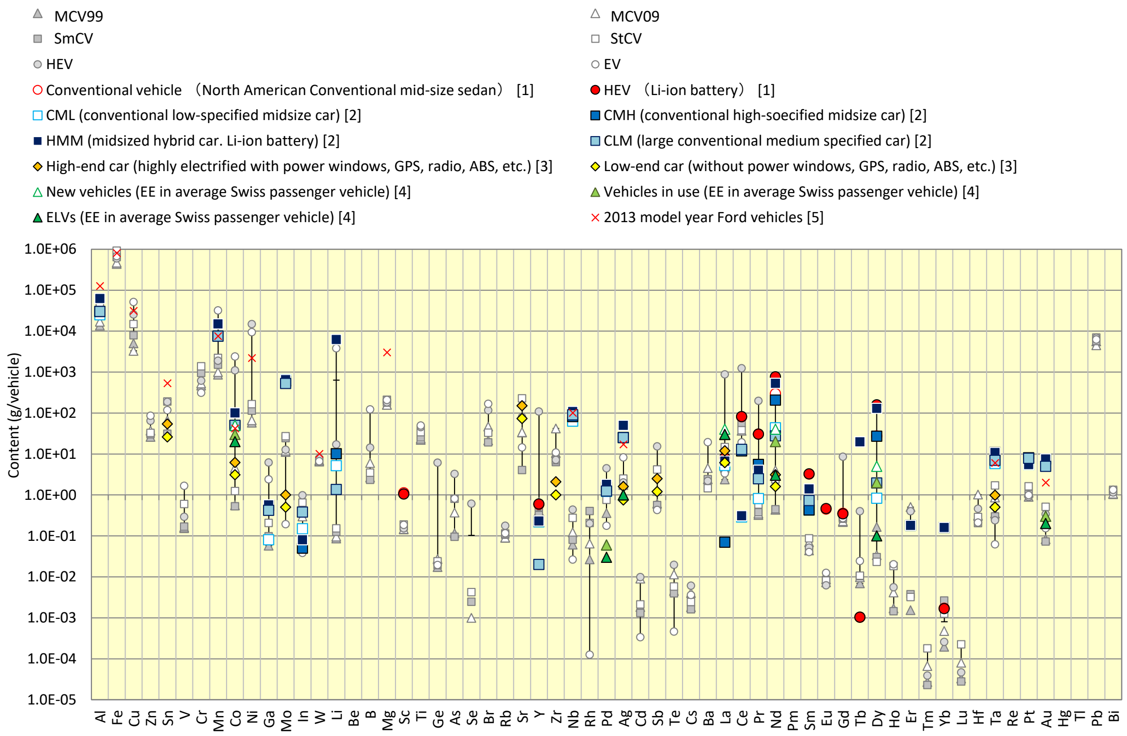

Figure 9. Comparison with previous studies. [1, 2, 3, 4, and 5] signify studies from Alonso et al.[15], Cullbrand and Magnusson[16], Widmer et al.[17], Restrepo et al.[18], and Field et al.[26], respectively.

Widmer et al.[17] found that the most common elements per vehicle were: 73–150 g Sr from magnets; 26–55 g Sn from resistors; 6.1–23 g La from magnets. Sr content in this study showed similar range 4–230 g and was derived from motor and speaker magnets. Although resistors were not considered in this study, Sn was contained in PWBs and ranged from 36 to 190 g. Alonso et al.[15] estimated that REEs content per vehicle was 440 g for a conventional vehicle, 4.5 kg for HEV equipped with a NiMH battery, and 1.1 kg for an HEV with a Li-ion battery. In comparison, the REEs content in the present study was 22–64 g for CVs. This difference was because the CVs in this study were relatively old and therefore used relatively few NdFeB magnets containing REEs. Nguyen et al.[31] estimated that REE contents in CVs and trucks were 4–30 g and 10–24 g, respectively. It showed similar results with this study. The REE content of HEV with NiMH battery and of EV with LIB in this study were 3.1 kg and 0.71 kg, respectively. These contents were also smaller than those reported by Alonso et al.[15], which might be attributed to factors such as the year of production and vehicle size (weight of parts). Cullbrand and Magnusson[16] also clarified the element contents of four types of vehicles.

Du et al.[25] compared 25 elements in conventional passenger vehicles between previous studies. Through the comparison, they indicated two challenges related to methodology and definition of subsystems. With regard to the dismantling process, the present study categorized a vehicle into three hierarchical assemblies: parts, subparts, and components after recording equipment locations. In comparison, Du et al.[25] modeled a vehicle as a hierarchical assembly of subsystem (like "steering"), parts, and components. The difference was that the present study considered equipment locations, whereas Du et al.[25] focused on the function of components (subsystem). It was pointed out that categorization methods differed between studies, therefore hindering comparison between the studies. Overall, it should be noted that these constituent elements might be lost without the introduction of a recovery system.

3.4. Uncertainty

There are uncertainties in each of the procedures in this study. Although it is impossible to eliminate these uncertainties completely, it is important to identify their causes. The chosen method of estimation also affects the results. The element contents of a vehicle can be estimated using: 1) data measured by chemical analysis including sampling collection and/or 2) statistical data from parts suppliers and automobile manufacturers. Furthermore, considering domestic resource potentials related to ELVs management, the amount of element *i* contained in an ELV generated in year *Y* can be described by developing Eq. 2 as below.

$$P_i^Y = \sum_y^Y \left(N^y \times \sum_j \left(R_i^y \times n_i^y \times w_j^y \times c_{i,j}^y \right) \right) \quad \text{Eq. 2}$$

Where Y is the counted year; y is the production year (considered as the first registered year); P_i^Y is the amount of element i contained in ELV generated in year Y; N^y is the number of ELV generated in year y; R_i^y is the installation rate of part or subpart j produced in y; n_i^y is the number of part or subpart j produced in y; w_j^y is the weight of part or subpart j; $c_{i,j}^y$ is the concentration of element i in part or subpart j.

The number of ELV generated in year y, N^y, can be obtained from statistical data. Installation rates, R_i^y of the types of parts recently fitted to vehicles, such as electronic parts, affect the resource potential. For instance, electronic power steering motors have been introduced since 2002. The installation rate of such system increased from 28% to 74% during 2003–2010.[41–43] The number of installations varies within the range n_i^y for optionally equipment such as (rear) speakers and seat adjustment motors. The weight of part w_j^y is affected by the technological level (the production year y). The concentration of contained element $c_{i,j}^y$ is affected by chemical analysis. Uncertainties during chemical analysis were associated with the representativeness of the samples; and with the pretreatment methods and accuracy of analysis. Generally, the representativeness of dismantled vehicles affects R_i^y, n_i^y, w_j^y, and $c_{i,j}^y$. As mentioned previously, some components were difficult to separate and/or to identify the constituent materials. This might lead to underestimation of the ferrous and non-ferrous metal compositions. The elements contents per ELV in this study might be underestimated because some parts such as sensors were excluded due to lack of data.

4. Conclusion

This study conducted a dismantling survey to obtain useful information for ELVs management. Element contents per ELV were estimated using data obtained from chemical analysis and from previous studies.

The conclusions of the study are as follows:

- The dismantling survey allowed identification of material composition and parts distribution; For instance, the PWB content ranged from 0.74 to 4.1 kg for CVs, 3.8 kg for HEV, and 7.9 kg for EV. Chemical analysis identified the various valuable and/or toxic elements contained in each part.
- Related to the element contents per ELV, next-generation vehicles tended to contain a wider variety of elements at higher amounts than did CVs. Furthermore, among the CVs, StCV tended to contain higher amounts of elements. Compared with previous studies, differences of one or two orders of magnitude were observed. Substance flow analysis and life cycle assessment studies for evaluating ELV recycling systems should consider these uncertainties.
- The use of next-generation vehicles has been promoted; however, as they contain specific components such as hybrid transmission and NiMH battery or LIB, it is necessary to develop ELV recycling systems that specifically consider the recovery of elements from next-generation vehicles.

In developing future ELV management systems, the following factors should be considered:

- Identifying and categorizing parts that contain valuable elements, and developing appropriate recovery and control schemes.

- Developing an ELVs management system to cover the increase in next-generation vehicles and highly electralized vehicles.
- Developing indicators to monitor and evaluate ELV recycling systems from the viewpoints of not only recycling rate but also resource efficiency.

Related to challenges for future ELV management systems, the findings of this study indicate that PWBs and magnets used in some motors (air compressor, electric power steering), and in next-generation-specific components (hybrid transmission and NiMH battery or LIB cells) contain larger amounts of valuable elements. Although catalyst systems also contained valuable elements such as Ce and Pd, this unit is already subject to stringent collection during the dismantling process and its recycle.[44] Some challenging to recovery REEs and other critical metals have been demonstrated and introduced in the world.[45,46] Considering that the number of end-of-life next-generation vehicles will increase in the future, the resource value of ELVs will increase significantly. To avoid consigning valuable and/or toxic elements to ASR, separate collection and recovery procedures will be important. Reuse of components will also contribute to reducing the demand for such elements by the automobile industry. Monitoring the concentrations of representative elements in ASR is one useful approach to evaluating an ELVs recycling system.

Chapter 3 Scenario analysis for recycling rare earth elements considering recovery technologies

1. Introduction

The REEs are very essential for modern industry because of their unique physical and chemical properties. Therefore, It has been said that for modern industrialized countries, oil is the "blood," steel is the "body," and REEs are the "vitamins". The European Commission deems REEs as the most critical raw materials group, with the highest supply risk.[47] As also acknowledged by the U.S. Department of Energy in their medium-term (2015–2025) criticality matrix, Dy, Nd, Tb, Eu, and Y are the top-five critical REEs in relation to their importance for clean energy, and also with respect to risk affecting their supply.[48] In automotive industry, REEs can be widely found in some components such as NiMH battery cells and motor NdFeB magnets.[3,4]

For the past two decades, the supply of REEs was in a near-monopolistic condition and related mining practices were environmentally unsustainable. China holds 50% of the global REE mineral reserves, and accounted for 94% of the market share in 2011.[49] However, because of a series of environmental concerns such as toxic water and radioactive mud,[50] and increased pressure on domestic demand, China lowered its REEs export quota from 50000 t in 2009 to 31000 t in 2012,[3] which is a reduction of about 38%. Consequently, in recent years a great deal of research seeking other sustainable use of REEs appeared worldwide.

It is estimated by Zhou et al.[51] that REE demand from clean technologies worldwide will reach 51900 t rare earth oxides in 2030, Nd and Dy, respectively, comprising 75% and 9%, while these two elements comprise 15% and 0.52% of the global REE resources, respectively. Moreover, Seo and Morimoto indicated that the domestic demand of Dy in Japan in 2030 was estimated to be 1100–1700 t, of which about 500–1000 t was to answer the demand of the automotive industry and 65–95 t to answer the demand for home appliances.[13]

Nowadays, many technologies have been developed for the recovery of NiMH batteries and NdFeB magnets, including pyrometallurgical (e.g., electro-slag refining, liquid-metal extraction glass-slag method, and direct melting), hydrometallurgical, and gas-phase extraction methods.[52–60] Additionally, some enterprises such as Hitachi, Honda Motor, Japan Metal & Chemicals, and Umicore have announced technologies for recovering REE, but the details have not yet been disclosed.[3,61–64] In 2010, less than 1% of REEs were actually recycled from end-of-life products.[65] Historically, only small amounts of REEs have been used, and are dispersed throughout many components of a vehicle. However, increasing amounts of REEs, especially the highly critical elements Dy and Nd, are being used in vehicles (motor NdFeB magnets and NiMH battery cells).[66]

This chapter and the followed Chapter 4 are the dynamic flow modeling for selected elements based on experiment data gathered in Chapter 2. This chapter focuses on REEs contained in components of passenger's CVs and HEVs. We developed quantified scenarios for exploring the recovery of REEs in near future, and aimed to provide comprehensive and scientific recommendations for managing ELVs. All 17 REEs (i.e. La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc, and Y) were targeted in estimation of REE contents in ELVs. We focused on interpretation of Nd and Dy in recovery scenarios owing to they are two of the most critical REEs.

2. Materials and methods

Data was obtained from our dismantling surveys including chemical analyses conducted during 2012–2015 (see Chapter 2), official statistics, and academic literatures. StCVs, SmCVs, and HEVs are considered in this chapter. The term "recovery" was defined as the amount of REEs regained from ELVs through certain refining technologies and processes. The time boundary was set between 2015 and 2030 based on the average lifespan (i.e. about 13 years[33]) of vehicles in Japan and schedule of the UN's Sustainable Development Goals on Responsible Consumption and Production (i.e., goal 12).[12]

2.1. Estimation of REEs per vehicle

There are three main factors determining the amount of REEs in vehicles: the size of vehicle, the year of production, and the installation rate. Installation rate was considered only for electric power steering and electric air compressor motors. In addition, the contents of REEs in a vehicle can vary according to its production year owing to reasons such as component upgrades. Based on this understanding, it was estimated that CPVs and HEVs underwent three cycles of component upgrades, and thus, fell under the following three categories: 1st period (before 2003), 2nd period (between 2003 and 2008), and 3rd period (after 2008).

Table 4. List of targeted parts and components in chapter 3

Type	StCV			SmCV			HEV		
Time period[a]:	1st	2nd	3rd	1st	2nd	3rd	1st	2nd	3rd
Common vehicle component									
PWBs of: engine, steering, brake, air big, audio/car navigation, switch in glove box, and harness	●	●	●	●	●	●	●	●	●
PWBs of: speed meter, mirror switch, and door locking receiver	–	●	●	●	●	●	–	●	●
PWBs of electronic toll collection	–	●	●	–	●	●	–	●	●
Electric air compressor motor magnets[b]	–	–	–	–	–	–	–	●	●
Electric power steering motor magnets[c]	–	●	●	–	●	●	–	●	●
Liquid-crystal display monitor	–	●	●	–	●	●	–	●	●
Catalyst	●	●	●	●	●	●	●	●	●
HEV specific component									
PWBs of: Hybrid computer, invertor, and NiMH battery	–	–	–	–	–	–	●	●	●
Hybrid transmission motor magnets	–	–	–	–	–	–	●	●	●
NiMH Battery cell	–	–	–	–	–	–	●	●	●

[a] 1st period: before 2003; 2nd period: between 2003 and 2008; 3rd period: after 2008.

[b] Electric air compressor motor magnets were mostly used in HEVs, owing to high associated electricity cost; therefore, CVs were assumed not to include these components.

[c] Electric power-steering motor magnets were assumed to be installed, in part based on the year of first registration.[43]

●: installed

The various components containing REEs in vehicles were identified by chemical analysis as listed in Table 4. They were classified into two large groups: common vehicle components and HEV specific components. The former group was defined as the components commonly equipped in both CVs and HEVs including motor magnets, PWBs, and catalysts. While the latter included the components equipped only in HEVs, for instance, NiMH battery cells and hybrid transmission motor magnets. LIB may replace NiMH batteries in the future, but it is difficult to predict the change of battery type so far. Therefore, only NiMH batteries fitted to HEVs were considered in this chapter.

Electric air compressor motors and electric power steering motors were assumed to be installed in the vehicles from 2nd period onward, thus the installation rate of them was considered. The term "installation rate" in this study indicates the proportion of the first registered vehicles that are installed with the given components. Based on the statistics, the electric power steering motor was used in the vehicles at a gradual rate. It started from 28% in 2003, and rose to 74% in 2010.[43] Based on an approximation logarithm, the rate was estimated to increase from 74% in 2011 to 91% by 2030, as shown in Figure 10. The historical data related to the installation rate of electric air compressor motor could not be found in the official statistics. However, electric air compressor motors were mostly used in HEVs owing to the high electricity cost associated with them; therefore, CVs were assumed not to equip them.

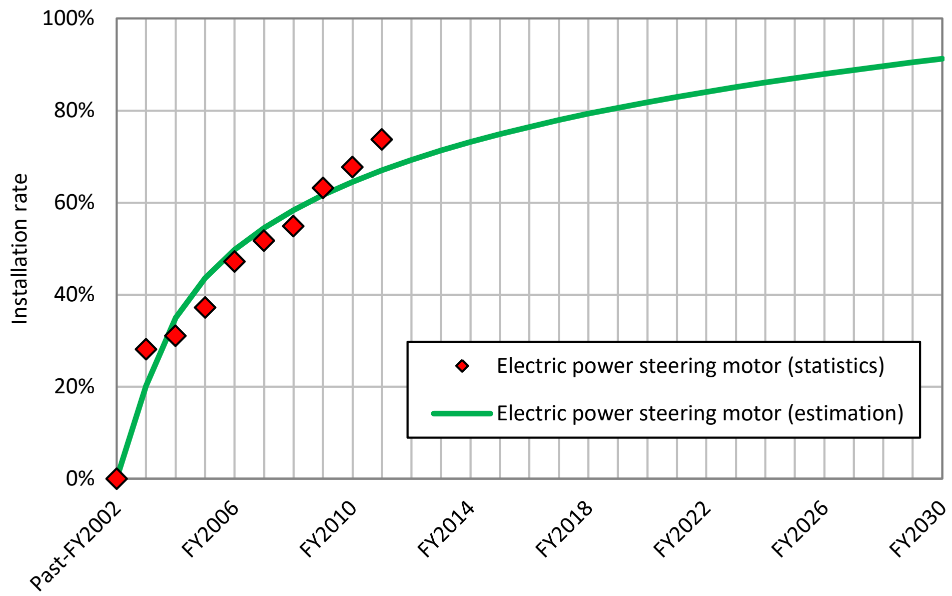

Figure 10. Installation rate of electric power steering motor. FY signifies fiscal year in statistics.

The contents of REEs in various components were calculated by multiplying the concentration of REEs in a specific component by the weight of that component. The contents of REEs per vehicle derived in this chapter are shown in Table 5.

Table 5. REE contents per ELV

Selected components and elements	SmCV			StCV			HEV		
	1st	2nd	3rdc	1st	2nd	3rdc	1st	2nd	3rdc
	(g/vehicle)								
				Component					
Electric power steering motor magnet[a]	–	64.2	64.2	–	94.7	94.7	–	78.8	78.8
Electric air compressor motor magnet[b]	–	–	–	–	–	–	–	18.5	13.3
PWBs	0.7	1.8	1.8	3.5	3.6	3.5	0.3	0.9	0.9
LCD monitor	–	0.001	0.001	–	0.002	0.002	–	0.001	0.001
Catalyst	9.2	9.2	9.2	13.6	13.6	13.6	11.3	11.3	11.3
Hybrid transmission motor magnet	–	–	–	–	–	–	486.3	456.9	335.2
NiHM Battery cells	–	–	–	–	–	–	2702.8	2445.4	1801.9
All components	9.9	75.2	75.2	17.1	111.9	111.9	3200.7	3011.8	2241.4
				Element					
La	1.3	1.4	1.4	2.3	2.3	2.3	870.4	787.8	580.9
Ce	7.9	7.9	7.9	11.7	11.7	11.7	1226.2	1110.3	820.7
Pr	0.04	45.6	45.6	0.2	67.4	67.4	188.6	228.8	135.2
Nd	0.5	19.2	19.2	2.6	28.9	28.9	649.2	660.9	554.6
Dy	0.00	0.9	0.9	0.01	1.3	1.3	148.5	117.5	71.4
Other REEs	0.1	0.2	0.2	0.3	0.3	0.3	117.8	106.6	78.6
Total REEs	9.9	75.2	75.2	17.1	111.9	111.9	3200.7	3011.8	2241.4

[a] The installation rate of electric power steering motors was considered.

