

Materials recycling technology for recovering rare earth fluorescent powder from fluorescent lamp sludge

— Pioneering near-future resource circulation —

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A materials recycling technology to recover green phosphor, including terbium, which is a heavy rare earth, from fluorescent powder in waste lamp sludge was realized by collaborating with researchers who specialize in materials and powder sorting. There are few cases worldwide in which materials circulation from post-consumer waste has been established in loops further inside than horizontal recycling. This is a recycling system which is described as an ideal circulation system for the near future in Europe's circular economy (CE)/resource efficiency (RE) policies, etc. This study is a successful example of Japan leading the world in urban mine development to establish a resource circulation system of various waste products, and becoming a pioneer in near-future resource recycling.

Keywords : Rare earth, fluorescent powder, high gradient magnetic separation, magnetic matrix, terbium, urban mine

1 Introduction

Japan relies on imports for most of metal resource materials. It is the world's largest importer of rare metals that are essential in manufacturing high-tech devices, and one may say that the manufacturing industry exists through the import of rare metal resources. The problems of rare metal supplies are that their production control is difficult as many metals are by-products, they invite oligopoly since the consumption volume is low, and they are influenced easily by international affairs. Compared to base metals that are heading toward gradual depletion, rare metals are susceptible to man-made factors such as the affairs of the producing countries, and it is difficult to forecast which rare metals will have the supplies cut off.

The rare earths (17 elements) are types of rare metals, and their market is dominated by China. The "rare earth crisis" is still fresh in our memory, as the domestic industries were hit hard, although temporarily, by the trade embargo against Japan and the following restrictions of export volume. Rare earth resources exist throughout the world including the USA and Australia, and recently, submarine deposits in the Japanese exclusive economic zone (EEZ) was in the news. The reason why China captured the current market is because they possess extremely good mines. One factor is that the products from the Chinese mines hardly contain any radioactive impurities seen in other rare earth mines. Therefore, only simple processing is necessary for production. Another factor is that they have heavy rare earth products such as dysprosium and terbium that are used as magnetic and fluorescent materials. Particularly, in the world

movement toward low carbonization, the heavy rare earth resources that are necessary for the manufacture of high-performance motors are attracting high attention.

This research was started before the rare earth crisis became serious. Around 2005, the price of rare earth was gradually increasing, and the urban mine resources in Japan that were practically untouched suddenly gained attention. At the time, the authors were conducting research and development of physical sorting technology of magnets and capacitors,^[1] while the objective of this research was to concentrate and recover green phosphor LAP that contains terbium abundantly from waste fluorescent lamps. The fluorescent lamps are roughly divided into white light types and three-wavelength (rare earth) types. Three types of fluorescent powder corresponding to RGB are used in the three-wavelength type lamps. The topics of this research are as follows: to remove white fluorescent powder, glass, and other impurities from the mixed sludge of white and three-wavelength waste fluorescent powders, to concentrate the green (G) LAP, and then to reuse it as fluorescent powder material.

Traditionally, fluorescent powder sorting was studied at universities, and sorting using particle density and surface hydrophobicity/hydrophilicity was attempted. However, it was difficult to separate the white and three-wavelength types, and no practical application was achieved. A hint for success of this research was found when the authors (Akai and Yamashita) saw that LAP selectively attaches to the magnet when it is placed under extremely strong magnetic force, although it seems to be unresponsive to magnets. To

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utilize this principle in industry, a high-gradient magnetic separator was improved and made into a device to achieve highly concentrated LAP (Oki). In this research, a separation system was developed and brought to practical realization through the collaboration of researchers specializing in materials and sorting. The following is an account of the road to practical realization particularly from the viewpoint of separator device development.

2 Building up to the development of a separator

2.1 Research prior to the selection of topics (2005–2006)

The author (Oki) started investigation of recycling technology for waste products that contain rare metals from the end of 2005. In a survey project of Japan Oil, Gas and Metals National Corporation (JOGMEC), advanced recycling method was investigated for small fuel cells, automobile catalysts, neodymium magnets, and fluorescent lamps. At that time in the recycling plants, automation technology for cutting the two ends of straight fluorescent lamps, recovering glass and aluminum, and then processing the waste fluorescent powder including mercury had been established. The transition from incandescent light bulbs to compact fluorescent lamps was occurring in Japan at the time, and it was necessary to establish a recycling method. Therefore, in the succeeding project, selective crushing and separation methods for fluorescent powder of compact fluorescent lamps were studied. However, compared to straight tubes, there were numerous types of compact or bulb type fluorescent lamps, and there was no clear idea for developing a comprehensive processing technology.

On the other hand, a researcher at AIST was looking for application of magnetic levitation in the bioscience field, and was referred to the authors (Akai and Yamashita) to see whether it could be used in this research. At the time, we were conducting glass research, and it seemed that using magnetic levitation by superconductive magnets (equivalent to a suspension magnetic separator by the wet method) for recycling waste glass would not be cost effective in terms of resource value. Therefore, we suggested using this technology for waste fluorescent powder because it was more valuable, and started joint investigation. At the same time, the Rare Metal Task Force at AIST was working on a method for recovering terbium from LAP by a vitrification method,^[2] but this also seemed to be difficult cost wise. An engineer at a major lamp company told us that there was a social demand for technology to recover reusable LAP from waste fluorescent powder since LAP deteriorated very little.

2.2 Identification of topics and start of R&D (2007–2011)

In 2007, the authors (Akai and Yamashita) conducted fluorescent powder separation tests by magnetic levitation, and confirmed that LAP could be levitated selectively by adding an appropriate surfactant. Magnetic levitation

tests for various fluorescent powders were conducted, and a separation method of fluorescent powder by magnetic levitation^[3] was established. At this time, the price of rare earths started to increase, and the lamp manufacturers became highly interested in this technology, and we started seriously preparing for a project proposal. During this preparation stage, there was a consultation with an author (Oki) who specialized in sorting technology and had been studying recycling technology of fluorescent lamps. Oki responded that realizability was higher for a high-gradient magnetic separator rather than magnetic levitation, and therefore, steps were taken towards the development of a high-gradient magnetic separator.

In 2009, this research was selected as the “Rare Metal Substitute Materials Development Project” of the New Energy and Industrial Technology Development Organization (NEDO), and the development of recycling technology using high-gradient magnetic separation started as part of this project. In the first three years, Akai and Yamashita conducted basic tests for model samples using a conventional batch type high-gradient magnetic separator, and established the basics of a LAP recovery method using the separator.^[4] A press release on this research was issued in May 2011. This was right in the middle of the rare earth crisis (the peak price was in October 2011), and we received numerous inquiries from the lamp manufacturers. While there were many voices pushing practical realization, the construction of a system contiguous to actual waste fluorescent powder processing became an immediate topic, and this led to the development of a device in the later NEDO project (2012–2013).