[b] Air compressor motors were assumed to be installed only in HEVs from the 2nd period.

1st period: pre-2003; 2nd period: 2003–2008; 3rd period: post-2008.

2.2. Forecast of ELV generations by 2030

This step is to project the number of ELVs and to estimate the amount of end-of-life common vehicle components. Firstly, the future size shares for the end-of-life SmCVs and StCVs were determined in 1989–2010, as shown in Figure 11. From 1989 onwards, the share of StCPVs increased until 2004, reaching its peak (33%); thereafter, it constantly decreased to 2010 (29%).[67] Owing to the increasing concerns around environmental and energy conservation, we assumed that this current trend of decrease in StCVs will last till 2030. By using linear gradient between 2004 and 2010, the share of StCVs was estimated to decrease to 23% by 2030.

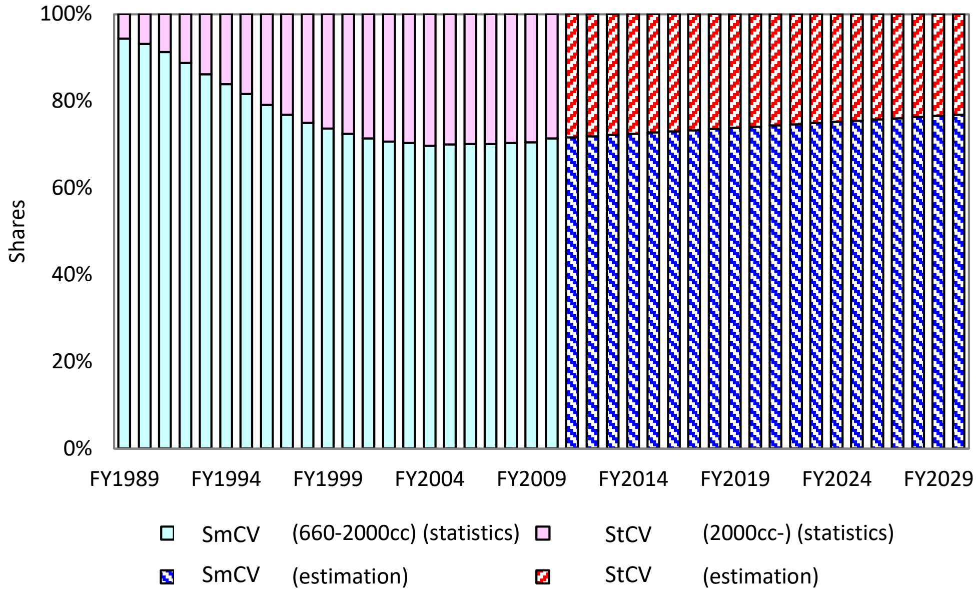

Figure 11. Size shares of end-of-life small size and standard size CVs. FY signifies fiscal year in statistics.

Secondly, the number of end-of-life CVs was projected by building a population balance model, which is a dynamic estimation model based on mass balance between input, stock and output of a material or product with a defined lifespan. The proportion of CVs among the first-registered vehicle was estimated to decrease to 48–58% (maximum–minimum situations for HEVs) in 2030, from the current 65%, based on government target for first-registered vehicles issued by Ministry of Economy, Trade and Industry, Japan.[68] The terms “maximum situation” and “minimum situation” derive from the proportion target set for HEVs. The lifespan distribution based on the first year of registration of a vehicle was considered following the Weibull distribution. The remaining rate function is shown in Eq. 3. Furthermore, the lifespans of CVs were assumed to be different depending on the year of first-registered years. The shape parameter u and scale parameter η were estimated by the least squares method according to the statistics on the ownership of vehicles in Japan.[43,69] Then the annual outputs of end-of-life CVs from stock during 2010–2030 were estimated by Eq. 4–6.

$$F(y,t) = exp\left\{-\left(\frac{t+0.5}{\eta}\right)^{u}\right\} \quad \text{Eq. 3}$$

Where $F(y,t)$ is the remaining rate of first-registered CVs in year *y*; *y* is the first-registered year (=1989–2030); *t* is the vehicle age; *u* is the shape parameter; η is the scale parameter.

$$N_{use}(y,t) = N_{1st_{reg.}}(y,0) \times F(y,t) \quad \text{Eq. 4}$$

$$N_{EoL\_CPV}(y,t) = N_{use}(y,t) - N_{use}(y,t+1) \quad \text{Eq. 5}$$

$$Total_{N_{EoL\_CPV}(Y)} = \sum_{Y=FY1985}^{Y} N_{EoL\_CPV}\,(y, t = Y - y) \quad \text{Eq. 6}$$

Where, *Y* is the counted year (=2010, 2011,…,2030); $N_{use}(y,t)$ is the annual number of CVs in use that are first-registered in year *y* of vehicle age *t*; $N_{1st_{reg.}}$ is the annual number of CVs that are first-registered in year *y* (=$N_{use}(y,0)$), $N_{EoL\_CPV}(y,t)$ is the annual number of end-of-life CVs that are first-registered in year *y* of vehicle age *t*; $Total_{N_{EoL\_CPV}(Y)}$ is the annual total number of end-of-life CVs in *Y*.

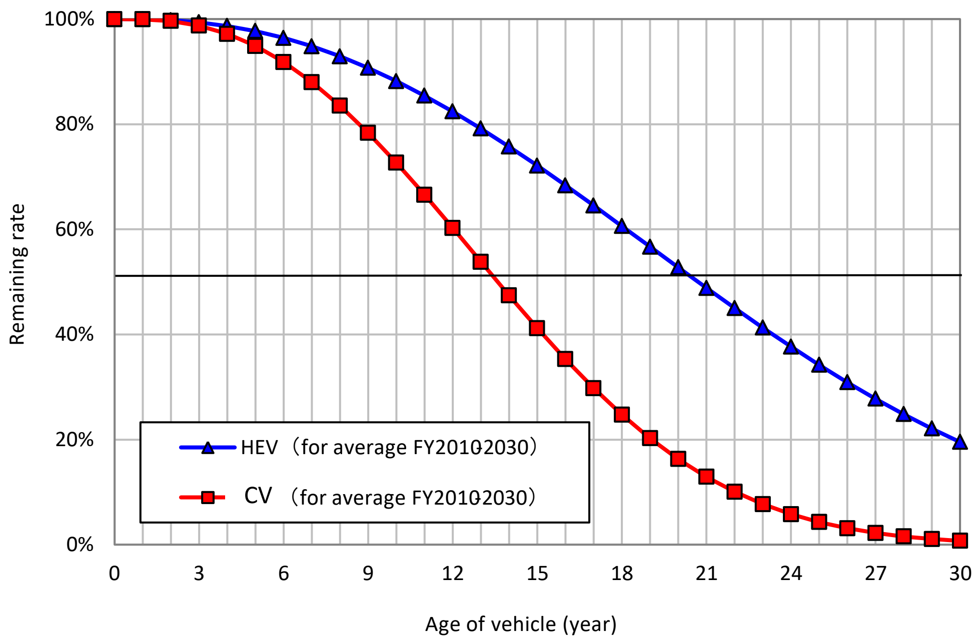

Figure 12. Estimated remaining curves of CVs and HEVs by 2030

Thirdly, by using the same method as for CVs, the number of end-of-life HEVs was projected. The estimated remaining curves of CVs and HEVs are shown in Figure 12, and the estimated lifespans (when the remaining rate falls to 50%) are 13.1 and 20.2 years for CVs and HEVs, respectively. Our estimation regarding CVs was basically in accord with the statistical data by Automobile Inspection & Registration Information Association, which reported that the average lifespan of Japanese vehicles is 13.0 years.[33] Additionally, Oguchi and Fuse[70] estimated the average lifespan of Japanese vehicles is 13.0 years. Overall, our findings showed that HEVs have a considerably longer lifespan than CVs.[24] This phenomenon might due to the HEV owners might have higher environmental awareness, accordingly they preserve and use their HEV longer. On the other hand, HEV entered the market around 2000, there is still lack of statistics of HEV between age 10 and 20 years. This could lead to some uncertainty with respect to the remaining rates of HEV between age 10 and 20 years.

Furthermore, by multiplying the content of REEs in common vehicle components per CV and HEV by the number of end-of-life CVs and HEVs, the annual amount of REEs in end-of-life common vehicle components during 2010–2030 was estimated. By adding the amount of REEs in end-of-life HEV specific components (result of from our previous study[24]) the annual amount of REEs contained in ELVs was estimated.

2.3. Review of recovery technologies

It was reported that the main barriers to recovering REEs are: lack of incentives, inefficient collection, and technological difficulties.[3] Although many laboratory-scale processes have been developed, few have been scaled-up to industrial capacity. A review of REE recovery technologies was conducted to support setting recovery scenarios, as summarized in Table 6.

In the case of NiMH battery recovery, the battery cells can be handled via a pyrometallurgical process using waste metallurgical slag (SiO_2 and CaO), where nickel and cobalt are recovered in priority, and REEs are captured within the slag as oxides. It was reported that the slag contains about 71% of REE content,[71] and that more than 99% of REEs remaining in the slag can be separated at high temperatures (1510–1620°C).[71] Subsequently, the remaining REEs oxides can be directly electrolyzed to high-purity REEs (metal phase) through fluorides melts.[72] The combined net recovery efficiency reaches 70%. On the other hand, NiMH battery cells can also be treated via hydrometallurgical processes. Selective separation of REEs by leaching and precipitation is developed, and about 80–99% of REEs can be gained in solution depending on their levels of solubility.[54,56,57] Overall, REE recovery rates are within the range 70–99%. A pyrometallurgical process is available for NdFeB magnets. In this process, 65% of the REEs in NdFeB magnets are dissolved in molten magnesium (800°C), then separated by vacuum distillation of the magnesium.[73] Similarly to NiMH batteries, REEs in NdFeB magnets can be recovered by hydrometallurgical process in which roasted magnets are dissolved in sulfuric and hydrochloric acid, thereby leaching 60–99% of REEs.[74,75] In this way, REE recovery rates are within the range 60–99%.

Table 6. Review of REEs recovery technologies

Source	Object	Technology type	Efficiency
		Lab-scale	
Tang et al.[55]	NiMH batteries	Pyrometallurgical	REEs collection ratio: 71.4% (oxide phase)
Tang et al.[71]	NiMH batteries	Pyrometallurgical	Separation ratio: 99% (oxide phase)
Morrice et al.[72]	NiMH batteries	Pyrometallurgical	Unknown
Pietrelli at al.[52]	NiMH batteries	Hydrometallurgical	Recovery ratio: about 80%
Li et al.[54]	NiMH batteries	Hydrometallurgical	Over all recovery ratio: 98%
Innocenzi and Vegliò[57]	NiMH batteries	Hydrometallurgical	Dissolution ratio: 99%
Okamoto[73]	NdFeB magnets	Pyrometallurgical	Dissolution ratio: up to 65%
Voßenkaul et al.[75]	NdFeB magnets scrap	Hydrometallurgical	REEs yield: up to 60% (86% purity)
Koyama et al.[74]	NdFeB magnets	Hydrometallurgical	Dissolution ratio: 80%
		Industry-scale	
Toyota[64]	NiMH batteries NdFeB magnets	Unknown	Unknown
Honda Motor[63]	NiMH batteries	Unknown	Recovery ratio: over 80%
Mitsubishi[76]	NdFeB magnets	Unknown	Unknown
Hitachi[62]	NdFeB magnets	A four-step preprocessing	Unknown
Rhodia/Solvay[77]	NiMH batteries	Pyrometallurgical	Recovery ratio: 80%
Umicore UBR plant[77]	NiMH batteries	Unknown	Unknown

For automotive industry, since 1998 Toyota has recovered NiMH batteries from approximately 30 thousands vehicles in Japan, but these batteries could be only recycled as raw materials for stainless steel. Until recently, in 2012 Toyota became the first automaker to develop technology with Sumitomo Metal Mining which allows reuse of

NiMH batteries. Also in 2012, Toyota created the world's first system for recycling Nd and Dy from magnets of HEV motors.[64] In 2013, Honda Motor started to work with TDK Corporation and Japan Metals & Chemicals recycling REEs from used NiMH batteries and reusing them in new batteries.[63] In addition, Mitsubishi Electric is now recycling magnets of air conditioners, the company has developed an automatic dismantling equipment allowing quick dismantling, sorting, and collection (30 seconds per unit). As Mitsubishi also having automobile division, they might apply the process to recycling ELVs.[76]

Addition to the traditional major participants of recovering REEs such as Japan and Western European countries, China is joining this "club" now promoted by its environmental plans and regulations. Ganzhou Recycle Hi-Tech Co., Ltd. (Ganzhou, China) is operating a plant which can treatment 8000 t magnet per year, and recovery 2000 t rare earth oxides per year.[78] GEM Co., Ltd. announced to build a network of recycling secondary battery from ELVs on the 8th Conference of Stockholm Convention in 2018. The network will comprise five facilities: Tianjin ELVs recycling plant, Wuhan ELVs recycling plant, Henan ELVs recycling plant, Wuxi materials base, and Shenzhen vehicle batteries recycling center.[79]

2.4. Scenario analysis

Accordingly, we developed four scenarios for the recovery of REEs, as shown in Figure 13. Our target components were NiMH battery and motors containing NdFeB magnets, as they are major carriers for REEs. In this section, collection and recovery rates of existing recovery technologies are considered. However, rather than selecting a representative technology (e.g. pyrometallurgical technology or hydrometallurgical technology) for each scenario, we considered the range of recovery rates between differing technologies for each scenario. In order to avoid uncertainty regarding future technological advances, we assumed the recovery rates to be constant by 2030. Finally, the amounts of REEs that could be recovered from ELVs during 2010–2030 were estimated.

Scenario 1 (no recovery) is the baseline scenario. We assumed that the current situation, in which REE recovery relies solely on spontaneous processes from end-of-life products such as hard disk drives and air conditioner compressors,[3] will not change until 2030. The commercial recovery of REEs from ELVs remains limited and unknown. Therefore, scenario 1 assumed that no REEs would be recovered from ELVs by 2030.

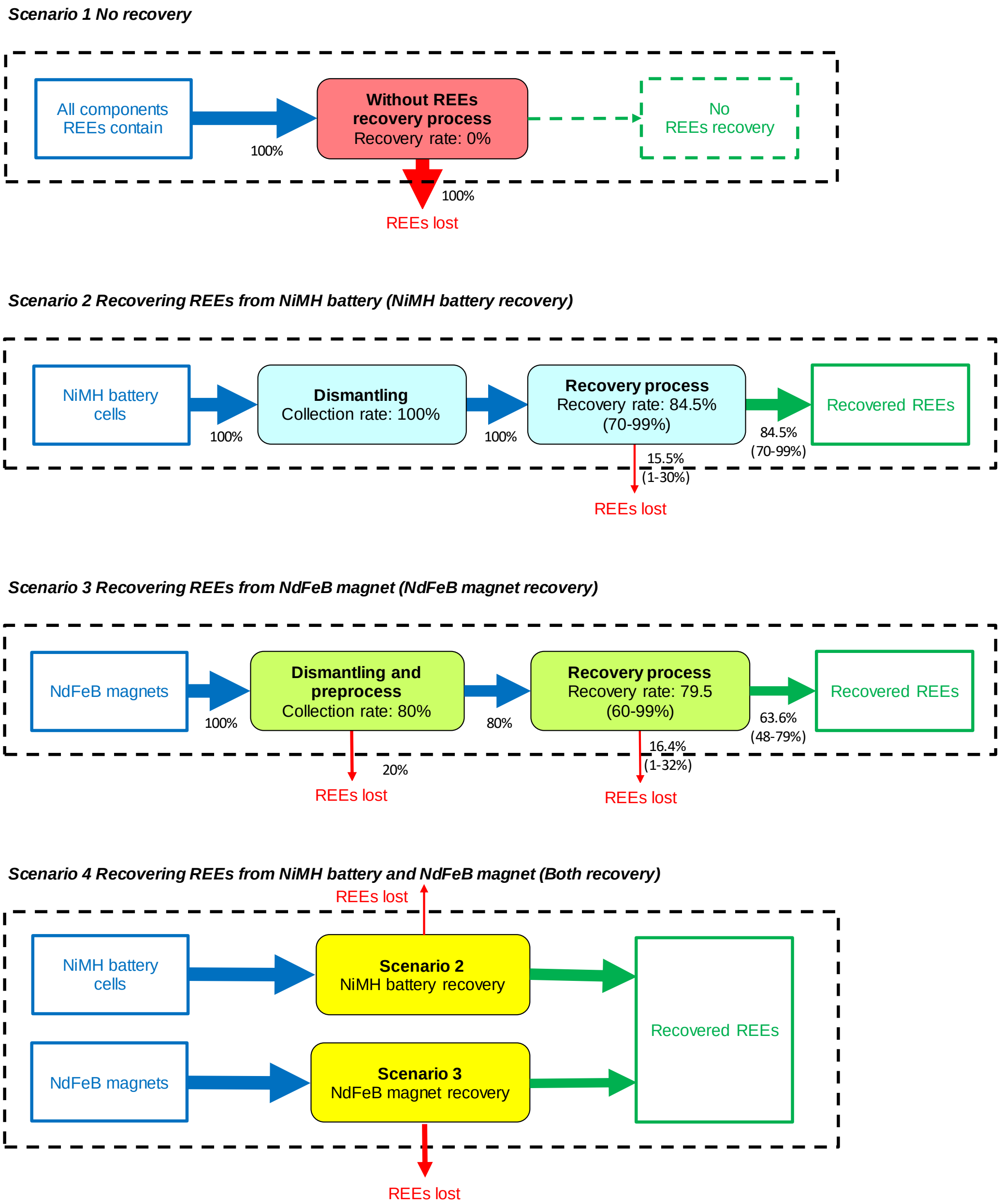

Figure 13. REEs recovery scenarios. Figures in bracket show the range of recovery efficiency.

Scenario 2 (NiMH battery recovery) includes recovery of NiMH battery cells. In Japan, the Law for the Recycling of End-of-Life Vehicles requires the recycling of specific components and materials (e.g., airbags and refrigerants).[2] According to this law, since 2012, secondary batteries were required to be separately collected during the dismantling process and recycled. This can facilitate recovery of the cells from NiMH batteries. In this scenario, NiMH battery cells are first collected at the dismantling plants. Because the battery is a comparatively independent and valuable component of HEVs, its collection rate was assumed to be 100%. Subsequently, the NiMH battery cells are subject to recovery processes. The various technologies for handling NiMH batteries offer recovery efficiencies within the range 70–99%, from which the median value of 84.5% was used as default value in the scenario. Scenario 3 (NdFeB magnet recovery) points at the recovery of motor NdFeB magnets. Compared with NiMH battery cells, motor NdFeB magnets are difficult to separate from vehicles, because they are usually glued in-place and have strong magnetic fields. Hence, an additional preprocessing step is required. The motors are first preprocessed and then the NdFeB magnets are recovered. In this scenario, a four-step preprocessing method is applied. Motors are first removed from vehicles and then passed through cutting machines, after which the NdFeB magnets are manually exposed. Secondly, rotors containing NdFeB magnets are disassembled. Thirdly, the NdFeB magnets are demagnetized for safe sorting. Finally, using NdFeB magnet removers, the rotors are subjected to a specific form of vibration at which only the NdFeB magnets re separated and collected.[62] The separation rate was assumed to be 80% (optimistic). The separated NdFeB magnets are then subject to recovery processes. The available technologies achieve total REE recovery efficiencies of 60–99%. Similarly to scenario 2, the default recovery rate was set as the median, which is 79.5%. Scenario 4 integrates scenario 2 and 3, and it was developed to test the gradient and maximum value (both NiMH batteries and NdFeB magnets) of REE recyclability from ELVs.