3 Topics in development of a separator

3.1 Mechanism of a high-gradient magnetic separator

Responding to the request of the NEDO Project under which basic research had already started, the author (Oki) joined in the development of the device. First, the principle of a high-gradient magnetic separator will be explained. The strength of magnetic trapping in the magnetic separator is generally expressed by the magnetic flux density (B) (unit, T: tesla; 1 T = 10,000 gauss) of the magnet surface. The surface magnetic flux density of a neodymium magnet is about 0.35 T, and about 1 T can be achieved partially when the magnetic circuit is established. Since electromagnets demagnetize at high temperature, it is normally about 2 T. On the other hand, a superconductive magnet can achieve magnetic flux density of over 10 T. Many of the magnetic separators are open systems, and the magnetic flux density declines inversely to the square of the distance. While it is not described as a specification, since the magnetic gradient (difference between magnetic flux density between two points) cannot be controlled, the “magnetic force” that actually attracts the particles is the product ($B \cdot \Delta B$) of magnetic flux density (B) and a magnetic gradient (ΔB) (Fig. 1 top).

On the other hand, the attraction to magnets on the particle side is expressed as the magnetic susceptibility (dimensionless number) per unit volume. This is an index that expresses the property of a substance, but the attraction to magnets is also dependent on the particle volume, and the particle is more easily attracted to magnets as it increases in size. In the case of separating a mixture of various substances as in mines or recycling, it is extremely difficult to accurately separate fine particles of 50 μm or smaller according to properties (specific gravity, magnetic force, etc.) of the particle bulk. When sorting such fine particle mixtures, slurry that is particles dispersed in water is used to simplify the transportation and separation. However, water viscosity or the property of the particle surface becomes dominant in fine particles, and it becomes difficult to generate differences in particle motion according to particle bulk properties. However, in an earlier study by the authors (Akai and Yamashita), it was confirmed in the lab test that highly precise LAP recovery was possible using magnetic levitation by superconductive magnets, from the fluorescent powder mixture with particle diameter of about 5 μm that did not have strong magnetic force. This magnetic levitation is the same mechanism as the suspension magnetic separator, and is an open system. That means, with magnetic force equivalent to about 10 T in an open system, 5 μm of LAP can be selectively recovered. To construct a practical system at low cost, it is necessary to produce the same magnetic force at normal

conductivity. In theory, if a magnetic gradient of about five times the open system is produced, it is possible to produce a magnetic force equivalent to 10 T in an open system, and the only method that can achieve this is a high-gradient magnetic separator.

Even if a magnetic body is placed in a position where 1 T parallel magnetic flux is produced when the N and S poles are set against each other, the particles do not move even in this extremely strong magnetic field. That is because there is no magnetic gradient (Fig. 1 bottom). Conversely, if a high magnetic gradient is produced, a strong magnetic force is produced even if the magnetic flux density is low. Although such an interpretation existed for a long time, H. H. Kolin *et al.* of the Massachusetts Institute of Technology proposed a mechanism for a high-gradient magnetic separator that actually realizes the phenomenon in 1968. This was the earliest case in which a separation mechanism itself was developed, and in recent years many physical separators have been developed. Kolin *et al.* succeeded in generating an extremely high magnetic gradient around thin wire by arranging the matrix of highly permeable thin wire in the parallel flux to concentrate the magnetic flux. This technology was realized as a de-ironing method (removal of colored minerals) of kaolin that is used in white pigment. In the 1980s, it was also applied to desulfurization/separation of coal (removal of sulfide iron

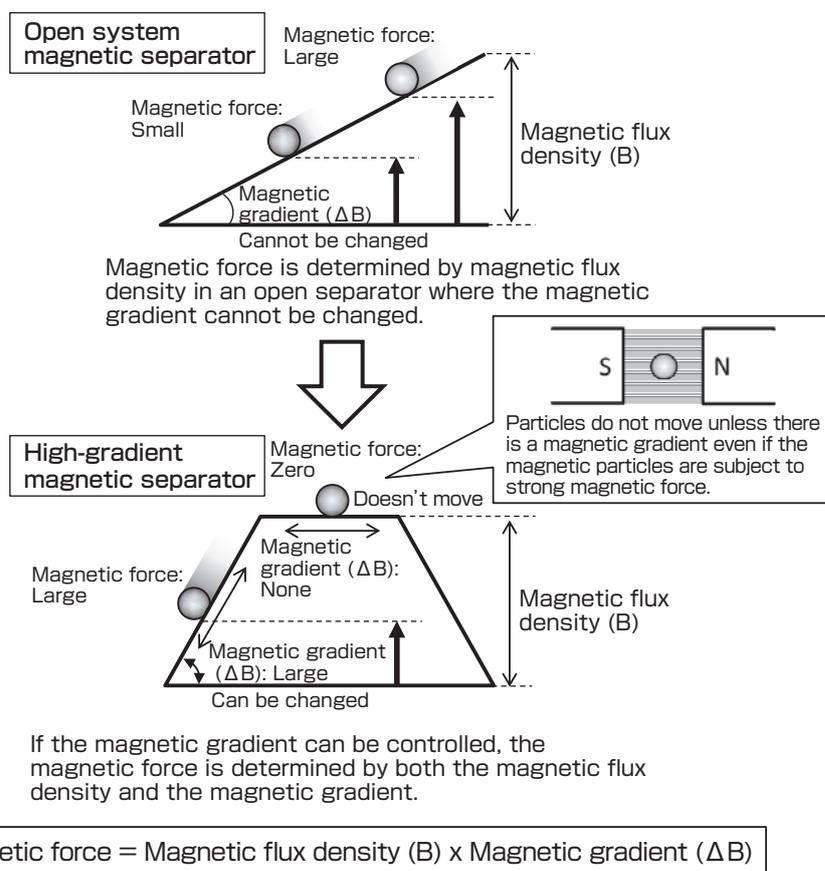


Fig. 1 Image of magnetic force in open system magnetic separator and high-gradient magnetic separator

minerals). Until then, the floatation method using particle surface properties was mainly used for sorting of particles 50 μm or less. With the development of this technology, the road to fine particle sorting was opened for sorting technology that utilized the particle bulk properties.

3.2 Topics of fluorescent powder sorting and development goals

Although a high-gradient magnetic separator has potential of sorting fluorescent powder at normal conductivity, there were two issues in developing the actual separator. The first issue is that this separator can provide a strong magnetic gradient but it cannot be precisely controlled. The expanded metal (the metal plates in which cuts are made and pulled to form diagonal nets) are used as a matrix, but the magnetic gradient is fixed according to magnetic flux density by magnetic permeability, thickness, special distribution between the thin wire, etc. of the material. That is, it is not possible to have control such as achieving desired magnetic flux density by voltage. Moreover, because there is unevenness of the magnetic force within the space and the matrix contracts due to excitation, it is impossible to accurately predict the unevenness. If the matrix is in a rough condition, the magnetic force the particle receives may differ depending on its travel route. Therefore, slurry is run in a dense matrix condition, all particles are attached to the matrix, and nonmagnetic particles are washed off in an excited condition. However, the engulfment of particles is not resolved and highly precise sorting is difficult. The second issue is the size of the device. Since the device was developed for use at mines, the continuously operating commercial separator is a large device, while the bench size device is a batch type lab

device. Considering the volume of fluorescent powder to be processed, a lab device is sufficient in terms of size, but it was necessary to newly develop a continuously operating device based on the lab device.

De-ironing of kaolin and desulfurization of coal are for the removal of impurities (magnetic recovery of substances to be removed), and the objective is to recover fine particles of iron (sulfide iron) as much as possible, by generating a strong magnetic force. There is no problem if some kaolin or coal was removed together with the impurities, and the objective is achieved even if the sorting selectivity is not that high. On the other hand, for fluorescent powder sorting, the idea is to selectively recover (magnetic recovery of valuable substances) LAP as a valuable substance. It was necessary to set a sorting limit between the slight difference of magnetic permeability of LAP and other particles, and the mission of the NEDO Project assigned to the author (Oki) was to achieve a prototype of a small continuously operating device in two years.

4 Automatic continuous operation of a small high-gradient magnetic separator

4.1 Outline of the automatic continuous operation system and comparison to the batch process

Based on the commercial batch type, a small high-gradient magnetic separator (a Jones wet strong magnetic separator, maximum 1 T), we aimed for the development of a magnetic separation system that could recover LAP at high precision, while allowing automatic continuous operation. Figure 2 shows the batch process and the newly developed automatic

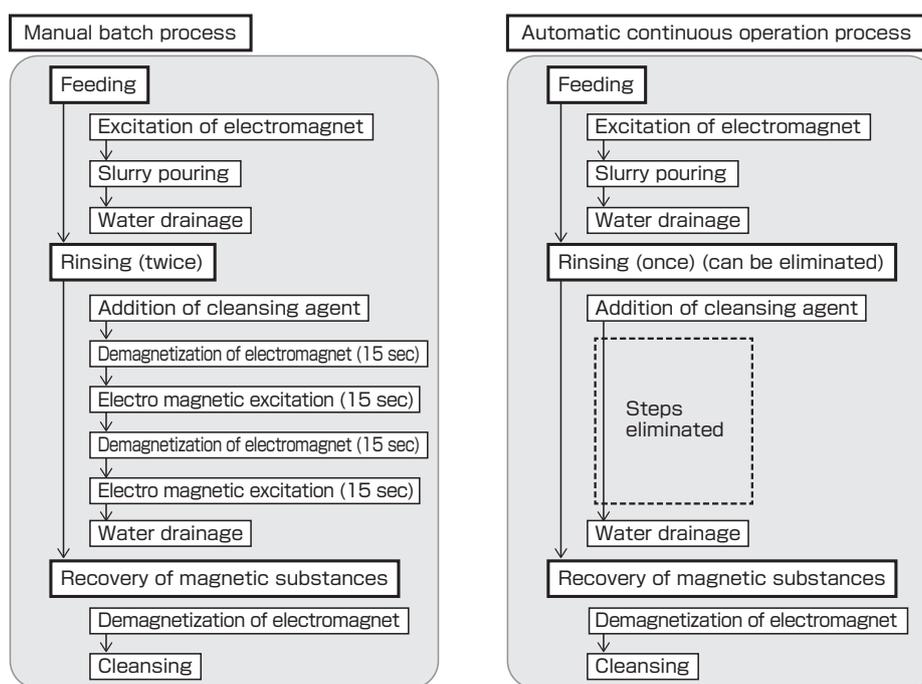


Fig. 2 Flow of conventional batch process and newly developed automatic continuous operation

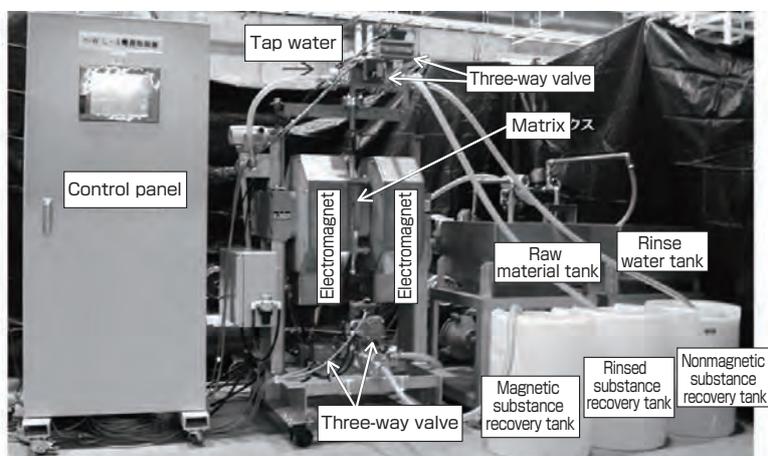
Table 1. Results of preliminary sorting test for batch device and automatic continuous device

	Recovery rate [Distribution ratio (%)]				Purity [Grade (%)]						
	Magnetic substance	Rinsed substance	Nonmagnetic substance	Total		Before magnetic separation	Magnetic substance	Rinsed substance	Nonmagnetic substance		
Manual batch test	Overall (yield)	18.0	20.5	61.5	100					As pretreatment, the following three processes are conducted: (1) Sieving at 38 μm opening (recover undersized material) (2) Magnetic separation to remove iron (0.05 T magnetic separation, nonmagnetic substance recovery) (3) Sedimentation and cleansing (remove upper layer) three times	
	LAP	86.4	4.8	8.7	100	LAP	12.0	60.7	3.0		1.8
	BAM+CAT	23.6	17.9	58.5	100	BAM+CAT	4.0	5.1	3.4		3.7
	SCA	4.9	18.2	76.9	100	SCA	6.5	1.9	6.2		8.7
	YOX	9.5	21.2	69.3	100	YOX	13.6	8.0	15.7		17.1
	Halo-phosphate	7.2	24.7	68.1	100	Halo-phosphate	60.7	23.4	70.9		64.9
	Glass	5.5	6.2	88.4	100	Glass	3.2	0.8	0.8		3.8
Automatic continuous operation test	Overall (yield)	17.9	0.0	82.1	100					As pretreatment, only one process is conducted: (1) Sieving at 20 μm opening (recover undersized material)	
	LAP	79.8	0.0	20.2	100	LAP	13.9	62.9	0.0		3.5
	BAM+CAT	27.7	0.0	72.3	100	BAM+CAT	7.3	9.7	0.0		5.5
	SCA	4.9	0.0	95.1	100	SCA	4.7	1.5	0.0		6.4
	YOX	7.0	0.0	93.0	100	YOX	16.9	7.7	0.0		22.3
	Halo-phosphate	5.8	0.0	94.2	100	Halo-phosphate	54.7	17.0	0.0		60.0
	Glass	6.4	0.0	93.6	100	Glass	2.3	0.7	0.0		2.2
	Iron oxide	66.4	0.0	33.6	100	Iron oxide	0.2	0.5	0.0		0.1