2.5. Sensitivity analysis

A sensitivity analysis was conducted in order to identify the crucial parameters of our model. In the section of forecast of the generation of ELVs, our estimation mainly relied on government target. Therefore, parameter (1): number of first-registered vehicles and (2): share rate between CVs and HEVs in first-registered vehicles were analyzed. In Addition, our model was built upon a population balance model. Thus, the set of parameter (3): shape parameters and 4: scale parameters were essential. Parameter (5): Content of REEs in common vehicle components may also lead to deviation, because if contents of REEs per vehicle were assumed to be constant after 2009, the effect of efforts to reduce REEs content according to national strategies such as the Strategy for Ensuring Stable Supplies of Rare Metals will be missed. Parameter (6): installation rate of electric power steering motor magnets and (7): number of NiMH battery cells were also concerned.

In total, seven parameters were analyzed to understand which parameter inaccuracies can significantly affect our estimation. By increasing one parameter by 10%, the change in results and the default results were compared. This sensitivity analysis involves the sensitivity of not recovered amount, but contents in ELVs. Then sensitivity was defined as according to Eq. 7.

$$S_r(i) = REEs_{r'}(i)/REEs_r(i) \quad \text{Eq. 7}$$

Where *i* is the REE (Nd or Dy), $S_r(i)$ is the sensitivity of element *i* when parameter *r* is increased by 10% (=*r*′), $REEs_{r'}(i)$ and $REEs_r(i)$ are the results, and amounts of element *i* in ELVs in year 2030 when parameter *r* and *r*′ are applied.

3. Results and discussion

3.1. ELV generations by 2030

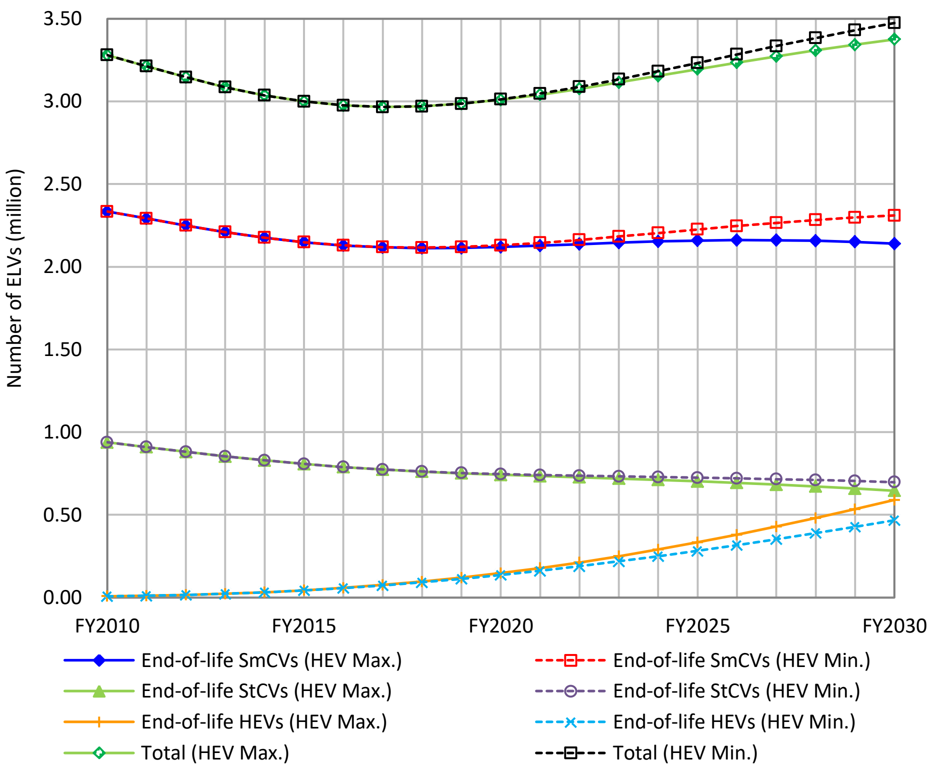

Figure 14. End-of-life SmCV, StCV, and HEV generations. HEV Max.: the maximum situation of HEVs, HEV Min.: the minimum situation of HEVs, FY: fiscal year.

Because we used the government target for our estimation including minimum and maximum situations, our estimations also had minimum and maximum situations as shown in Figure 14. The solid curves reflect the situation where the number of HEVs increases the most in 2030. The dotted curves represent a contrasting situation where the number of HEVs follows the minimum increase route.

The result shows that the number of end-of-life SmCVs during 2010–2030 follows a down and up route, and it will decrease to 2.1 million by 2030. The number of end-of-life StCVs is constantly in a decreasing route, and it will decrease from 0.9 million to 0.6 million during 2010–2030. Regarding end-of-life HEVs, its number will continuously rise till 2030, from 10 thousand to a number to that of as StCVs, which is 0.5 million. In general, the number of ELVs during 2010–2030 appears to follow a slight U-shape concave. Approximately 3.3 million ELVs will be generated in 2030, which is nearly equal to the number of ELVs generated in 2010. End-of-life HEVs accounted for only 0.3% of total number of ELVs generated in 2010, but it will increase to 18% in 2030. Accordingly, the number of end-of-life HEV specific components will increase rapidly in these 20 years.

3.2. REEs in ELVs between 2010 and 2030

The result was estimated by adding the amount of REEs in common vehicle components and in HEV specific components[24] as shown in Figure 15. Considering the actual case in Japan, which is the one of the largest HEV producing countries in the world, we mainly interpreted the HEV maximum situation, from this section.

The amount of REEs in ELVs will increase dramatically during 2010–2030, from 130 t in 2010 to 3400 t in 2030. NiMH battery cells had the biggest share, and accounts for 87% of the total amount in 2030. Meanwhile, 12% of the total is occupied by motor magnets, including 6% by hybrid transmission motor magnet, approximately 6% by electric power steering motor magnet, and only 0.3% by electric air compressor motor magnet. Additionally, 1% is occupied by other components in 2030. Regarding the type of element, Ce and La had the largest shares because they are the dominant elements in NiMH battery cells, and these battery cells are the main components of REEs in HEV. In 2030, Ce and La occupy 41% and 29%, respectively, of the total amount of REEs in ELVs. There are 670 t and 49 t of Nd and Dy. More details on the amount of each REE in vehicle components have been included in the electronic supplemental materials (Appendix 9).

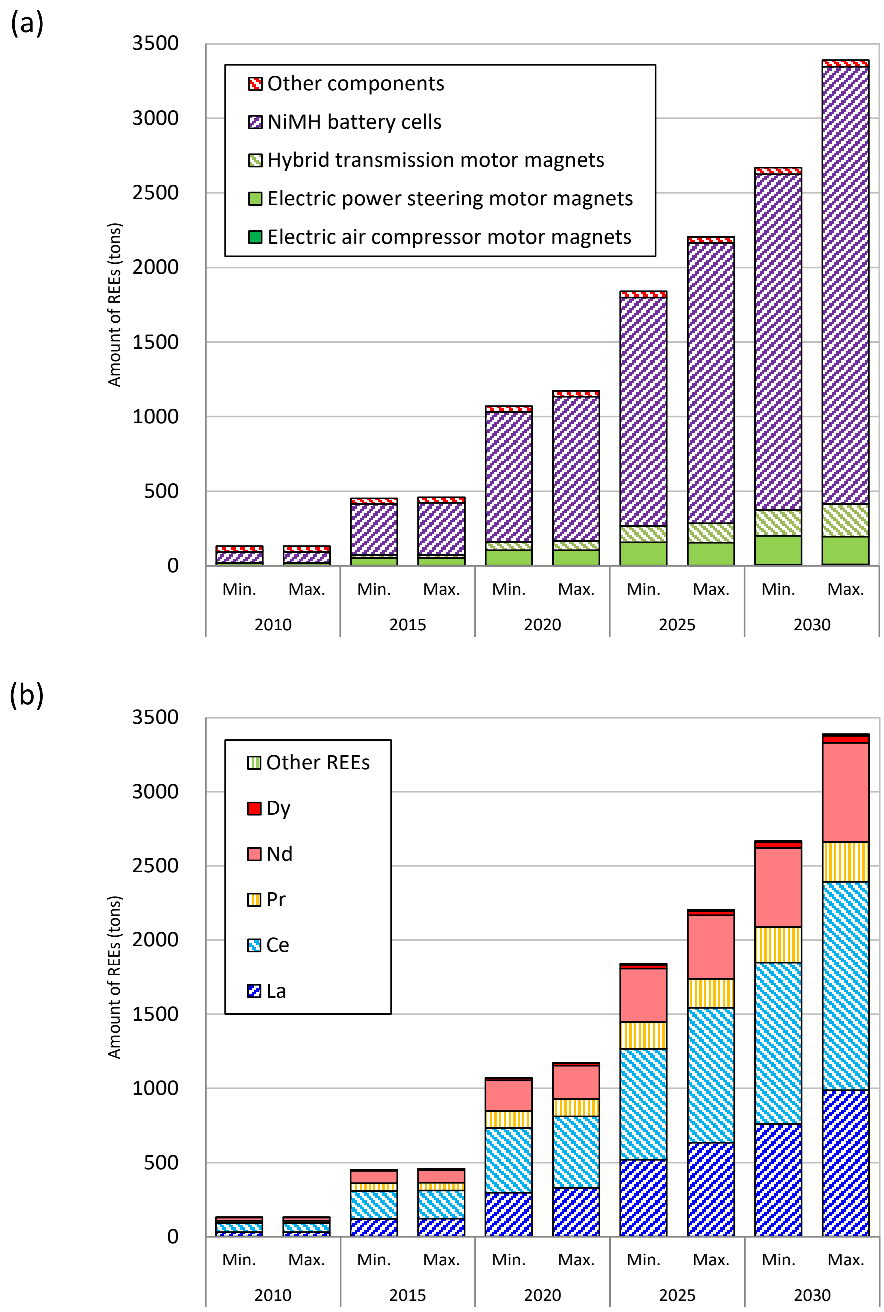

Figure 15. REE contents of ELVs by (a) components and (b) elements

3.3. Recovered REEs by scenarios

The outcomes of the recovery scenarios in 2030 are shown in Figure 16. Scenario 1 is the baseline, where no activity of REEs was carried out. Accordingly, the collection and recovery rates are 0%. In scenario 2 focusing NiMH battery recovery, the amounts of Ce, La, Pr, and Nd recovered from NiMH battery cells were 1200 (±200), 830 (±140), 110 (±20), and 370 (±60) t in 2030, respectively. The absolute amount of other recovered

REEs is low, at only 9 (±1) t scenario 3 focuses on recovery of NdFeB magnets. Approximately 85 (±20) t of Pr, 150 (±40) t of Nd, and 31 (±7) t of Dy can be recovered in 2030. According to scenario 4 (Both recovery), 2700 (±500) t of REEs could be recovered from NiMH battery cells and NdFeB magnets in 2030. The net recovery efficiency in 2030 is about 81% (±14%).

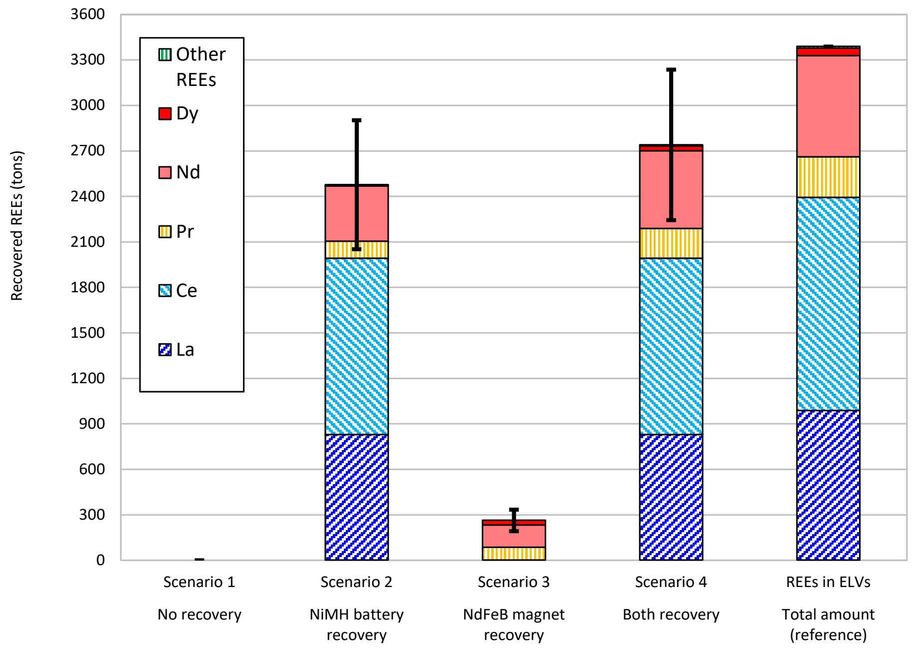

Figure 16. REEs recovered from ELVs under each scenario in 2030. Ranges reflect the recovered REEs by different recovery efficiencies.

Regarding the recovery of REEs, Nd and Dy are essential from the viewpoint of criticality and practice.[3,48] Furthermore, their recovery is potentially profitable: Dent reported average Ce and La prices of 9 USD/kg, whereas those of Nd and Dy once were more than 450 and 3400 USD/kg, respectively as of 2011.[80,81] Although the prices were dropped recently, they are still very valuable. A comparison between components in scenario 4 in 2030 is shown in Figure 17. For both of recovering Nd and Dy, hybrid transmission motors magnets contribute the most which account 21% and 91% of the total amount. Referring other motor magnets, electric power steering motor magnets and electric air compressor motor magnets account for 7% (±0.4%) of the total Nd recovery, and the Nd recovered from the former is about 7 times larger than the latter. On the other hand, these two motor magnets share 8% (<±0.1%) of total Dy recovery, and the Dy recovered from electric power steering motor magnets is almost 2 times of it recovered from electric air compressor motor magnets. Moreover, considering that the installation rate of electric power steering (and hence motor magnets) in vehicles increased from 44% in 2005 to 67% in 2010, the amount of Nd and Dy recovered from that component would increase over time. Therefore, collection of these magnets from both end-of-life CVs and HEVs is likely to contribute to the recovery of Nd and Dy. The recovery priority in motor magnets should be sequentially given to hybrid motor magnets, electric power steering motor magnets and electric air compressor motor magnets in the perspective of quantity

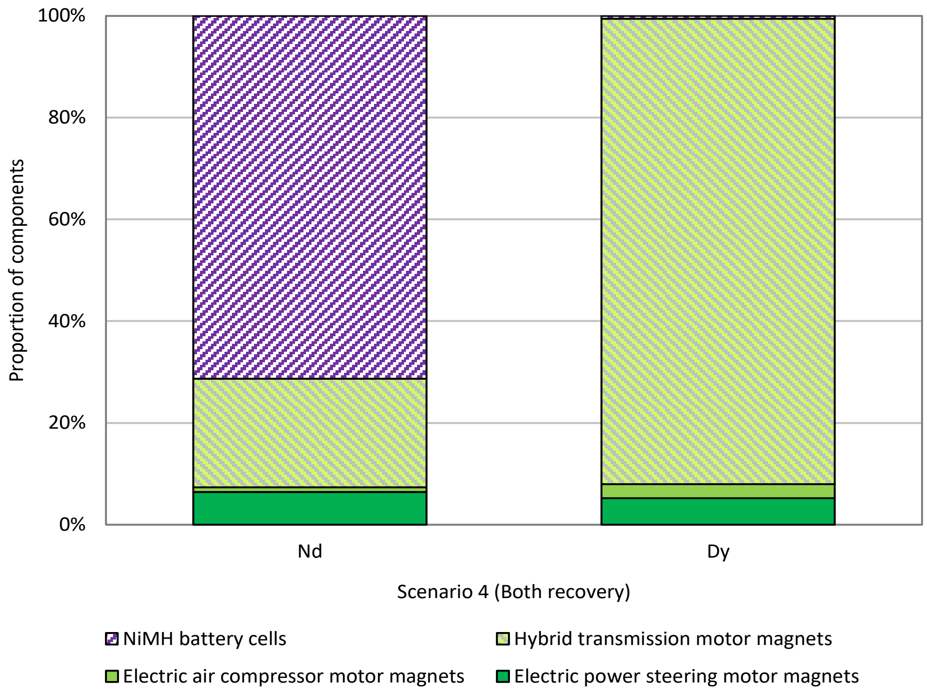

Figure 17. Comparison of components with respect to Nd and Dy recovered under scenario 4 in 2030

Using the number of first-registered vehicles and content of REEs per vehicle, the demands for Nd and Dy for vehicles in Japan could be also estimated through our model. Improvements in NiMH battery technology result in less use of Nd in batteries, but our dismantling survey indicated that there was no substantial reduction in the use of Nd for NdFeB magnets. We estimated that 470 t of Nd will be required to produce NiMH battery cells, and 590 t of Nd and 130 t of Dy will be required for NdFeB magnets in motors in 2030. The recovered Nd from ELVs can satisfy 49% (±9%) of the demands for production of NdFeB magnets and NiMH battery cells. In addition, 23% (±6%) of

Dy demand for automobiles (motor magnets) can be satisfied by the recovery from ELVs. Further details on demand and recovery in 2010 and 2020 have been included in the electronic supplemental materials (Appendix 10).

Secondary Nd and Dy sources from ELVs in Japan appear to be enough to meet a considerable fraction of the domestic demand and to relieve the dependence on import of raw materials, which is similar to the situation in the EU.[7]

3.4. Sensitivities and uncertainty

Table 7. Results of sensitivity analysis in chapter 3

No.	Parameter	$S_r(Nd)$ Sensitivity to Nd content in ELVs	$S_r(Dy)$ Sensitivity to Dy content in ELVs
1	Number of first-registered vehicles	1.07	1.05
2	Share rate between CVs and HEVs in first-registered vehicles	1.06	1.04
3	Shape parameters of remaining rate function	-1.08	-1.03
4	Scale parameters of remaining rate function	-1.05	-1.11
5	Content of REEs in common vehicle components	1.05	1.10
6	Installation rate of electric power steering motor magnets	1.02	1.03
7	Amount of NiMH battery cells	1.06	1.00

$S_r(i)$ is the sensitivity of element *i* (= Nd, Dy) when parameter r is increased by 10% compared with the default value.

As shown in Table 7, the most sensitive parameter for the Nd content of ELVs is parameter (3); a 10% increase in this parameter results in approximately -8% of Nd change in ELVs. In addition, parameter (1), (2), and (7) are also sensitive, and can account for 7%, 6%, and 6% of changes, respectively. The most sensitive parameters for Dy content in ELVs are parameter (4) and (5), which account for -11% and 10% changes in Dy content, respectively. Parameter (1) is also sensitive, accounting for 5% change.

In our estimation model where recovery efficiency was set to be constant during 2010–2030 and reuse of components was not concerned. They are remaining uncertainties. Moreover, parameter (5) may cause large uncertainties because the content of REEs per vehicle has a wider range between previous studies as shown in Chapter 1. Especially, Nguyen et al.[31] in USA reported a notable finding that they found only speakers containing NdFeB magnets among 91 components including motor magnets.