Expanded metal is used as matrix for both tests.
 ※LAP (LaPO₄: Ce,Tb), BAM (BaMgAl₁₀O₁₇: Eu²⁺), CAT (CeMgAl₁₁O₁₉: Tb³⁺), SCA ((SrBaCa)₁₀(PO₄)₆Cl₁₂: Eu), YOX (Y₂O₃: Eu³⁺)

continuous operation flow. There are several points that differ from the batch type, but the main difference is that in the batch type, the nonmagnetic substances are released in the water phase through repeated demagnetization and magnetization, while in the continuous system, the nonmagnetic substances are washed away with water only. Later, it was found that a rinsing effect appeared when the slurry flow rate was optimized, and sufficient sorting precision could be maintained even when the rinsing process was entirely eliminated. To verify this system, three sets of automatic, quantitative water supply and drainage systems and an automatic continuously operating high-gradient magnetic separator that incorporated those systems were fabricated (Fig. 3).^[5] To optimize the system, first, preliminary investigation was conducted to see whether the continuous system would have the sorting precision equivalent to the batch system. In the batch system, three processes were

conducted as pretreatment: (1) sieving at 38 μm opening, (2) magnetic separation to remove iron, and (3) precipitation and cleansing three times. On the other hand, in the automatic continuous system, it was found that major simplification of pretreatment could be done by precise control of the supply flow rate and time and sieving at 20 μm opening. Table 1 shows the results of the batch system and the automatic continuous system using the expanded metal. Even when the processes of (2) and (3) were eliminated, in the automatic continuous system, the residues of iron, binding agent, and alumina powder were very small. Comparing the sorting results of the two systems, the recovery was 6.6 % lower in the continuous system while the LAP purity (grade quality) was 2.2 % higher in the continuous system, and it was confirmed that approximately the same sorting performance could be obtained as the batch system, even in the continuous system from which most of the pretreatment processes were


Fig. 3 Automatic continuously operated high-gradient magnetic separator (prototype)

eliminated.

4.2 Control optimization using a conventional matrix (expanded metal)

A commercial expanded metal matrix is shown in Fig. 4. The stacked expanded metal is inserted in the column so the slurry flow is interrupted somewhere within the matrix. The particles collide with the thin wire without exception, and the magnetic substances are captured. The nonmagnetic substances will attach to the matrix on the thin wire or become sandwiched between the magnetic substances, but are sorted after being washed by slurry or rinsing water. That is, whether the particles are recovered as “magnetic substances” after accumulating in the matrix or are recovered as “nonmagnetic substances” after falling off is determined by the relationship between the magnetic force and the flow rate of water. However, since it was initially developed as a device to remove magnetic impurities, there was not much consideration for the rinse flow rate control to improve the purity of magnetic substances, and this function was not incorporated in the commercial devices. In order to appropriately separate the particles with slightly different magnetic susceptibility into magnetic and nonmagnetic substances, it is necessary to control both the magnetic flux density and the fluid rate and time. Due to intellectual property concern, we shall omit the details of the investigative process, but after repeating various tests, we conducted a test to determine the optimal magnetic substance volume in one cycle. Figure 5 shows the magnetic substance volume of the matrix in one magnetic separation cycle, and the separation efficiency of LAP and non-LAP substances. The separation efficiency reaches maximum at around 50 g of magnetic substance volume. The magnetic force

distribution of the matrix is extremely uneven, and initially, non-LAP substances attach to the sites where the magnetic force is extremely strong, and the selectivity somewhat decreases. Later, when a certain amount is attached, magnetic attachment occurs at the site where the magnetic force is even, and the selectivity improves. However, as the magnetic substance volume increases, the magnetic force decreases as the magnetic and nonmagnetic substances are stacked onto the matrix, and the desired LAP substances are no longer magnetically attracted and the separation efficiency decreases again.

From the above results, the amount of magnetic substances attached to the expanded metal per cycle used in this test was determined to be about 50 g. The results of the systematic sorting test when the magnetic attached volume is set at 50 g are summarized in Fig. 6. In the first magnetic separation (rough sorting), the LAP purity increases from 16.5 % to 61.9 %. The LAP recovery is 79.4 %, followed by BAM + CAT 27.3 % and iron oxide 15.2 %. Other components are all 8 % or less, and at this point 92–95 % are removed as nonmagnetic substances. When the magnetic separation (fine sorting) is applied again to magnetic substances, the total LAP recovery decreases to 64.2 %, but the LAP purity increases to 82.0 %. When this is repeated again (fine re-sorting), the total LAP recovery decreases to 56.4 %, but the LAP purity increases to 85.9 %, and the removal rate of SCA, YOX, and halo-phosphate fluorescent powders reaches 99 % or higher. From the above results, it is determined that the appropriate condition when LAP recovery is prioritized should be “magnetic separation conducted once,” and when LAP purity is prioritized, “magnetic separation conducted twice,” considering the decrease in recovery. These were

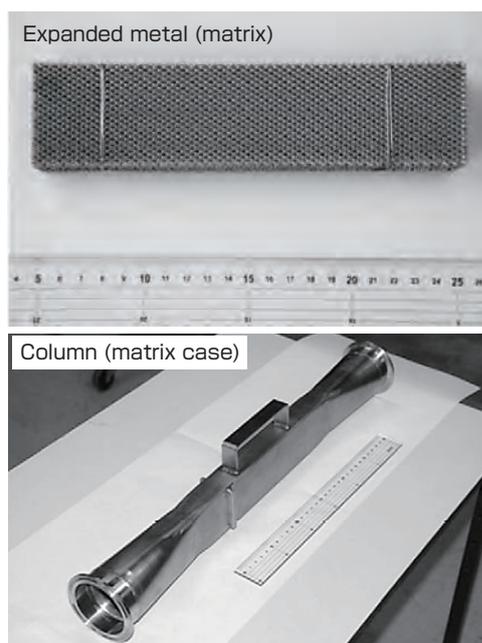


Fig. 4 Expanded metal (matrix) and column (matrix case)

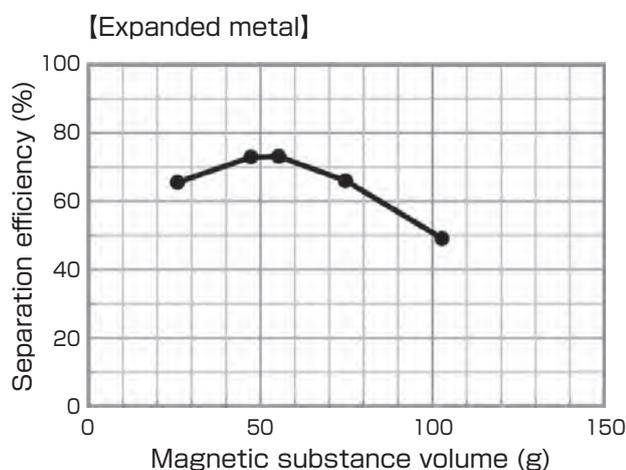


Fig. 5 Magnetic substance volume and selectivity of LAP (expanded metal used)

incorporated in the program of the automatic system.

5 Development of a high-selectivity matrix

5.1 Outline of a high-selectivity matrix

In the previous chapter, we developed an automatic continuous operation system using a traditional matrix to bring out its maximum performance, but we also developed a matrix with high selectivity to prepare for cases in which there is not enough sorting precision even when the developed system is used. In general usage of magnetic separation, the magnetic susceptibility difference between magnetic and nonmagnetic substances is extremely high, and in practice, it becomes the separation of magnetic and nonmagnetic

particles (Fig. 7). Even in a high-gradient magnetic separator, the emphasis is on the removal of magnetic fine particles using strong magnetic force, and the unevenness of the magnetic force within the matrix is not considered. On the other hand, although LAP has the highest magnetic susceptibility among fluorescent powders, it is a weak magnetic body, and the difference in magnetic susceptibility from other fluorescent powders is small. Therefore, to improve the selectivity, we started to develop a matrix with a new way of thinking. In the matrix developed in this research,^[6] the particles do not mechanically collide as in the conventional matrix, but even magnetic force is produced in the flow channel, and when the particles pass through the channel without barriers, only the particles with certain

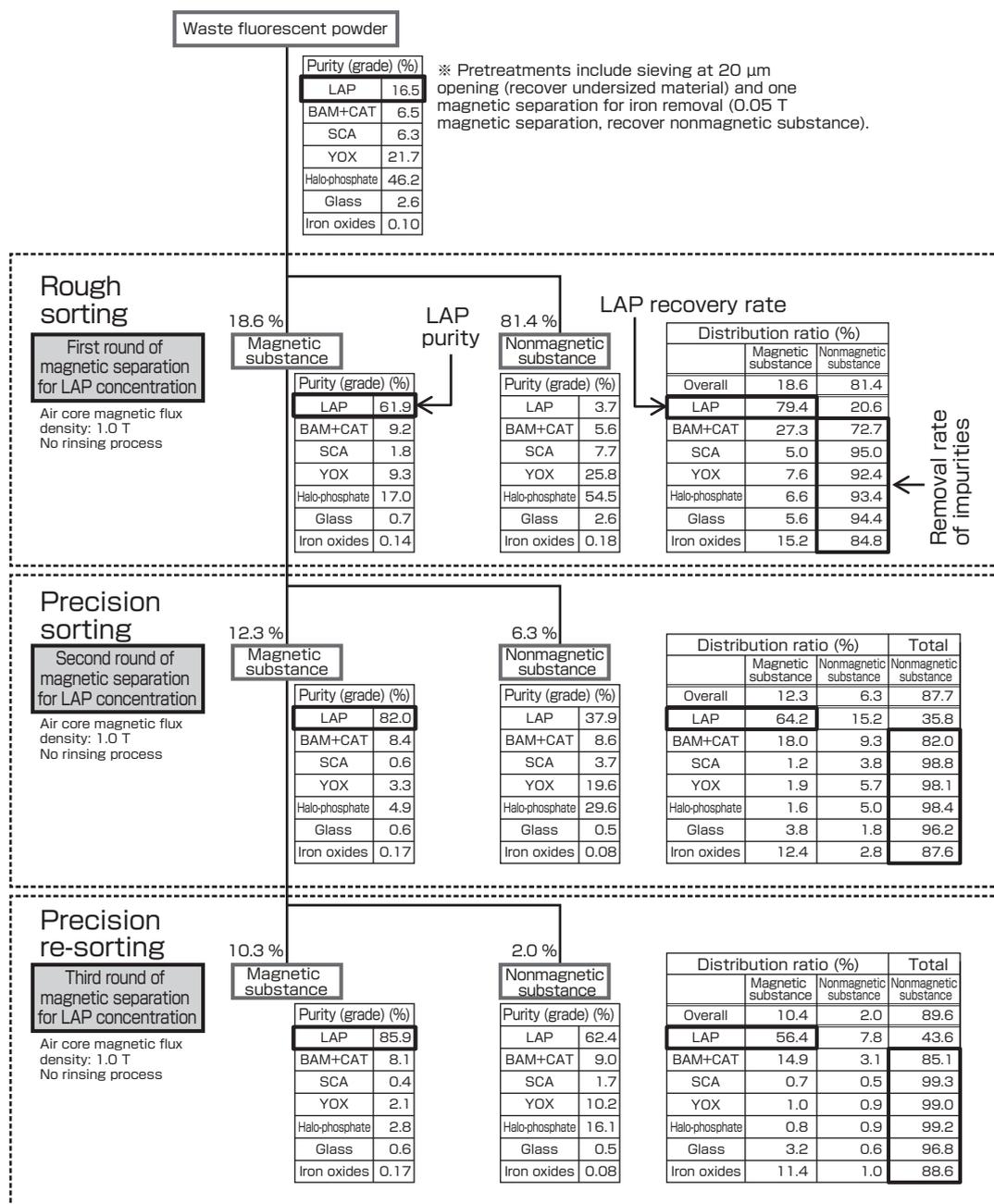


Fig. 6 Results of magnetic separation to obtain highly pure LAP using expanded metal

magnetic susceptibility are attracted and attach to the matrix by magnetic force. Therefore, the particles with magnetic susceptibility less than a certain threshold pass through the matrix, and it is expected that highly selective separation can be achieved. The developed matrix (Fig. 8) is designed by optimizing the internal magnetic force distribution through magnetic field simulation by a finite element method. It has a structure in which the waveform magnetic body walls of high rigidity and with 1 mm or lower height difference are placed facing each other, and regular and almost even magnetic force can be generated in the matrix space. It is also designed so the magnetic force strengthens only near the wall surface, to ensure the magnetically attached particles will not detach. As an example of the calculation results, the magnetic flux density (B) distribution around the matrix under device magnetic flux density 0.9 T, and the magnetic force ($B \cdot \Delta B$) distribution within the matrix are shown in Fig. 9. During the NEDO Project there was regular unevenness in the magnetic force distribution, but in the developed latest matrix, we succeeded in achieving almost even magnetic force, and now,

an extremely precise sorting threshold setting is possible.