Three reasons can be identified as follows:

- The first is that manufacturers differ from each other, and therefore the components differ.
- The second is that the scales of samples also differ from each other.
- The third is that the calculation methods and considered components are different.

4. Conclusion

This study conducted a scenario analysis aimed to clarify the REEs in ELVs in Japan in the near future, and then explored the recovery of REEs through scenarios. The obtained conclusions were shown below:

- The total amounts of REEs in ELVs will increase from 130 to 3400 t during 2010–2030. In total, 2700 (±500) t can be recovered from NiMH battery cells and motor NdFeB magnets in 2030, of which 520 (±100) and 31 (±7) t will be contributed by Nd and Dy, respectively. The net recovery efficiency in 2030 is approximately 81% (±14%).
- Comparing common vehicle components and HEV specific components, the former accounts about 7% (±0.4%) of the total recovery of Nd, and about 8% (<±0.1%) of the total Dy; electric power steering motor is the dominant component in Comparing common vehicle components.
- Dy recovered from ELVs can satisfy 23% (±6%) of the demand for automobile production of Japan in 2030; meanwhile, Nd from ELVs can satisfy 49% (±9%) of the production demands for motor NdFeB magnets and NiMH battery cells.

It is beneficial to recover magnets from electric power steering and electric air compressors units, together with those from hybrid transmission motors. In addition, by fully activating the incentive in our scenarios, net recovery rate has a wide range of 67–94%. However, the technologies required to achieve such high recovery rates are mainly limited to laboratory-scale at present. Thus, further progress is required to improve the viability of REE recovery processes. The recovery potential of 3400 t REEs from ELVs by 2030, estimated in the study, provides support for policies to expand facilities for: i) the dismantling of HEVs, ii) the recovery of REEs from motor NdFeB magnets and

NiMH batteries cells. The use of different vehicles and methodologies results in widely various estimation of the REE contents of vehicle components. Hence, REE recovery schemes should consider the types of automobiles and other factors relevant to local circumstances.

Chapter 4 Recycling potentials of precious metals by selective dismantling

1. Introduction

Precious metals are rare, naturally occurring metallic chemical elements of high economic value. They usually have high lustre and are ductile. Historically, precious metals were important as currency but now, they are mainly regarded as investment and industrial commodities. Au, Ag, Pt, and Pd each have its own ISO 4217 currency code.[82]

Graedel et al.[83] assessed that Ag, Au, and PGMs are critical to varying degrees with regard to supply risk, vulnerability to supply restriction, and environmental implications. For example, Au has large geological reserves and a low companion fraction, so supply risk is low, but its high cradle-to-gate environmental impacts per kilogram of metal (related to extraction and processing from ore deposits) and its high vulnerability to supply restriction (related to its near-universal use in electronics, jewelry, and investments and its lack of available suitable substitutes) render it of special interest. Additionally, Pt has similar energy and environmental challenges, and its deposits are geopolitically highly concentrated.

In automotive industry, PMs are extendedly used in some components such as catalyst and PWBs of various parts.[84–86] So far, it has so far been difficult to efficiently recycle all PMs in ELVs because many REE/PM-containing components are treated by shredding,[2,87] where their PM contents are highly diluted in complex shredder fractions (e.g. Fe and Al scrap and ASR).[17] At the industrial-scale, mature options for recycling scarce metals (e.g. refining and reuse) need to largely rely on pre-shredding separations of REE/PM-containing parts/components.[76,88,89] However, for reasons of simplicity and speed, the current industrialized ELVs recycling system tends to focus primarily on the collection of battery cells and catalysts, and neglects other REE/PM-containing components such as motor magnets and PWBs.[90,91] Hence, there is a risk that many of the PMs in ELVs may not be returned to their material streams for reutilization.

Currently, there is still lack of sufficient quantification of future PMs recycling potentials. This chapter therefore, enlightened by the concept of “easy to dismantle vehicles”[92] (vehicles that are specially designed to facilitate disassembly) initiated by some automobile manufactures for the future, chose selective dismantling as the main improvement means, focused on PM-containing parts that are both common in all vehicles and special in only next-generation vehicles, and provided an estimation of the recycling potentials of PMs from ELVs by 2040 to provide comprehensive and scientific recommendations for ELVs management.

2. Materials and methods

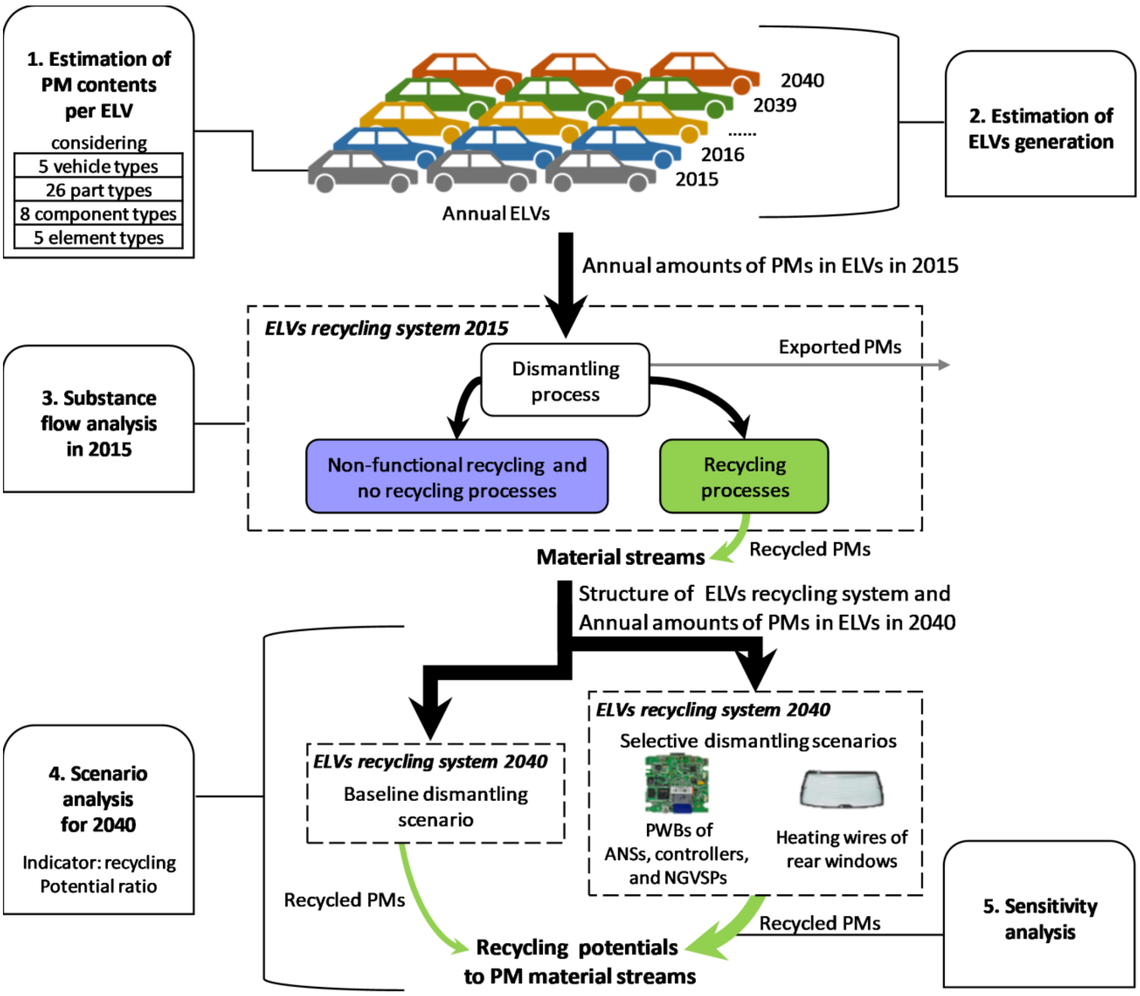

Figure 18. Overview of research procedure in chapter 4. ANS and NGVSP signify audio and navigation system and next-generation vehicle specific part, and, respectively.

In this chapter, the term "recycling potential" is defined as the amount of an embodied element that could be returned to material streams (in the form of refined metal and contained in reused parts) where its properties are utilized again functionally.[6,19,93,94] Details about the potential from ELV parts can be visualized in Appendix 11. Another term "selective dismantling" is defined as the dismantling process selectively focused on certain special items and providing intensive dismantling operations on them. To clarify the recycling potentials of PMs from ELV parts by selective dismantling in Japan, a substance flow analysis (SFA) and a scenario analysis were developed based on estimations of PM contents per ELV and ELV generations. We set 2015 as the reference year and 2040 as the target year to reflect the current and future situations. Moreover, a sensitivity analysis was carried out to identify the uncertainties. Research materials were mainly derived from experimental works including an ELVs dismantling survey (see Chapter 1) and an automotive PM-containing components collection questionnaire, and extraneous information including government statistics and targets, associations and enterprises information (e.g. reports and official websites), and academic articles. Figure 18 illustrates the research procedure of this study.

2.1. Estimation of PMs per vehicle

A layered model that describes the physical relationship between goods and substances was developed specifically to estimate PM contents per ELV.[16,18,95] The layered model comprised four structural layers: vehicle layer, part layer, component layer, and element layer. Because we focused on passenger vehicles, other vehicle types, such as buses and trucks, were not included in this chapter as the same as Chapter 3. Considering vehicles classification under the Road Vehicles Act[32] (Appendix 1) and the recent rise of next-generation vehicles in Japan, five vehicle types were included in the vehicle layer: StCVs, SmCVs, MCVs, HEVs, and EVs. The size difference with respect to HEVs and EVs was not considered, as EVs are mainly represented by a few vehicle models such as Nissan Leaf in Japan. Based on industrial information[76,85,86,89,90] and advice from the dismantler, in total 26 parts and the eight components they contained were categorized into the part layer and component layer, respectively. A detailed list of these parts and components is provided in Table 8. Furthermore, five PMs: Au, Ag, Pt, Pd, and Rh were selected in the element layer as a result of their importance to the automotive industry.[86]

To maintain the original shape of the data from experimental works, no further assumption regarding the elements concentration change by industrial dynamics (e.g. metal sustainability and technology advance) was made in the estimation. In addition, because some parts are optional in vehicles, part installation ratios were taken into consideration. In total, six parts: steering controller, anti-lock braking system (ABS) controller, navigation system, compact disc (CD) changer (subpart) in the audio system, electronic toll collection (ETC) controller, and center console monitor were considered and assigned differing installation ratios over time.[18] Subparts refer to parts assemblies that still consist of some components. The details of the part installation ratios are provided in Table 9.

Table 8. List of targeted parts and components in Chapter 4

Component	Part		Vehicle		
			CV	HEV	EV
PWB	PWB-containing ANSs	Navigation system	○	○	○
		Audio system	●*	●	●
	PWB-containing Controllers	Engine controller	●	●	–
		Steering controller	○	○	○
		ABS controller	○	○	○
		Air-bag controller	●	●	●
		Speedometer	●	●	●
		ETC controller	○	○	○
	PWB-containing NGVSPs	NiMH battery unit	–	●	–
		LIB unit	–	–	●
		Hybrid computer	–	●	–
		Electric motor computer	–	–	●
		Inverter	–	●	●
	Other PWB-containing parts: e.g. shift lock, door (switch), wiring harness, unidentified parts		●	●	●
Battery cell	NGVSPs	NiMH battery			
Catalyst	Catalytic converter		●	●	–
LCD	ANSs	Center console monitor	○	○	○
Ceramic	O_2 sensor		●	●	–
Metal terminal	Rear window		●	●	●
Heating wire			●	●	●
Earth electrode	Igniter		●	●	–

●: installed,

○: partially installed with parts installation ratios

–: no installation

* Sub-part CD changer of audio system is an old technology, and we considered it only be installed in StCVs partially.

Table 9. Parts installation ratios in ELVs in Japan. The ratios for steering controllers and ABS controllers were assumed.

Part	Installation ratio		
	2015	2030	2040
Navigation system	30%	60%*	80%
CD changer in audio system	30%	12%*	0%
Center console monitor	30%	60%*	80%
ETC controller	30%	60%*	80%
Steering controller	30%	66%*	90%
ABS controller	30%	66%*	90%

* Values were calculated linearly using ratios between 2015 and 2040.

2.2. Update of ELV generations by 2040

Chapter 3 estimated the end-of-life StCVs, SmCVs, and HEVs generations in Japan between 2010 and 2030. In this chapter, we additionally included MCVs and EVs (a key vehicle type affecting future PMs recycling potentials owing to the absence of catalytic convertors and the appearance of EVs specific parts) in the estimation, and updated two remaining rates, one for conventional vehicles (i.e. StCVs, SmCVs, and MCVs), another for next-generation vehicles (i.e. HEVs and EVs) using statistics from government and associations.[11,96–98] In Japan, EVs were commercially introduced in the market since 2010 which is about ten years later than HEVs. Accordingly, we extended time boundary by ten years for the target year as 2040 to conduct an effective estimation on EVs. Detailed estimated ELVs generation in Japan during the target years is shown in Appendix 12.

2.3. Substance flow analysis

An SFA was conducted to clarify the performance of present PMs recycling from ELV parts in Japan. After being deregistered, the used vehicles in Japan generally have two fates: some of those that are in relatively good condition can be exported overseas as commodities for their second lifespan, others, referred to as ELVs, are sent to domestic ELVs recycling system.[87] The former was excluded from system boundary. Vehicles that are domestically reused in entirety were not considered because they (without entering the ELVs recycling system) are still vehicles in use in Japan despite change of ownership. This study focused on ELVs; we considered eight processes related to PMs: dismantling, domestic parts reuse market, PMs refining (currently available technologies in industrial-scale mainly include pyrometallurgical and hydrometallurgical technologies),[86,88,99] storage, shredding and separation, other metals market, ASR recycling and energy recovery, and landfill.[2,87]

The company Dowa recycles PMs from various industrial products including computers, cellphones, and automobiles. They have built five facilities throughout the world to recycling PMs; three are located in Japan, other two are in China and Singapore, respectively. Among them, two facilities in Japan have abilities to recycling PMs from automotive catalysts and PWBs, and both of them are using pyrometallurgical processes.[99] Asahi Holding can process wider range of goods, except computers, cellphones, and automobiles, they can also refining PMs from dental and photographic equipment and jewelry. Their business has been expanded to North American already.[89] Tanaka is enable to recycle PMs from not only solids but also liquids. By using hydrometallurgical processes, they manage to produce high purity PMs from waste plating solution and so on.[86] In lab-scale, new technologies: biometallurgical process and electrochemical process developed due to their advantages in terms of recovery efficiency and pollution control. For instance, biotechnological processes involve interactions between microorganisms and metals or metal-bearing minerals,[100] and there are two main fields of biometallurgy for recovery of metals, namely biosorption and bioleaching. The biosorption process is a physico-chemical and metabolism-independent process enabling removal of substances from solution using biological materials; on the other hand, bioleaching is technically feasible by the use of bacteria-assisted reaction to extract precious metals from PWBs.[88]

Note that owing to the fact that illegal ELVs recycling in Japan is rare, following enforcement of the Law for the Recycling of End-of-life Vehicles in 2005,[94] all ELVs were assumed to go through the dismantling process. The storage process was established for temporal conservation of NiMH battery cells, partly because the amount of end-of-life NiMH battery cells is still low at present, and partly because the systematic recycling of the cells might require large investments in facilities initially. However, it should be replaced by other metals market processes in the future to recycle metals such as the rare earth elements inside the NiMH battery cells. In addition, because recycling policy and technology vary by country, and it is difficult to identify the exact final fates and recycling efficiencies of the exported used parts/components overseas, another treatment option for the parts/components, called export market for reuse and refining purposes (hereafter, for short, all references to export market refer to this process), was excluded from the system as an unclear process. More details about each related substance flow and stock in the ELVs recycling system can be found in Appendix 11.

The total amount of PMs in ELVs (i.e. total inflow to the system) was calculated by combining the estimated PM contents per ELV and ELV generations. Moreover, an automotive components collection questionnaire was also completed by the commercial dismantler in 2015 to calculate PM flows and stocks that directly connect to the dismantling process. Two main investigation items: collection ratios and fate ratios were surveyed. According to advice from the dismantler, the questionnaire divided collection ratios into five levels: 0% (i.e. no collection), 1–24%, 25–49%, 50–74%, and 75–100%, and determined the following four fates: export market (outside the system boundary), domestic parts reuse market, PMs refining, and storage for the collected parts/components. In 2015, an automotive PM-containing components collection questionnaire funded by the Environment Research and Technology Development Fund, Ministry of the Environment, Japan, was conducted with the aim of identifying the situation of removal of the PM-containing components during dismantling. Summarized component collection and fate ratios can be found in below Table 10.

Table 10. Components collection ratios and their fates after dismantling. Components can be collected as individual components or contained in their parts.

Component (part)	**Collection ratio**		**Fate ratio**			
	Min	**Max**	**Export market**	**Reuse part market**	**PMs refining**	**Storage**
PWBs (ANSs, controllers, and NGVSPs)	25%	49%	25%	25%	50%	–
PWBs (other parts)	0%	0%	–	–	–	–
Battery cells (NiMH batteries)	75%	100%	–	5%	–	95%
Catalysts (catalytic converters)	75%	100%	–	10%	90%	–
LCDs (center console monitors)	50%	74%	–	95%	5%	–
Ceramic (O_2 sensors)	75%	100%	–	5%	95%	–
Earth electrodes (igniters)	0%	0%	–	–	–	–
Metal terminals (rear windows)	1%	24%	–	–	100%	–
Heating wires (rear windows)	1%	24%	–	–	100%	–

0%: components were considered, but no commercial collection.

Table 11. Calculation methods of PM flows and stocks in SFA.

Flow/stock	Quantification method	Calculation equation
–	PM contents and ELVs generation	*f_deregistered vehicles* = $\sum C_{veh.}(r,p) \times N_{der}(p)$
–	Association statistics	*f_exported used vehicles* = $\sum C_{veh.}(r,p) \times N_{exp.}(p)$
–	Questionnaire	*f_parts/components to export market* = $\sum C_{com.}(r,j) \times N_{E.}(p) \times O(j) \times F_{exp.}(j)$
f1_2		*f1_2* = $\sum C_{com.}(r,j) \times N_{E.}(p) \times O(j) \times F_{reu.}(j)$
f1_3		*f1_3* = $\sum C_{com.}(r,j) \times N_{E.}(p) \times O(j) \times F_{ref.}(j)$
f1_4		*f1_4* = $\sum C_{com.}(r,j) \times N_{E.}(p) \times O(j) \times F_{sto.}(j)$
f3_0	Academic articles	*f3_0* = $\sum f1\_3(r) \times R_{ref.}(r)$
f5_6		*f5_6* = $\sum f1\_5(r) \times S_{shr.}(r)$
f7_6		*f7_6* = $\sum f5\_7(r) \times S_{rec.}(r)$
–	Mass balance	*f_exported used parts/components = f_parts/components to export market*
f0_1		*f0_1 = f_deregistered veh. - f_exported used veh.*
f2_0		*f2_0 = f1_2*
f3_8		*f3_8 = f1_3 - f3_0*
f1_5		*f1_5 = f0_1 - (f1_2 + f1_3 + f1_4 + f_parts/components to export market)*
f5_7		*f5_7 = f1_5 - f5_6*
f7_8		*f7_8 = f5_7 - f7_6*
f6_0		*f6_0 = f5_6 + f7_6*
s4	Mass balance	*s4 = f1_4*
s8		*s8 = f7_8 + f3_8*

In the equations, r is the type of PM; p is the type of vehicle; j is the type of component; $C_{veh.}$ and $C_{com.}$ are the PM content in a vehicle and a component respectively; N_{der}, N_{exp}, and $N_{E.}$ are the number of deregistered vehicles, exported used vehicles and ELVs; O is the collection ratio of component; $F_{exp.}$, $F_{reu.}$, $F_{ref.}$, and $F_{sto.}$ are the fates; $R_{ref.}$ is the refining ratio in PM refining; $S_{shr.}$ and $S_{rec.}$ are the separate ratios of PMs in shredding and separation, ASR recycling and energy recovery to other metals market. For example, $C_{veh.}$(r.p) is the content of PM type r in vehicle type p, N_E(p) is the number of ELVs type p, O(j) is the collection ratio of component type j, $F_{exp.}$(j) is the fate of component type j after collection, $R_{ref.}$(r) is the refining ratio of PM type r in PM recycling, $S_{shr.}$(r) is the separate ratio of PM type r in shredding and separation to other metals market, f1_4(r) is the type of PM r contained in the flow f1_4. $R_{ref.}$(r), $S_{shr.}$(r), and $S_{rec.}$(r) were estimated from previous studies, of which $R_{ref.}$(r) is 70% for Rh, 95% for Ag, Au, Pd, and Pt; $S_{shr.}$(r) is 87% for Ag, 99% for Au, 95% for PGMs; $S_{rec.}$(r) is assumed as 92% for each PM.