5.2 Sorting performance of new high-selectivity matrix

To understand the selectivity property of the new matrix, we investigated the optimal magnetic substance volume of one cycle while setting the condition to be the same as the expanded metal of the previous chapter. The magnetic substance volume of the new matrix and the separation efficiency of LAP and non-LAP substances are shown in Fig. 10. Compared to the expanded metal, there is less contact opportunity with the particles, so the separation efficiency when the magnetic substance volume is low is a slightly low value. However, the separation efficiency hardly decreases even when the attachment volume reaches 50 g or more, and maintains 70 % even around 180 g. This is thought to be because constant long-range magnetic force occurs where the particles are attracted to the wall surface no matter where the particles are in the matrix space, and also LAP does not detach even when the magnetic substance forms a layer on the wall due to the strong magnetic force near the

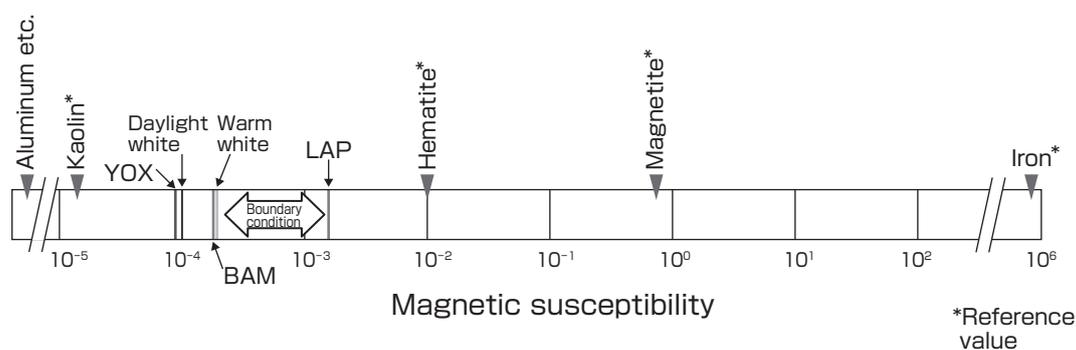


Fig. 7 Magnetic susceptibility of various substances and sorting boundary conditions for this research

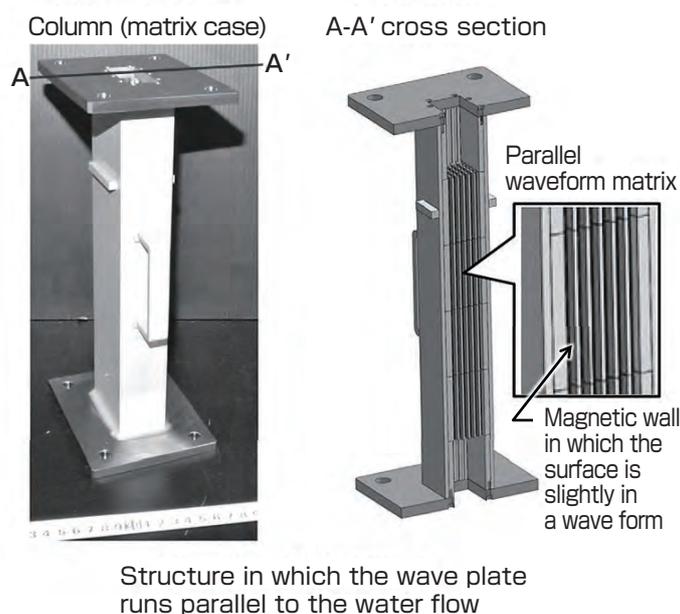


Fig. 8 Newly developed high-selectivity matrix

wall surface. Table 2 compares the selectivity results of the conventional expanded metal and the new matrix. From the results of the preliminary test, three cases of conditions prioritizing recovery and purity are given for both matrices. Although there are no great differences between the recovery and separation efficiency of the two, for the LAP purity and LAP concentration ratio, the new matrix shows about 10–30 % better results for both values.

On the other hand, there is a weakness, in principle, in this matrix. Conventionally, fluid transports the particles, but in the new matrix, the particle itself must reach the matrix by its own motion. Therefore, with finer particles with slow particle velocity, the time required for one cycle increases. While there is hardly any effect on particles of several ten μm or more that is the normal target of magnetic separation, waste fluorescent powder has extremely small particle size

of about $5\ \mu\text{m}$, and it is necessary to set the slurry flow rate to about 1/20 of the expanded metal. However, with the new matrix, there are two possibilities to supplement this. One is that the magnetic substance volume per cycle can be increased. As mentioned earlier, if the size is the same, about four times the amount of substances can be captured and recovered compared to the expanded metal. Second is that the generated magnetic flux can be distributed without loss in the matrix space, and the output of the electromagnet needed to capture LAP can be greatly reduced. As shown in Table 2, to obtain the same sorting results, the expanded metal requires 1.0 T (output 73 %), while the new matrix requires only 0.4 T (output 15 %). That is, if 1.0 T equivalent to the former is generated in the new matrix, a matrix with greater capacity can be utilized. The fluorescent powder processing volume per hour is 12.9 kg/h for the expanded metal, and 1.1 kg/h, or about 1/12, for the new matrix, and the same processing

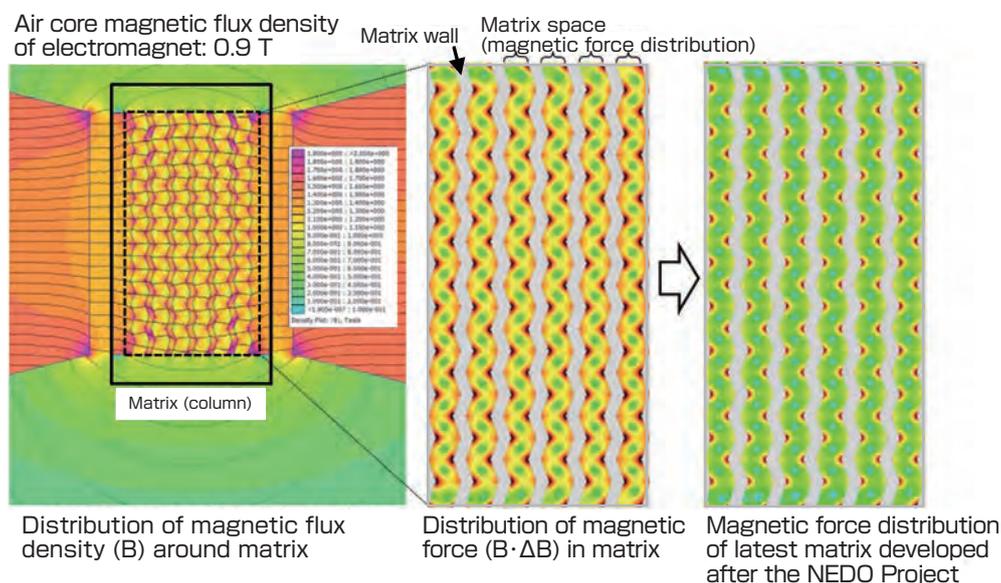


Fig. 9 An example of simulation result of magnetic flux density around high-selectivity matrix and magnetic force in the matrix

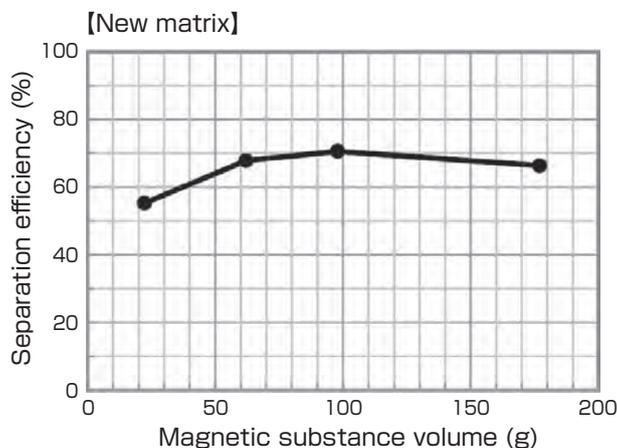


Fig. 10 Magnetic substance volume and selectivity of LAP (in new matrix)

Table 2. Results of high-purity magnetic separation for LAP using expanded metal and new matrix

Expanded metal					New matrix					
Processing condition	Air core magnetic flux density (T)	1.0			Air core magnetic flux density (T)	0.4				
	Feeding flow speed rate	1	1.2	1.45	Feeding flow speed rate	0.05				
	Rinsing flow speed rate	No rinsing			Rinsing flow speed rate	なし	1.0	2.9		
Yield (%)		20.6	17.9	16.3	Yield (%)		15.4	13.8	13.4	
LAP recovery rate (%)		84.2	79.8	78.8	LAP recovery rate (%)		83.1	79.7	73.0	
LAP separation efficiency (%)		73.1	72.0	72.1	LAP separation efficiency (%)		76.2	74.1	68.4	
LAP concentration ratio		4.1	4.5	4.8	LAP concentration ratio		5.4	5.8	5.5	
Purity (grade) (%)		Before separation	Substance after magnetic separation				Before separation	Substance after magnetic separation		
	LAP	13.9	53.0	62.8	64.2	LAP	13.9	60.2	64.5	70.1
	BAM+CAT	7.3	10.3	9.7	10.0	BAM+CAT	7.3	9.7	9.8	9.6
	SCA	4.7	1.8	1.5	1.4	SCA	4.7	1.6	1.5	1.1
	YOX	16.9	8.8	7.7	6.9	YOX	16.9	6.9	6.5	5.2
	Halo-phosphate	54.7	24.5	17.0	16.2	Halo-phosphate	54.7	19.5	15.9	12.3
	Glass	2.3	1.0	0.7	0.7	Glass	2.3	1.5	1.3	1.1
	Iron oxide	0.2	0.6	0.5	0.6	Iron oxide	0.2	0.6	0.6	0.6



Priority on recovery rate ← → Priority on grade



Priority on recovery rate ← → Priority on grade

LAP concentration ratio = LAP recovery rate/yield

volume can be obtained if the volume is increased 12 times.

The new matrix is still in development, but various characteristics and points that are superior to the conventional matrix have been demonstrated. Also, as mentioned earlier, development of higher precision type has been successfully created after the NEDO Project, and is being utilized in national projects for submarine hydrothermal deposits, copper ore de-arsination, and others. Compared to the conventional matrix, using the characteristic that allows sorting at very little magnetic susceptibility, it is expected to be used in various ways in the future.

6 Summary: Practical realization of the sorting system and the prospect for near-future resource circulation

In the years after the NEDO Project, automatic continuous operation of a small high-gradient magnetic separator was achieved by the author (Oki), and later, a license agreement was concluded with a magnetic separator company. During this time, the authors (Akai and Yamashita) conducted an investigation of the pre- and post-treatment processes. Also, a LAP lamp prototype test was done using waste fluorescent powder recovered as post-consumer products, and a result that luminous flux of 98.9 % could be achieved was obtained. There was no problem in the luminous flux maintenance rate, and it was found that LAP from which impurities were removed showed almost the same luminance as brand-new LAP, and was reusable. For this technology,

Nomura Kohsan Co., Ltd., which is the major company that engages in waste fluorescent lamp processing and which also engages in mercury processing in Japan, was selected for the NEDO grant in 2014, and the developed technology was transferred to the Itomuka Mine of the company. From the original batch system to the development of the continuous system, the overview of the fluorescent powder sorting system that was installed in February 2015 is shown in Fig. 11. Flow from the beginning of this research to practical realization is summarized in Fig. 12. During this time, the “Minamata Convention on Mercury” was adopted in October 2013 and was concluded in February 2016, and there was great expectation for the Nomura Kohsan’s ability to process mercury. On the other hand, in November 2015, the government announced a policy of promoting energy saving in lighting equipment, and the lighting equipment companies one after another announced termination of the production of fluorescent lamp lighting equipment. Although production of fluorescent lamp will be continued, it will be on a declining trend in Japan. The environment surrounding waste products is changing daily, and recycling technology must be able to adapt to the changes in society. Moreover, energy-saving properties of products should include possibilities for resource reuse and reduced energy consumed for disposal, as well as energy saving during manufacturing and usage. The age when a new product can be developed with an easy mind because recycling technology is established will arrive.

In this paper, the course of events was summarized from the

viewpoint of sorting device development, but some points should also be mentioned about how recycling is done. In general, there are two types of recycling. One is in-process recycling during manufacturing (of in-process waste), and the other is the post-consumer recycling (of post-consumer waste). In this research, both can be realized, but in general, the hurdle for the latter is extremely high compared to the former. As shown in Fig. 13, in-process recycling is the reuse of waste material that is process managed, and in most cases the purity is maintained to some degree. Although important as a production improvement process, these materials have

never been used in society as products and do not contribute to resource circulation. On the other hand, the latter starts from a condition where waste products from various sources are mixed, and it is difficult to reuse the resource from this stage. The latter can be roughly divided into cascade recycling and horizontal recycling by methods of resource use. The waste disposal that Japan has been working on for a long time is a process where the objective is to not make products into waste material, and does not focus on their reuse. Although an extremely high recycling rate has been achieved, most of the recycling is cascade recycling.

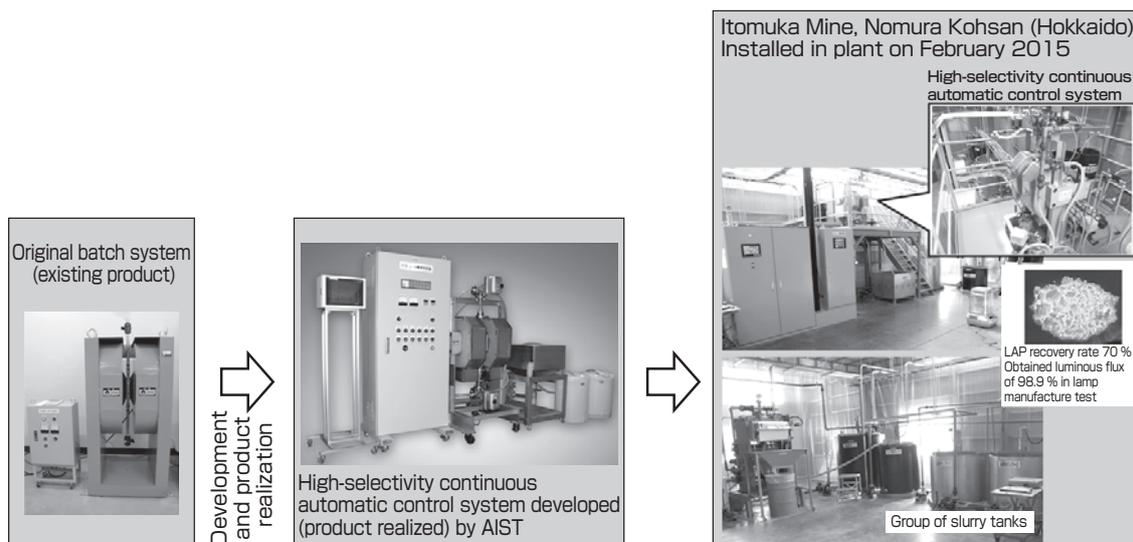


Fig. 11 Overview of the device from original batch system to fluorescent powder sorting system actually installed