The commercial ELV dismantler (with an 11000 unit per year capacity) with whom we collaborated in the ELVs dismantling survey was queried. The targeted 26 parts and eight components in the layered ELV model were included in this questionnaire. According to advice from the dismantler, we set a five collection levels questionnaire: 0% (i.e. no collection), 1–24%, 25–49%, 50–74%, and 75–100%. In the meantime, four fates after collection: export market for parts reuse and PMs refining purposes (hereafter, for short all references to export market mean this process), domestic parts reuse market, PMs refining, and storage for the parts/components were considered. All the questions in the questionnaire were answered by experts in the dismantler, based on annual situation. Other intermediate and final PM flows and stocks were calculated based on data from associations and enterprises[98] and academic articles.[17,19,88,101] Detailed calculation methods for each PM flow and stock considered in the SFA are provided in Table 11.

2.4. Scenario analysis

According to the results of our questionnaire, there are still large improvements that could be made to the dismantling process, in particular with respect to various PWBs and heating wires in the rear window. Therefore, two selective dismantling scenarios aimed at PWBs and heating wires were developed for recycling PMs from ELVs in 2040. Recycling potential ratio (RPR) was set as an indicator to show the performance of scenarios by using Eq. 8.

$$RPR(r,s) = \frac{A_{reu.}(r,s) + A_{ref.}(r,s)}{A_{inp.}(r,s)} \quad \text{Eq. 8}$$

where *r* is the PM element (e.g. Ag); s is the scenarios (e.g. S3); *RPR(r, s)* is the RPR of element *r* in the scenarios, $A_{reu.}(r,s)$ and $A_{ref.}(r,s)$ are the amount of domestic reused (contained in parts) and refined element *r* in the scenarios, and $A_{inp.}(r,s)$ is the amount of element *r* inputted into the ELVs recycling system in the scenarios.

An overview of the scenarios is illustrated in Figure 19. This scenario analysis focused on improving the collection ratios of the PM-containing components in the dismantling process.

Scenario 1 assumed that the dismantling process in 2015 will not change until 2040, and serves as a baseline scenario for comparison. Scenario 2 selectively focuses on PWBs. In total, 20 PWB-containing parts were considered, and they can be grouped into four groups: audio and navigation systems (ANSs), controllers, next-generation vehicle specific parts (NGVSPs), and the other parts, based on their functions and locations in vehicles. Accordingly, we further divided scenario 2 into three sub-scenarios: scenario 2a assumed that the collection ratio of PWBs in ANSs ideally improves to 75–100% (the highest level in our questionnaire), through dismantling operation reforms by 2040; scenario 2b focused on PWBs in controllers, and made the same assumption on the collection ratio (to 75–100%) by 2040; scenario 2c targeted PWBs in NGVSPs, it also assumed that their collection ratios will improve to 75–100%. Scenario 3 focused on heating wires; it assumed that the assembling and dismantling technologies will upgrade to allow the collection ratio of heating wires to increase to 75–100% by 2040.

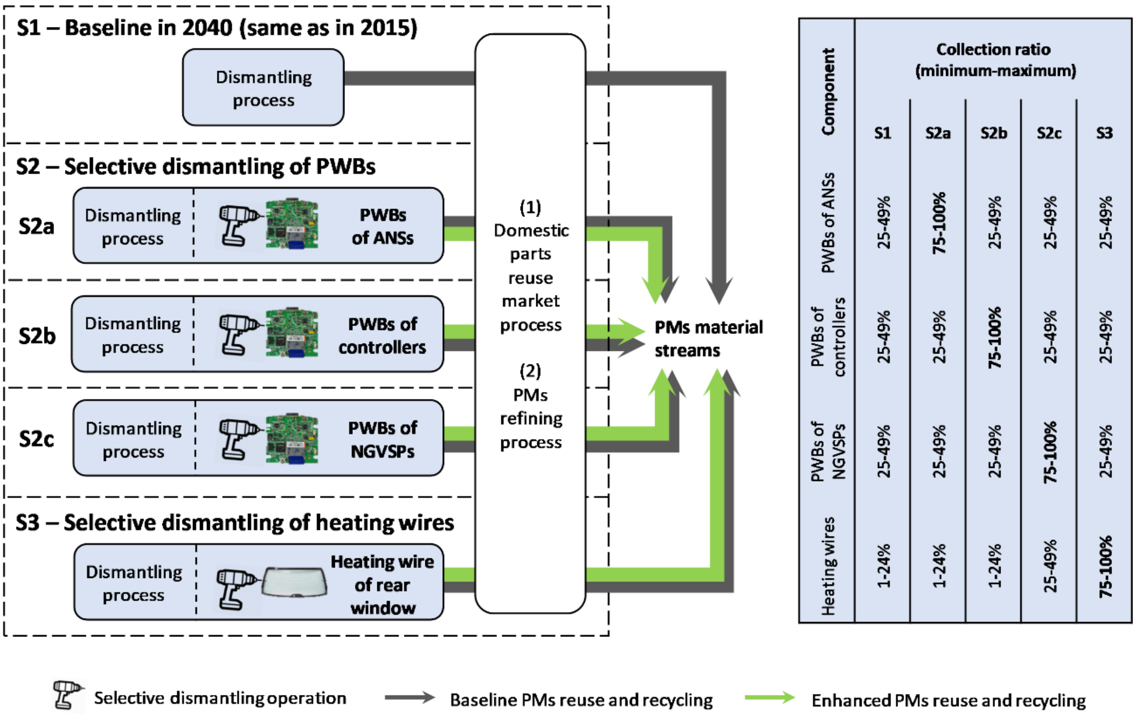

Figure 19. Overview of selective dismantling scenarios in 2040. ANS and NGVSP signify audio and navigation system and next-generation vehicle specific part, respectively. Domestic parts reuse market and PMs refining are two recycling processes for PMs.

2.5. Sensitivity analysis

A sensitivity analysis was carried out to identify the key parameters that affect the recycling potentials. Parameters (1) component weight including all targeted components and targeted components in NGVSPs, (2) PM concentration including PGMs in catalysts and Pd in PWBs (owing to the concern of industrial dynamics such as vehicle emission standards and substitution of elements[13,102,103]), and (3) part installation ratio were included, because they could be decisive to outcomes generated by the layered model and they might change considerably in 2040 owing to the evolution of the automotive industry. Parameters (4) newly registered vehicle number, (5) government's next-generation vehicle target, and (6) exported used vehicle number in the population balance model were included because they are vital to the calculation of ELVs generation. Parameter (7) component collection ratio is crucial to flow and stock calculations, thus it was included. Parameter (8) PM flow of export market to many destination countries, remains largely unclear so far, for this reason it was also included. Following the changing of a parameter by ±10%, the changed results (i.e. the total recycling potential of PMs in 2040) and default result were compared. The sensitivity was defined as in Eq. 9.

$$S(x) = \frac{RP_{PMs}(xv')}{RP_{PMs}(x)} \quad \text{Eq. 9}$$

Where x is the analyzed parameter, S is the sensitivity, RP_{PMs} is the total recycling potential (amount) of PMs, $RP_{PMs}(x)$ is the default result, v' is $\pm10\%$, and $RP_{PMs}(xv')$ is the changed result.

3. Results and discussion

3.1. PM contents per vehicle

Figure 20 shows the estimated PM contents per ELV in Japan from 2015 to 2040. The content of PMs per ELV ranges from 2.2 to 5.9 g in this period, of which StCVs (5.8−5.9 g) and HEVs (5.2−5.4 g) have the highest PM contents, followed by SmCVs (3.9−4.1 g), and EVs (3.2−3.5 g), while the content of PMs is lowest in MCVs (2.2−2.4 g). Notable results of the estimation include that (i) the vehicle types (including sizes) largely affect PM contents per ELV, and (ii) the changes of PM contents per ELV from 2015 to 2040 influenced by the part installation ratios (representing effects of time changing in this study) are relatively small. Furthermore, the PM contents per vehicle are tended to smaller than PM contents per vehicle sample in Chapter 2. The PM contents in samples were measured; on the other hand, the PM contents in vehicles were estimated. This was mainly due to the consideration of installation ratios and utilization of average values.

In terms of parts, catalytic converters and rear windows are the most PMs abundant, with PM contents reaching 0.7–2.5 g (excluding EVs) and 1.0–1.9 g respectively. The 24 other parts holding varying PM contents might be less significant individually. However, by grouping them, it becomes clear that ANSs (0.2–0.8 g), NGVSPs (around 0.7 g) in HEVs and EVs, and controllers (0.1–0.4 g) are also important in terms of PM mass. Narrowing the scale down to components, obviously catalysts and heating wires have the highest PM contents, because they are the key PM carriers in catalytic converters and rear windows, respectively. Additionally, PWBs also are main PM carriers, owing to their existence in ANSs, controllers, and NGVSPs. From the viewpoint of elements, the contents of Ag, Pd, Au, Pt, and Rh in an ELV are 1.2–2.7 g, 0.3–2.6 g, 0.2–0.4 g, 0.03–0.2 g, and 0.002–0.3 g, respectively. In addition, EVs have the lowest PGM contents (0.3–0.5 g), because they do not have catalytic convertors. Information about the PMs in each component of the various parts is provided in Appendix 13–17.

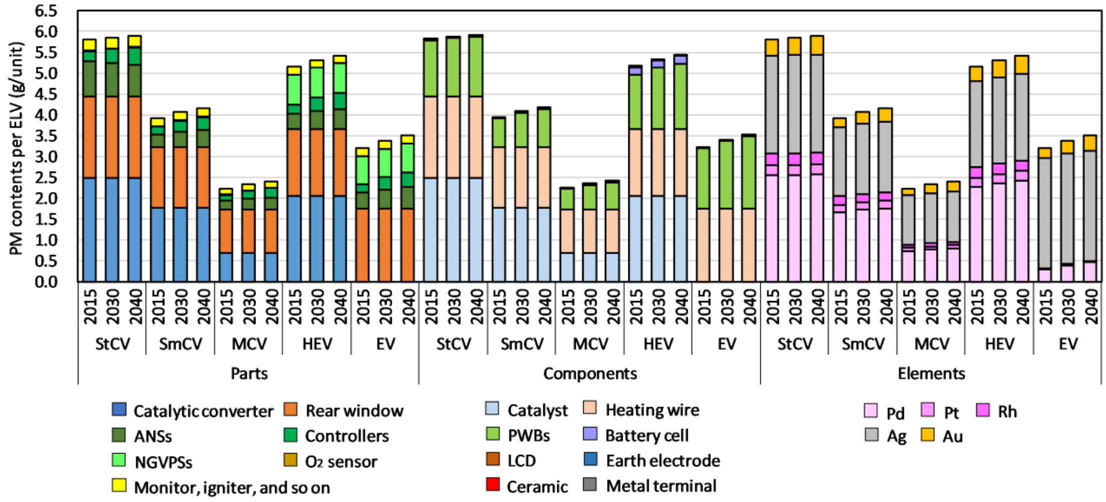

Figure 20. PM contents per ELV in Japan from 2015 to 2040. ANS and NGVSP signify audio and navigation system and next-generation vehicle specific part, respectively.

3.2. PMs in ELVs between 2015 and 2040

The change of vehicle types leads directly to changes in part and component types, as shown in Figure 21b and 21c. The PMs in the catalysts of catalytic convertors decrease by 26% to 4.5 t in 2040. In contrast, the PMs in PWBs of NGVSPs increase 16 times to 1.1 t in 2040, shares 7% of the total amount of PMs at that time. Meanwhile, the mass of PMs in PWBs of controllers also respectively increases by 49% to 1.1 t in 2040. Furthermore, the mass of PMs in the heating wires of rear windows remains relatively steady at around 5 t, because the change of vehicle types only minimally affects this component.

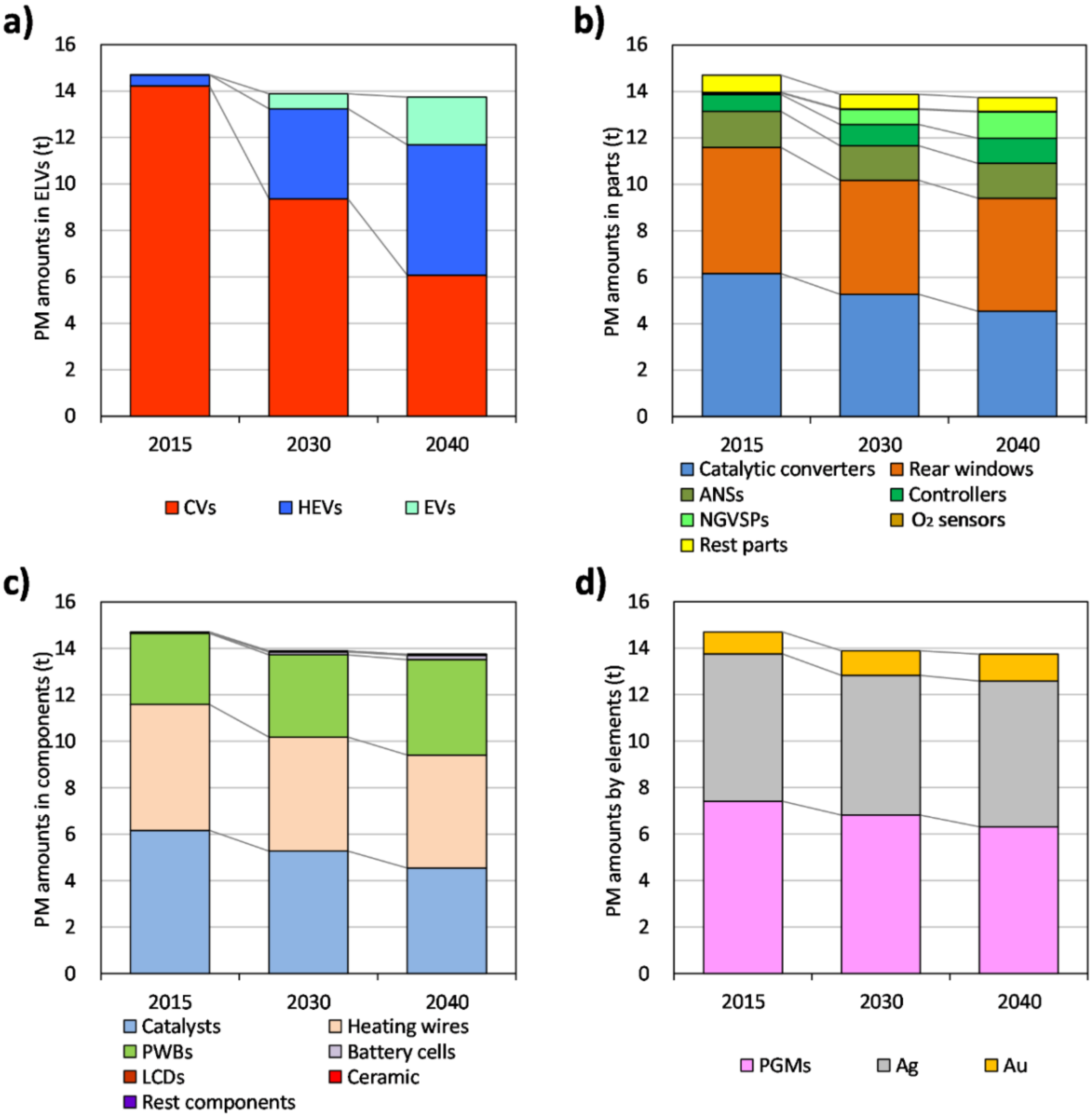

Figure 21. Annual PM amounts in ELVs in Japan from 2015 to 2040 subdivided by vehicles (a), parts (b), components (c), and elements (d). Vehicles: StCVs, SmCVs, and MCVs were grouped into CVs, elements: Pt, Pd, and Rh were grouped into PGMs, since CVs and PGMs have similar futures, and can share similar dismantling and recycling processes.

With respect to element type, as shown in Figure 21d, we estimated that from 2015 to 2040, the amounts of PGMs will decline by 15% to 6.3 t, Au increases by 22% to 1.2 t, meanwhile, Ag remains at about 6 t. The annual amounts of Ag and Au in Japan in 2015 were 6.3 t and 0.9 t, respectively. Restrepo et al.[18] estimated that the annual amounts of

Au and Ag were 9–30 kg and 30–80 kg, sourced from 0.1 million ELVs (considering only automotive electrics and electronics) in Switzerland in 2014. Based on these values, if Switzerland had the same ELV generation (i.e. 3.8 million) as Japan, then the annual amount of Au and Ag recovered in Switzerland would be 0.3–1.1 t and 1.1–3.0 t respectively for 2014. This matches the Au calculated in this study, but our Ag estimation is about twice to five times theirs. The main reason for this mismatch is due to the fact that we included the heating wires (containing 5.4 t Ag). The most notable features of plots of annual PM amounts in ELVs between 2015 and 2040 are that (i) although the total amount remains largely stable, (ii) PM proportions in vehicles, parts, and components are gradually changed, for instance, increased proportion occurs in next-generation vehicles. Therefore, the ELVs recycling system should adapt to the changing recycling needs, in particular with respect to the increased PWBs and heating wires in rear windows.

Furthermore, there will be about 104 t Ag, 94 t PGMs, and 17 t Au in 49 million unit vehicles in use in Japan as of 2040 while there were 87 t Ag, 103 t PGMs, and 15 t Au as of 2015.

3.3. PM flows and stocks in ELVs recycling system in 2015

Figure 22 shows the estimated PM flows and stocks in the ELVs recycling system in Japan, 2015. In total, all deregistered vehicles contained 17.2 t PMs. Among them, 14.7 t PMs (including 7.4 t PGMs, 6.3 t Ag, and 1.0 t Au) in ELVs flowed into the recycling system and were treated within the system; of these 33–53% (4.8–7.8 t PMs) were recycled through the domestic parts reuse market and PMs refining processes functionally. PMs refining is the crucial process (which recycled 4.2–6.9 t PMs) contributed to mostly by the PGMs in catalysts, and it dwarfs the domestic parts reuse market (which recycled 0.6–0.9 t PMs). Some of the reasons for this phenomenon from the dismantler side are difficulty dismantling many parts without damaging them and the time-consuming process of matching the used parts with vehicles; thus, for simplicity and speed, dismantlers seem to prefer PMs refining. We estimated that 4.0–5.3 t PGMs were refined nationwide in 2015. According to O'Connell et al.[104], 6.0 t PGMs were refined from ELVs in Japan in 2015. Comparing this value, our estimation is 0.7–2.0 t smaller, a possible reason might be because we only included passenger vehicles, and other vehicle types such as buses and trucks were excluded.

In addition, approximately 2.5 t PMs contained in 0.6 million unit (15% of total deregistered amount) used vehicles. Similarly, 0.2 t PMs contained in used parts/components. Their final fates and recycling efficiencies in the destination countries are unclear. As recycling of PMs is a global concern, these flows could be important for global management.

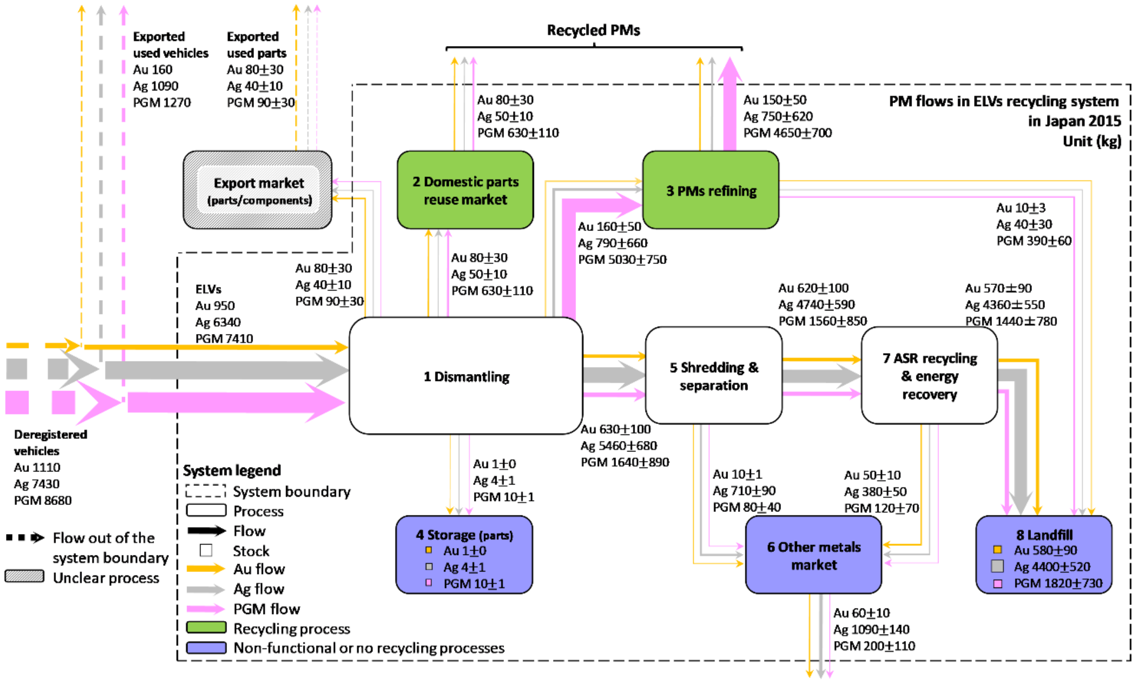

Figure 22. PM flows and stocks in the ELVs recycling system in Japan 2015. PGM signify platinum group metal. The mean values (between minimum and maximum collection ratios) are shown. The export market process is investigated in the questionnaire, but excluded from the estimation of recycling potentials. In addition, the emissions to the environment during ASR recycling and energy recovery are not considered.

More than half of the available Au and Ag were unable to be returned to their material streams in 2015. Of these, 3.9–4.9 t Ag and 0.5–0.7 t Au were sunk into the system through either storage or the landfill process, while 1.0–1.2 t Ag and around 0.05 t Au became impurities in other recycled metals. In total, the lost (non-functionally recycled and no recycled) Ag and Au in ELVs reached 4.8–6.1 t (76–97%) and 0.5–0.7 t (57–78%), respectively. It is reported that Japan demanded an average of 37 t Au annually for electrics, electronics, and machineries between 2010 and 2016, and imported 2.8 t Au per year in average to fill the gap between supply and demand in this period.[105] The loss of Au from ELVs accounts for 1–2% of the average Au used for electrics, electronics, and machineries, and 19–26% of the average Au import. Following the UN's Sustainable Development Goal 12,[12] 21 it is of great sustainable value to improve the current system as soon as possible to prevent accumulated loss over time.

3.4. Recycling potentials of PMs from ELVs in 2040: Amount in PWBs

Figure 23 shows the estimated recycling potential amounts of PMs from PWBs of ELVs in Japan, 2040. In 2040, a total of 1.9−2.5 t PMs could be potentially recycled from PWBs of various parts through selective dismantling. Audio systems (0.5−0.7 t) contribute the most, followed by navigation systems (0.3−0.4 t) and inverters (0.2−0.3 t). Additionally, hybrid computers (0.1−0.2 t), LIB units (around 0.1 t), etc. also provide considerable contributions. Considering the vehicle types, the recycling potential amounts of PMs from PWBs of next-generation vehicles (1.3−1.7 t) will be about twice that of conventional vehicles (0.6−0.9 t). For the amount from PWBs of next-generation vehicles, 41%, 33%, and 26% are shared by NGVSPs, ANSs, and controllers, respectively.

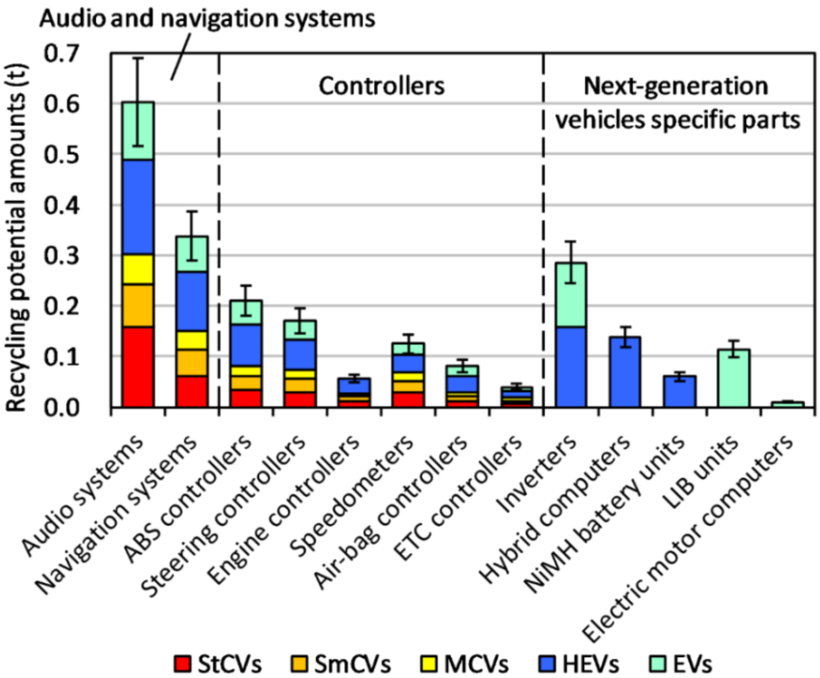

Figure 23. Recycling potential amounts of PMs from PWBs of ELVs in Japan 2040. ABS, ETC, NiMH, LIB and PWB signify anti-lock braking system, electronic toll collection, Nickel metal hydride (battery), Lithium-ion battery, and printed wiring board, respectively. The mean values (between minimum and maximum collection ratios) are shown.

3.5. Recycling potentials of PMs from ELVs in 2040: Potential ratio of selective dismantling scenarios

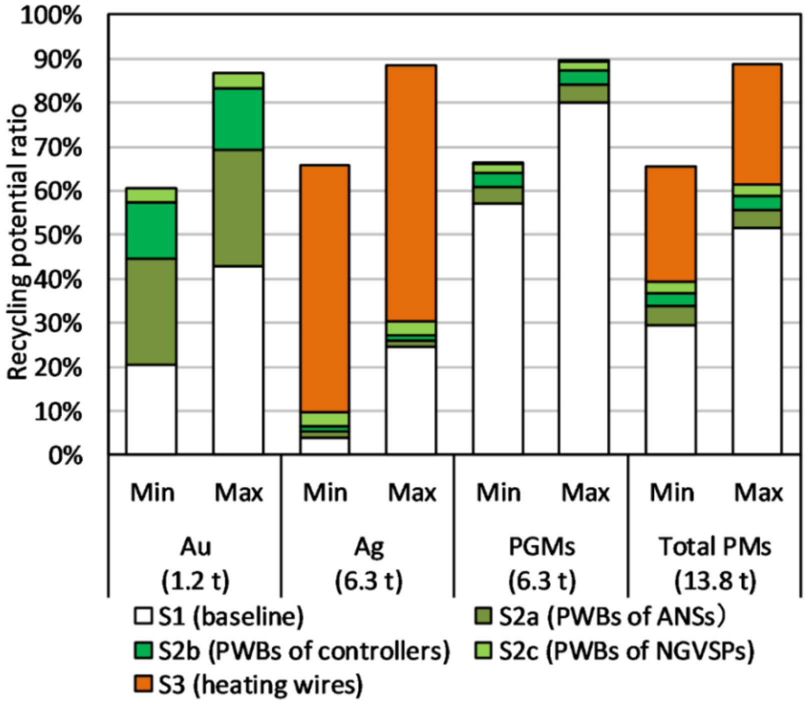

Figure 24. Recycling potential ratios of PMs by selective dismantling scenarios in Japan 2040. The minimum and maximum ratios reflect the minimum and maximum collections.

Figure 24 shows the estimated RPRs by selective dismantling scenarios in Japan 2040. We estimated 6.3 t Ag, 6.3 t PGMs, and 1.2 t Au will be contained in ELVs at that time. S1 which is the baseline scenario can offer 55–75% (3.4–4.8 t), 17–33% (0.2–0.4 t), and 4–24% (0.2–1.5 t) recycling of PGMs, Au, and Ag respectively. Through applying scenario 2a, which targeted the PWBs of ANSs, the RPR of Au can increase by 20% over the baseline scenario; it can also bring about a 4% improvement on PGMs and a 1% improvement on Ag recycling. Scenario 2b, which focused on the PWBs of controllers also can increase the RPR of Au by 11% compared with the baseline scenario to 28–44%; additionally, this scenario can deliver 3% and 1% enhancement on PGMs and Ag, respectively. Scenario 2c, which addressed the PWBs in NGVSPs, is the most efficient on Ag among the three sub-scenarios under scenario 2, and it can increase the RPR of Ag by 3% above the baseline scenario to about 7–27% (0.4–1.7 t). Further, recycling PWBs from NGVSPs can provide 3% and 2% enhancement on Au and PGMs, respectively. On the other hand, adoption of scenario 3 which is aimed at heating wires is remark able for recycling Ag, and under this scenario, the RPR of Ag can soar by 55%, compared with that of scenario 1, to about 58–80%. Optimistically, if scenario 2a, 2b, 2c, and 3 are combined and fully implemented simultaneously, the joint RPR of Au, Ag, and PGMs can reach, 50–67% (0.6–0.8 t), 64–85% (4.0–5.3 t), and 63–84% (4.0–5.3 t), respectively. In addition, the joint RPR of the total PMs can reach 62–83% (9–11 t).

For Japan towards 2040, (i) S2a is the most promising scenario under current industrialized ELVs recycling systems,[90,91] because it only focuses on PWBs of ANSs. Audio systems and navigation systems are normally centralized at a center console panel, and attached by some flexible fasteners (e.g. screws and grooves). Accordingly, this scenario requires low operational intensity, with only a few operations such as disconnecting, disassembly, and sorting to be added. (ii) scenario 2b and 2c are more challenging compared with scenario 2a, because there are more PWBs of controllers and NGVSPs, and they are usually dispersed and hidden in vehicles. In order to collect them efficiently, more operations compared with scenario 2a, such as cutting, breaking, and discharging (for batteries), are needed by various tools (e.g. pneumatic hammer and hydraulic plier). Thus, they are disadvantaged by the cost of intensive operations. However, if the concept of the "easy to dismantle vehicle"[92] is widely spread in vehicle design to minimize the cost of dismantling PWBs from controllers and NGVSPs, then, scenario 2b and 2c can become less challengeable. It is desirable to design and manufacture next-generation vehicles that allow easy identification and collection of their PM contents after use. According to the commercial dismantler we surveyed, parts in recent vehicles are indeed becoming easier to dismantle. (iii) The main strength of scenario 3 is that only heating wires need to be selectively dismantled which makes them object-oriented and simple. However, it also has the major weakness of heating wires being frequently printed on rear window glass;[106] currently there is still a lack of economically efficient methods to separate them. Therefore, whether this scenario can succeed, is largely reliant upon the manufacturers to develop some new assembly methods, allowing easier removal of heating wires from rear windows.

3.6. Sensitivities and uncertainty

The sensitivities of the selected parameters can be found in Table 12. Parameters (1) weights of all targeted components, (3) newly registered vehicle numbers, and (7) component collection ratios have clearly higher sensitivities than others. Thus, they are considered as sensitive parameters for this study. We have a relatively high confidence level on parameter (1) and (7), because they were derived directly from on-site sampling and questionnaire. For parameter (3), we assumed that it will remain 4.0 million (SI, Figure S2) from 2020 to 2040, based on the estimation from the Japanese Automobile Dealers Association. However, it has been reported that the population of Japan will decrease from 127 million in 2015 to 113 million in 2040, which probably will cause a decrease in newly registered vehicles. This might, in turn, lead to an overestimation of the potential.

Table 12. Sensitivities of selected parameters in chapter 4. The closer the sensitivity is to one, the greater the sensitivity is.

No.	**Parameter, *x***		**Sensitivity, *S(x)***	
			When parameter +10%	**When parameter -10%**
1	Component weight	All components	1.10	0.90
		Components in NGVSPs	1.01	0.99
2	PM concentration	Pd in PWBs	1.01	0.99
		PGMs in catalysts	1.04	0.96
3	Part installation ratio		1.01	0.99
4	Newly registered vehicle number		1.12	0.88
5	Next-generation vehicle target		0.99	1.01
6	Exported used vehicle number		0.98	1.02
7	Component collection ratio*		1.06	0.90
8	PM flow of export (parts/components) market^		1.01	–

* Collection ratio has an upper limit of 100%, therefore, when the collection level has reached 75–100%, it can only increase to 83–100%.

^ We conservatively assumed that exported parts/components are neither reused nor refined overseas, because their final fates and recycling efficiencies are largely unclear; the default value is 0, thus only +10% is effective in the case that 10% of PM in parts/components can be recycled overseas.

In addition, for parameter (1) weights of targeted components in NGVSPs, we did not consider some new emerging parts that also contain PMs, such as fuel cells from fuel cell vehicles.[107] The effects of such factors are difficult to predict, but the absence of them might lead to underestimation of the potential. Although parameter (2) Pd concentrations in PWBs, is not obviously sensitive, it was estimated that about two-thirds of Pd in PWBs (mainly containing low performance capacitors) in the market could be substituted by Ni since 2003.[102] Note that the substitution was started largely owing to the dramatic rise of Pd prices at that time, but the fact is that Pd still performs better than the substituted elements, which means that some high performance capacitors might still need to use Pd.[102] Some of our sample ELVs were produced before 2003. If we also assume that two-thirds of the Pd contents in these samples' PWBs were substituted by Ni since 2003, up to 2040, the recycling potential of PGMs will decrease by 13%. This might lead to an overestimation of the potentials.

4. Conclusion

We developed substance flow and scenario analyses based on estimations of PM contents per ELV and ELV generations to explore their recycling potentials in Japan. The obtained conclusions were shown below:

- We predict that in Japan, from 2015 to 2040, the content of PMs per ELV will range from 2 to 6 g, and the annual amount of PMs in ELVs will remain largely stable, at 14–15 t,.
- The proportions of PMs utilized in different vehicles, parts, and components will gradually change; in particular, increased proportions will occur in the printed wiring boards (PWBs) of next-generation vehicles.
- The results also show that in Japan, totals of 33–53% of PMs in ELVs were recycled in 2015, and that by selective dismantling of PWBs and heating wires in the rear windows of ELVs, the recycling potentials of PMs could be optimally increased to a maximum of 62–83% by 2040.

In all, considering the situations, we recommend that at the downstream, stakeholders (e.g. government and dismantlers) can start to establish a complete dismantling procedure on collecting PWBs from ANSs. In the meantime, they can try to enhance the dismantling procedure to collect more PWBs from controllers and NGVSPs; at the upstream, stakeholders (e.g. manufactures) should note the PM-containing components in vehicles such as heating wires in rear windows, and put effort into designing and producing those components that facilitate disassembly in the end-of-life stage.

Chapter 5 Conclusion

Recycling of some minor but essential scarce metals used in vehicles may serve as an important strategy to strengthen the sustainable management of natural resources. Accordingly, this study focused on two groups of scarce metals: REEs (i.e. La, Ce, Pr, Nd, and Dy) and PMs (Pt, Pd, Rh, Au, and Ag) in both end-of-life conventional and next-generation vehicles. To explore the potential amount of those scarce metals that could be recycled from ELVs in Japan, the on-site ELVs dismantling survey including six ELV samples were carried out firstly to collect the raw data for modeling. Then, based on the data of element contents found by the survey, as well as, the estimations of ELV generations up to 2040, we mapped the element distributions in current ELVs recycling system by means of SFA, Finally, the potential recycling amounts (2030 for REEs and 2040 for PMs) were estimated through scenario analyses.

This study focused on the case in Japan. However, the estimation model could also be applicable for many other countries in developing polices and technologies for ELVs management.

1. Conclusions of the thesis

The main findings are:

- The REE and PM contents per sample range 22−3100 g and 3.1−9.6 g, respectively, based on the dismantling survey. By considering factors such as installation ratio and average value, the estimated REE and PM content per vehicle are 53−2800 g (without considering MCVs and EVs) and 2.2−5.9 g. Next-generation vehicles and their specific parts tended to contain higher amounts of REEs and PMs.
- The annual amount of REEs in ELVs will increases dramatically from 130 t in 2010 to 3400 t in 2030, mostly owing to the increasing use of HEVs. Regarding PMs, the annual amount in ELVs will remain largely stable at 14−15 t between 2015 and 2040, but the proportions of PMs utilized in different vehicle parts will gradually change, in particular, increased proportions will occur in the PWBs of next-generation vehicles.
- In the current ELVs recycling system, the amount of REEs can be recycled was quite low in recent years, this is mainly due to the small number of end-of-life HEVs generation and the storage without treatment of secondary batteries. Additionally, 4.8−7.8 t (33−35% of total) PMs were recycled by the system in 2015.

Still more than half of Au and Ag are unable to be returned to their material streams.

- In the perspective of mass, it is of great sustainable value to pay attentions on NiMH batteries, motors in terms of REEs, as well as, various electronics containing PWBs and heating wires of rear windows in terms of PMs. By collecting and recycling of them, in total 2200−3200 t REEs in 2030 and can 9−11 t PMs can be optimistically recycled in 2040. Furthermore, it is better to improve the current system as soon as possible to prevent the accumulated loss over time.

2. Suggestions for ELVs management and future studies

- The global automotive industry is now undergoing a rapid evaluation era. Major changes (e.g. substitution of materials and development of new technologies) might happen in near further. In this study, we observed the reduction of REEs in battery cells through vehicle generations. At the upstream, manufactures and suppliers should note and updated information related to the REE/PM-containing components and put efforts on designing “easy to dismantling vehicles”.
- At the downstream, dismantlers and refineries can start to and establish a complete dismantling procedure for collecting the NdFeB magnets and PWBs in ELVs. In particular, NdFeB magnets in hybrid transmission motors, power steering motors, and PWBs in audio and navigation systems, controllers, and next-generation specific parts should be addressed on.
- As automotive industry continues evolving, new type of vehicles and/or parts might be largely applied in future. Therefore, it is recommended as future studies to conduct new dismantling surveys focusing on the latest developed promising vehicles and/or parts. Accumulate bottom-up data are helpful for adjusting the dynamical substance flows.
- Cooperating with a commercial dismantler, the automotive PMs-containing components collection questionnaire was carried out to identify recycling flows. To improving the accuracy of recycled REE and PM flows in Japan, we recommend that future studies to investigate more about the collection ratios and final fates by cooperating with dismantlers and manufactures (use of recycled materials in production). In addition, further discussions of how to manage secondary batteries which contain critical metals (e.g. REEs, PMs, and others) should be made in perspectives of function, composition, and recycling technology.

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First of all, I would like to express my sincere gratitude to my supervisor Prof. Sakai for the continuous support of my PhD study and related researches, for his patience, motivation, and immense knowledge. His guidance helped me in all the time of research. This thesis could not have been done without him.

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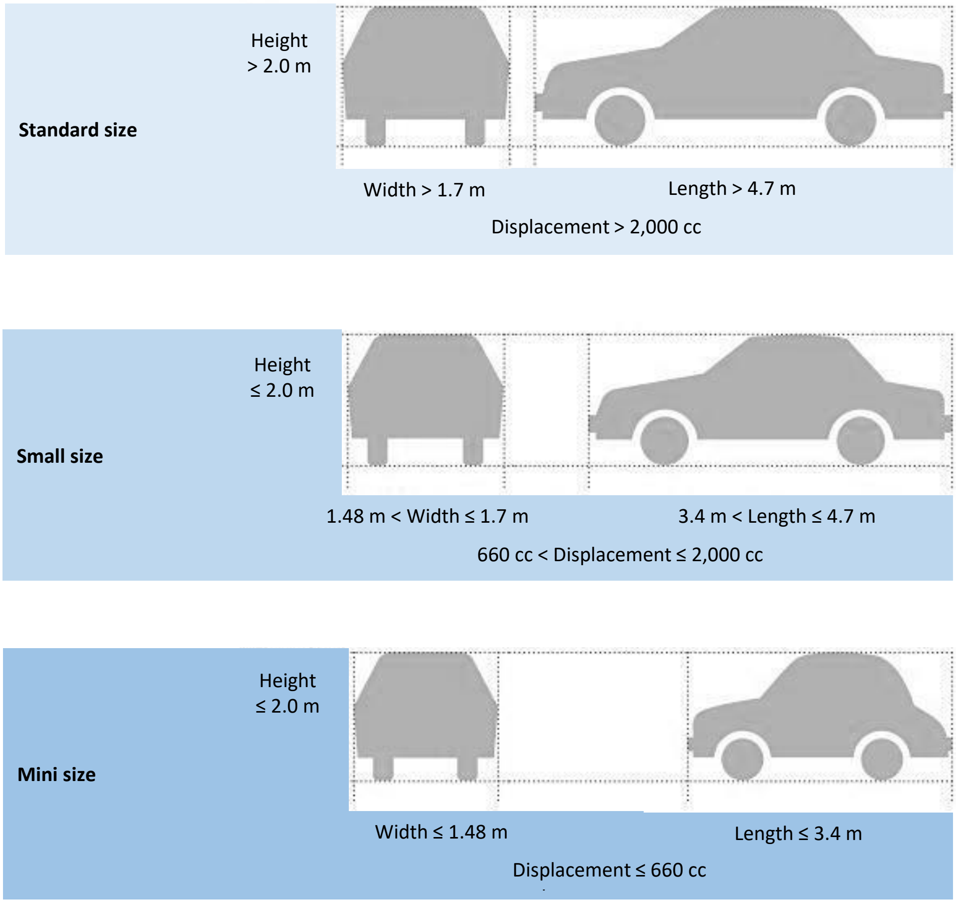
Appendix

Appendix 1. Size classification of passenger vehicles under the Road Vehicle Act in Japan. This classification is used for registration and inspection. If a vehicle exceeds any one of the stipulations in a size, that vehicle is automatically classified into the larger size.



Appendix 2. Components collected from HEVs between 1st, 2nd and 3rd generations

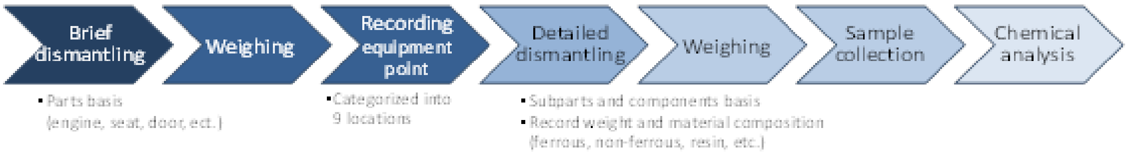
Parts	Components collected for chemical analysis	HEV generation		
		1997–2003	2003–2009	2009–2015
		1st HEV	2nd HEV	3rd HEV
NiMH battery unit	Battery cell, PWBs	●	●	●
Inverter	PWBs	●	●	–
Hybrid transmission[a]	Magnets	●	●	●
Electric power steering motor	Magnets	●	●	●
Air compressor motor	Magnets	–[b]	●	●

[a] Hybrid transmission has two motors: a driving motor and a generator.

[b] The air compressor is non-electrical, it lacks motor magnets.

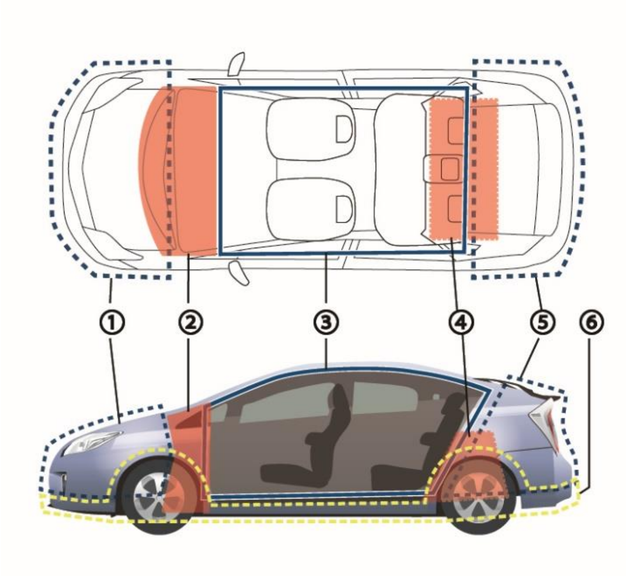
●: installed and –: no installation

Appendix 3. Procedure of dismantling survey

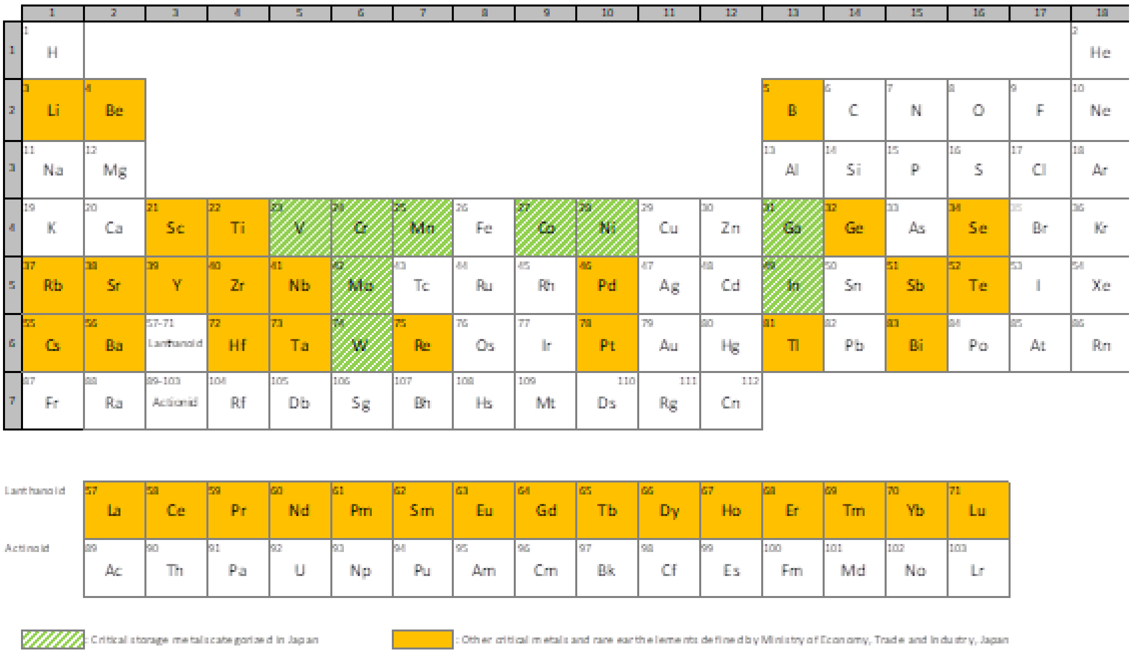


Appendix 4. Parts classification based on location

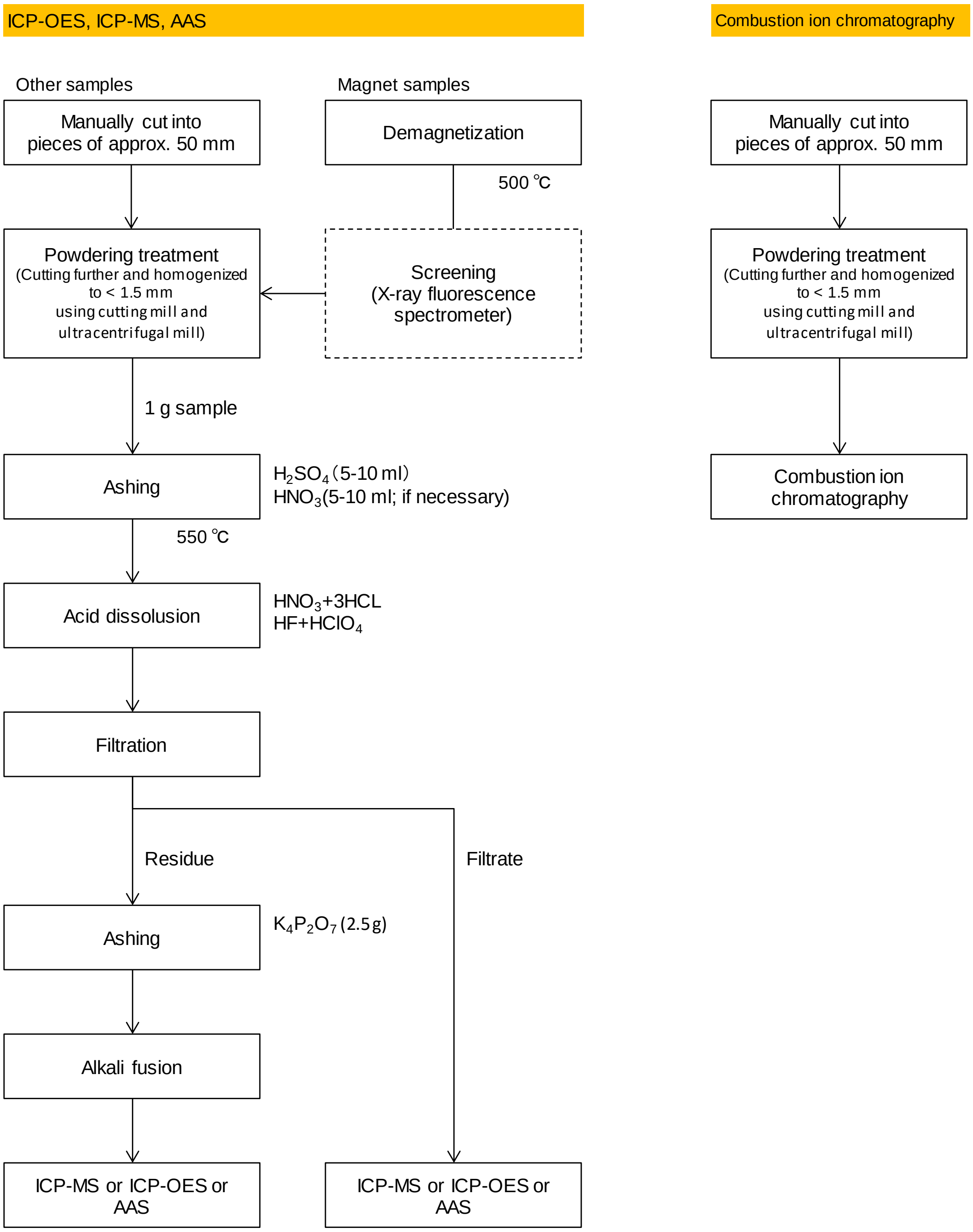
Category (Location)		No.
Engine room		1
Interior	Front	2
	Seat area	3
	Rear	4
	Trunk	5
Exterior	Suspension	6
	Other exterior	–
Remaining body		–
Others		–

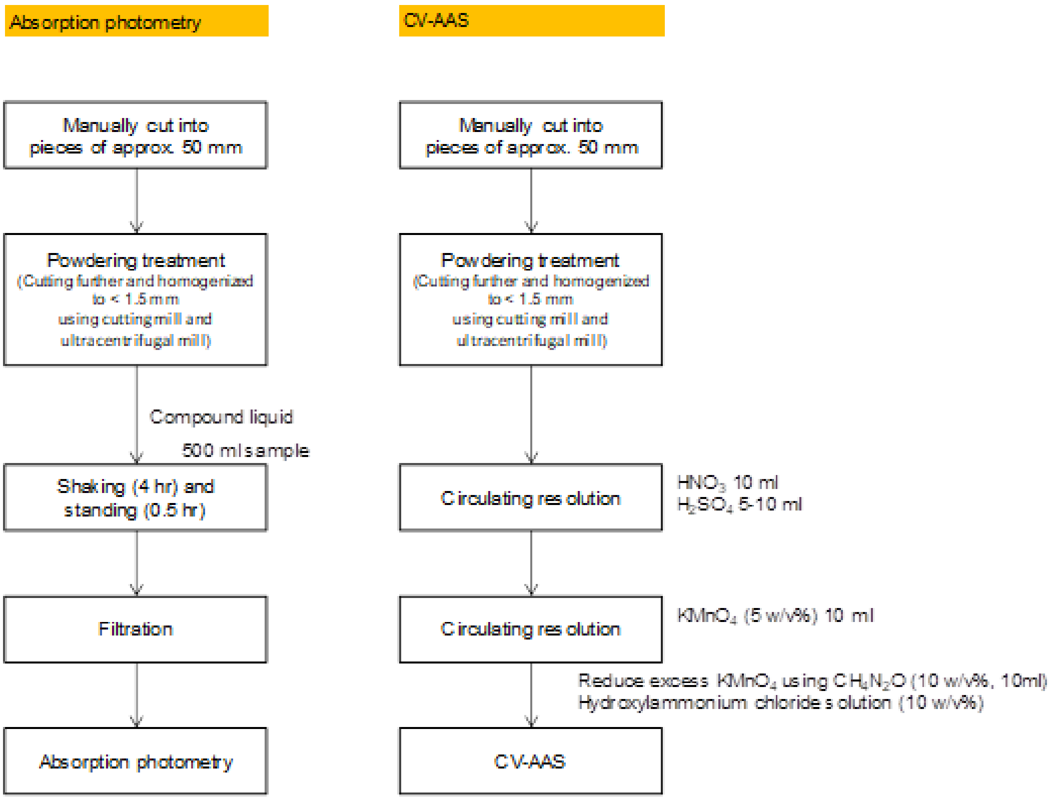


Appendix 5. Critical metals defined in Japan by METI Ministry of Economy, Trade and Industry, Japan



Appendix 6. Procedure of chemical analysis. ICP-OES: inductively coupled plasma optical emission spectrometry, ICP-MS: inductively coupled plasma mass spectrometry, AAS: atomic absorption spectrophotometry, CV-AAS: cold vapor atomic absorption spectrophotometry, Combustion-IC: combustion ion chromatography.




Absorption photometry
CV-AAS
Manually cut into pieces of approx. 50 mm
Powdering treatment (Cutting further and homogenized to < 1.5 mm using cutting mill and ultracentrifugal mill)
Compound liquid
500 ml sample
Shaking (4 hr) and standing (0.5 hr)
Filtration
Absorption photometry
Manually cut into pieces of approx. 50 mm
Powdering treatment (Cutting further and homogenized to < 1.5 mm using cutting mill and ultracentrifugal mill)
Circulating resolution
HNO3 10 ml
H2SO4 5-10 ml
Circulating resolution
KMnO4 (5 w/v%) 10 ml
Reduce excess KMnO4 using CH4N2O (10 w/v%, 10ml)
Hydroxylammonium chloride solution (10 w/v%)
CV-AAS

Appendix 7. Material composition of each vehicle

Type	Production year	Metals (%)				Plastics (%)					Textiles (%)		Glass (%)	Rubbers (%)	Others (%)	Unidentified (%)
		Ferrous	Copper	Aluminum	Others	PE	PP	ABS	Urethane	Others	Homogenized	Mixed				
Conventional vehicle:																
MCV99	1999	67.5	0.8	2.0	5.1	0.4	4.1	0.4	3.3	0.5	1.3	1.1	4.7	3.3	0.1	5.4
MCV09	2009	70.0	0.5	2.4	5.0	0.4	3.9	0.5	3.4	1.0	1.2	0.6	4.4	2.8	0.1	3.9
SmCV	1997	70.8	0.9	2.6	7.4	0.3	3.2	0.3	1.7	0.9	1.9	0.3	3.4	3.4	0.0	2.9
StCV	1997	69.5	1.1	3.3	5.3	0.4	4.3	0.3	2.8	0.8	1.4	0.3	4.6	3.0	0.6	2.2
Next-generation vehicle:																
HEV	1998	66.3	2.9	2.9	6.2	0/4	2.8	0.0	2.1	1.4	1.5	0.6	3.7	3.5	0.3	5.5
EV	2011	58.7	4.3	3.7	2.8	0.1	3.2	0.3	2.0	1.3	1.0	0.3	2.8	2.8	0.2	16.4

PE: polyethylene

PP: polypropylene

ABS: acrylonitrile-butadiene-styrene

“Others” refers to materials that were identified but not allocated to a category (metals, etc.).

“unidentified” refers to unidentified materials and materials contained in complex parts.

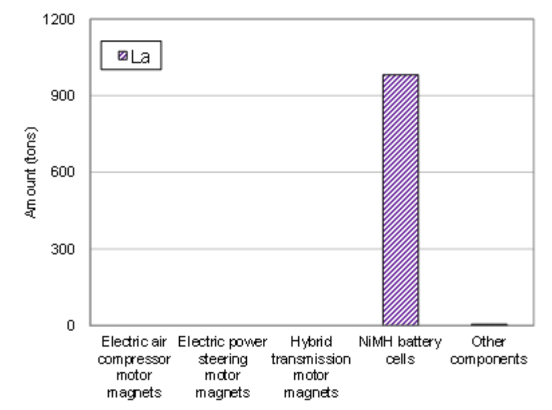
Appendix 8. Overview of the element concentrations of magnets in ELVs. * Samples were analyzed by wavelength dispersive X-ray fluorescence spectrometer. ICP was not used.

Type		Wiper motor	Power window motor	Radiator fan motor	Blower fan motor	Rear brake	Speaker	Electric power steering motor					
		EV*	EV*	EV*	EV*	EV*	1st HEV*	MCV99*	MCV09*	1st HEV*	2nd HEV*	3rd HEV*	EV*
[illegible]	(g)	60	119	52	145	4	292	140	193	216	415	418	102
Fe	(ppm)	850000	960000	990000	870000	570000	910000	860000	840000	870000	870000	870000	670000
Co		13000	5000	12000		30000			16000	2000			31000
Sr		89000			110000		80000	110000	91000	110000	110000	120000	
La		38000	25000										
Pr													210000
Nd						260000							82000
Dy						58000							4000

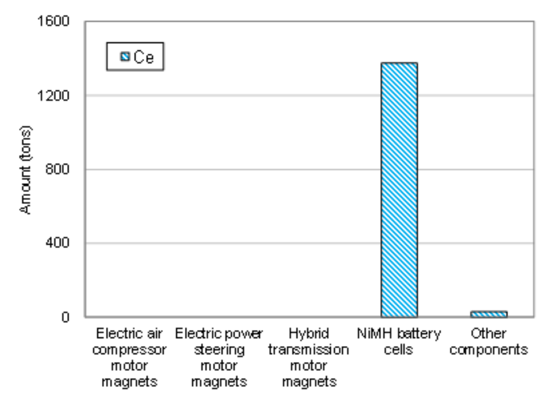
Table continuing

Type		Air compressor motor				Driving motor				Generator		
		MCV09*	2nd HEV	3rd HEV	EV*	1st HEV	2nd HEV	3rd HEV	EV	1st HEV	2nd HEV	3rd HEV
[illegible]	(g)	116	62	43	78	1134	1183	732	1889	368	274	396
Fe	(ppm)	850000	700000	690000	660000	680000	680000	670000	650000	700000	680000	730000
Co		9000	9600	17000	18000	12000	11000	22000	22000	11000	4800	5300
Sr		95000										
La												
Pr			53000	500		47000	53000	1100		49000	2500	470
Nd			180000	260000	280000	180000	180000	250000	270000	170000	250000	210000
Dy			67000	48000	38000	99000	82000	52000	65000	98000	55000	76000

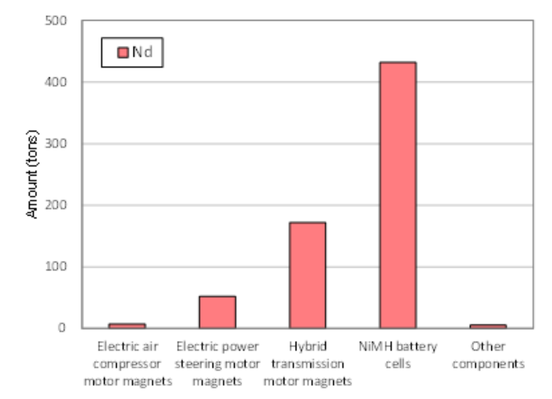
Appendix 9. Distribution of each REE in vehicle components in 2030



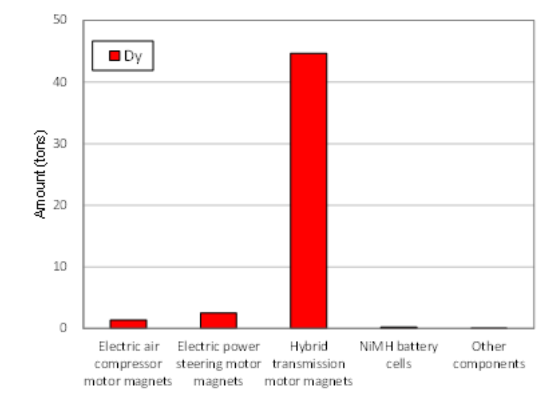
(a)



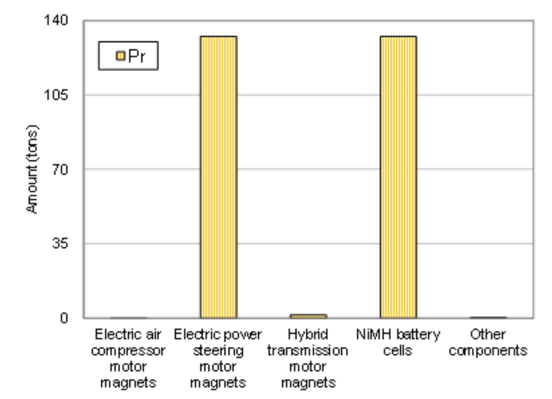
(b)



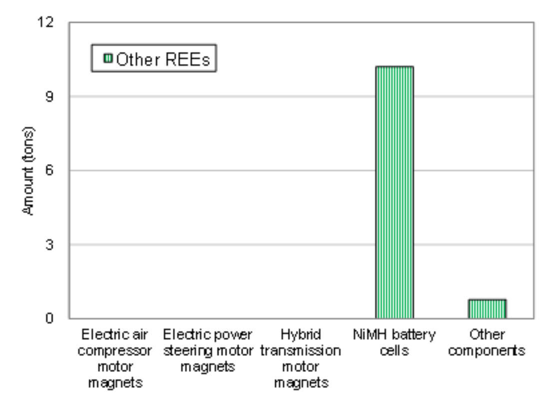
(c)



(d)



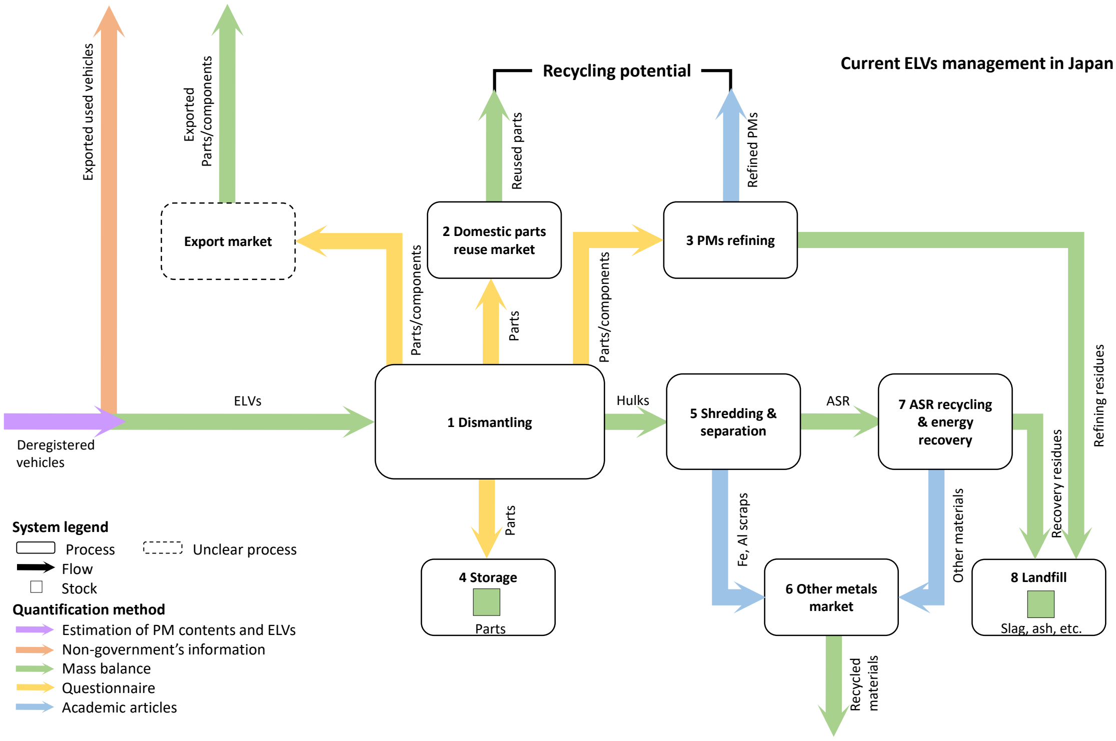
(e)



(f)

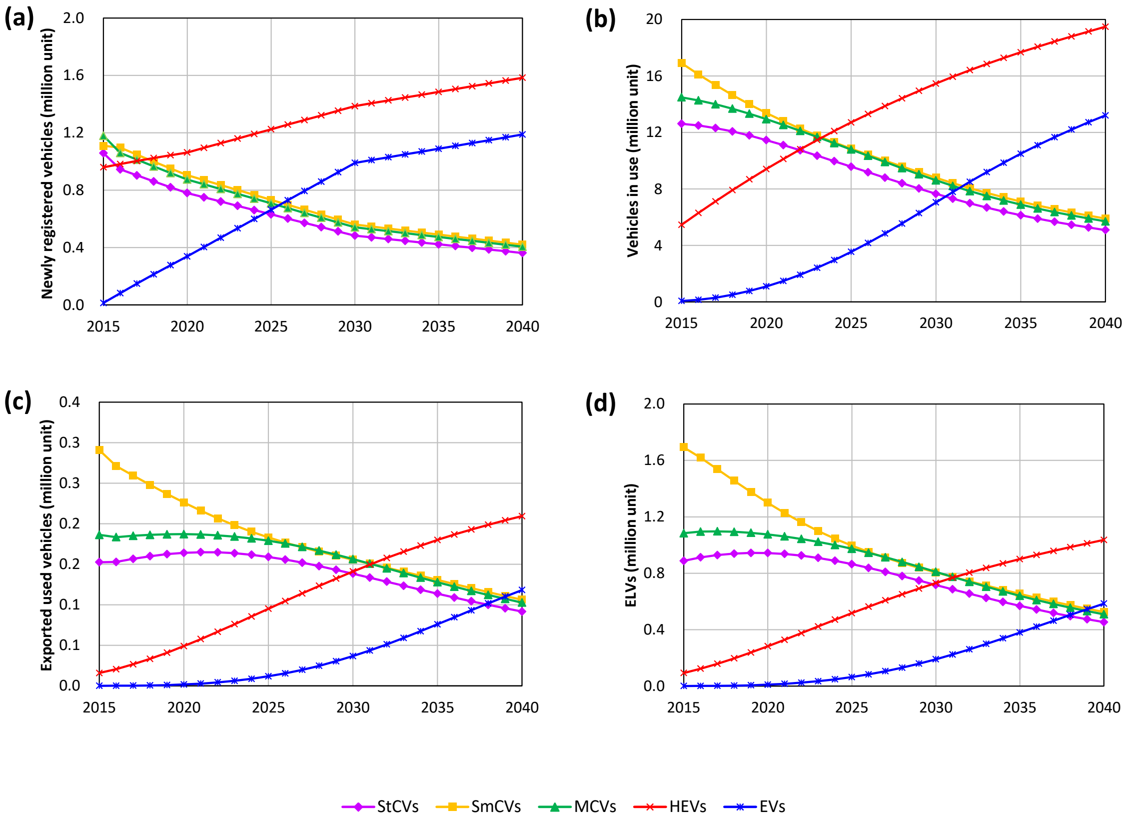
Appendix 10. Relationship between REE demand and recovery for vehicles in Japan. Demand: required element to meet the production of first registered vehicle in a specific year. Ranges reflect the recovered REEs by different recovery efficiencies.

Type			**Unit**	**Year**		
				2010	**2020**	**2030**
Demand for vehicle: D	Nd	For NiMH battery cell	t	115	352	469
		For NdFeB magnet		168	450	587
	Dy	For NdFeB magnet		34	100	133
Recovery for ELVs: R	Nd	From NiMH battery cell		8—11	101—141	307—428
		From NdFeB magnet		3—6	37—62	111—183
	Dy	From NdFeB magnet		0.7—1.2	7—12	23—38
R/D	Nd	NiMH battery cell	%	7—10	29—40	65—91
		NdFeB magnet		2—4	8—14	19—31
		Total		4—6	17—25	40—57
	Dy	NdFeB magnet		2—4	7—12	17—29



Appendix 11. Processes, flows, and stocks of PMs in current ELVs management in Japan. The emissions to the environment during ASR recycling and energy recovery are not considered.

Appendix 12. Estimated newly registered vehicles (a), vehicles in use (b), exported used vehicles (c), and ELV generations (d) in Japan between 2015 and 2040. Total number of ELVs in 2015, 2030, and 2040 is 3.8 million, 3.3 million, and 3.1 million, respectively.



Appendix 13. Estimated PM contents in components of StCVs. Note that in this table and following Appendix 22, 23, 24, and 25, weight refers to a set of components; parts installation ratios are not considered; in addition, the heating wire was sampled and analyzed together with the rear window glass; thus, their total weight is shown.

Component (part)	**Weight**	**PMs**					
		Ag	**Rh**	**Pd**	**Pt**	**Au**	**Total**
PWBs (ANSs: audio system* and navigation system)	2785	0.28	0.00	0.70	0.00	0.53	1.52
PWBs (controllers: Engine controller, Steering controller, ABS controller, air-bag controller, speedometer, ETC controller)	1486	0.12	0.00	0.18	0.00	0.14	0.44
PWBs (NGVSPs: NiMH battery unit, LIB unit, hybrid computer, electric motor computer, inverter)	–	–	–	–	–	–	–
PWBs (other parts)	398	0.15	0.00	0.06	0.02	0.04	0.27
Battery cell (NiMH battery)	–	–	–	–	–	–	–
Catalyst (catalytic converter)	1068	0.02	0.28	1.98	0.21	0.00	2.48
LCD (center console monitor)	102	0.00	0.00	0.00	0.00	0.00	0.01
Ceramic (O_2 sensor)	5	0.00	0.00	0.00	0.01	0.00	0.01
Earth electrode (igniter)	2	0.00	0.00	0.00	0.00	0.00	0.00
Metal terminal (rear window)	12	0.00	0.00	0.00	0.00	0.00	0.00
Heating wire (rear window)	7486	1.95	0.00	0.00	0.00	0.00	1.95

Unit (g)

* Sub-part CD changer of audio system is considered to be only installed in StCVs partially.

Appendix 14. Estimated PM contents in components of SmCVs

Component (part)	Weight	PMs					
		Ag	Rh	Pd	Pt	Au	Total
PWBs (ANSs: audio system and navigation system)	1173	0.06	0.00	0.18	0.00	0.20	0.45
PWBs (controllers: Engine controller, Steering controller, ABS controller, air-bag controller, speedometer, ETC controller)	1109	0.09	0.00	0.13	0.00	0.11	0.33
PWBs (NGVSPs: NiMH battery unit, LIB unit, hybrid computer, electric motor computer, inverter)	–	–	–	–	–	–	–
PWBs (other parts)	294	0.11	0.00	0.05	0.01	0.03	0.20
Battery cell (NiMH battery)	–	–	–	–	–	–	–
Catalyst (catalytic converter)	766	0.01	0.20	1.42	0.15	0.00	1.78
LCD (center console monitor)	75	0.00	0.00	0.00	0.00	0.00	0.01
Ceramic (O_2 sensor)	4	0.00	0.00	0.00	0.01	0.00	0.01
Earth electrode (igniter)	1	0.00	0.00	0.00	0.00	0.00	0.00
Metal terminal (rear window)	9	0.00	0.00	0.00	0.00	0.00	0.00
Heating wire (rear window)	5527	1.44	0.00	0.00	0.00	0.00	1.44
Unit (g)							

Appendix 15. Estimated PM contents in components of MCVs. Values are the average values of two MCV samples.

Component (part)	Weight	PMs					
		Ag	Rh	Pd	Pt	Au	Total
PWBs (ANSs: audio system and navigation system)	837	0.04	0.00	0.13	0.00	0.15	0.32
PWBs (controllers: Engine controller, Steering controller, ABS controller, air-bag controller, speedometer, ETC controller)	805	0.06	0.00	0.10	0.00	0.08	0.25
PWBs (NGVSPs: NiMH battery unit, LIB unit, hybrid computer, electric motor computer, inverter)	–	–	–	–	–	–	–
PWBs (other parts)	210	0.08	0.00	0.03	0.01	0.02	0.14
Battery cell (NiMH battery)	–	–	–	–	–	–	–
Catalyst (catalytic converter)	299	0.00	0.08	0.55	0.06	0.00	0.69
LCD (center console monitor)	54	0.00	0.00	0.00	0.00	0.00	0.00
Ceramic (O_2 sensor)	3	0.00	0.00	0.00	0.01	0.00	0.01
Earth electrode (igniter)	1	0.00	0.00	0.00	0.00	0.00	0.00
Metal terminal (rear window)	6	0.00	0.00	0.00	0.00	0.00	0.00
Heating wire (rear window)	3944	1.03	0.00	0.00	0.00	0.00	1.03
Unit (g)							

Appendix 16. Estimated PM contents in components of HEVs

Component (part)	Weight	PMs					
		Ag	Rh	Pd	Pt	Au	Total
PWBs (ANSs: audio system and navigation system)	1323	0.07	0.00	0.21	0.00	0.23	0.51
PWBs (controllers: Engine controller, Steering controller, ABS controller, air-bag controller, speedometer, ETC controller)	1352	0.04	0.00	0.23	0.00	0.14	0.41
PWBs (NGVSPs: NiMH battery unit, LIB unit, hybrid computer, electric motor computer, inverter)	1459	0.24	0.00	0.27	0.01	0.06	0.54
PWBs (other parts)	534	0.12	0.00	0.02	0.02	0.02	0.18
Battery cell (NiMH battery)	43300	0.03	0.02	0.04	0.02	0.02	0.11
Catalyst (catalytic converter)	880	0.01	0.23	1.63	0.17	0.00	2.05
LCD (center console monitor)	71	0.00	0.00	0.00	0.01	0.00	0.01
Ceramic (O_2 sensor)	4	0.00	0.00	0.00	0.01	0.00	0.01
Earth electrode (igniter)	2	0.00	0.00	0.00	0.00	0.00	0.00
Metal terminal (rear window)	10	0.00	0.00	0.00	0.00	0.00	0.00
Heating wire (rear window)	6230	1.62	0.00	0.00	0.00	0.00	1.62
Unit (g)							

Appendix 17. Estimated PM contents in components of EVs

Component (part)	Weight	PMs					
		Ag	Rh	Pd	Pt	Au	Total
PWBs (ANSs: audio system and navigation system)	1429	0.07	0.00	0.22	0.00	0.25	0.55
PWBs (controllers: Engine controller, Steering controller, ABS controller, air-bag controller, speedometer, ETC controller)	1136	0.04	0.00	0.24	0.00	0.10	0.39
PWBs (NGVSPs: NiMH battery unit, LIB unit, hybrid computer, electric motor computer, inverter)	428	0.65	0.00	0.01	0.00	0.01	0.68
PWBs (other parts)	577	0.13	0.00	0.02	0.02	0.02	0.20
Battery cell (NiMH battery)	–	–	–	–	–	–	–
Catalyst (catalytic converter)	–	–	–	–	–	–	–
LCD (center console monitor)	77	0.00	0.00	0.00	0.01	0.00	0.01
Ceramic (O_2 sensor)	–	–	–	–	–	–	–
Earth electrode (igniter)	–	–	–	–	–	–	–
Metal terminal (rear window)	11	0.00	0.00	0.00	0.00	0.00	0.00
Heating wire (rear window)	6730	1.75	0.00	0.00	0.00	0.00	1.75
Unit (g)							