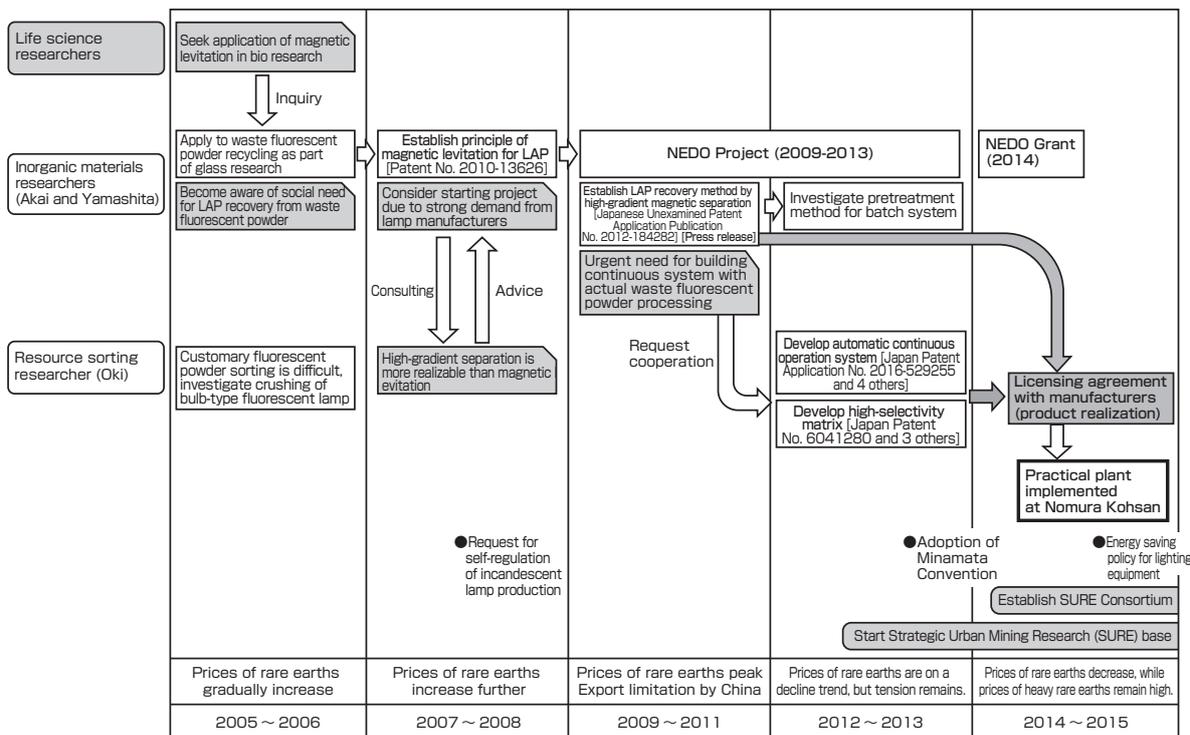


Fig. 12 Flow from beginning of research to practical realization

Particularly for metals, the main focus is on detoxification, and in many cases they are used as roadbed material and the values the metals possess is not really utilized. Even if cascade recycling increases, there is hardly any effect of reducing the import volume of natural metal resources, and in order to use waste products as urban mines, it is absolutely necessary to promote horizontal recycling. Fortunately, unlike organic materials, metal can be returned completely to original metal if it can be used as smelting material. However, metal excluding precious metals and copper does not have high value as raw material even if it undergoes troublesome recycling processes, and may not be feasible cost wise. The point that makes this research unique is that terbium metal is not recovered from the waste fluorescent powder but is made

into LAP, raw material for high-functional materials (Fig. 14). A sorting process that enables reuse of waste fluorescent powder is the first in the world, but more than that, a case in which metal circulation is established not for metal but for high-functional material with higher value in the inner loop of horizontal recycling is almost nonexistent in the world. It leads as a system that can be considered an ideal circulation system for the near future by European CE/RE policies.

After many twists and turns, the advanced recycling technology for waste fluorescent powder described in this paper was brought to practical realization as several favorable conditions came about. On the other hand, for the establishment of future urban mines, it is necessary to

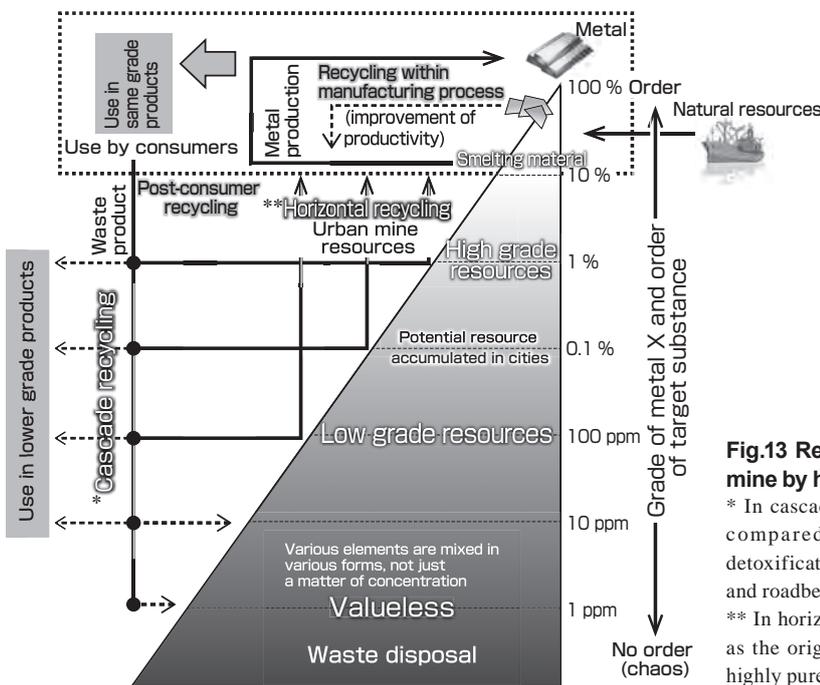


Fig.13 Resource potential and establishment of urban mine by horizontal recycling

* In cascade recycling, metal is reused in lower grade products compared to the original products. Its main purpose is detoxification, and the process includes mixing metal in cement and roadbed materials.

** In horizontal recycling, resources are reused at the same grade as the original products. In case of metals, it involves reuse as highly pure metal through refining of smelting material.

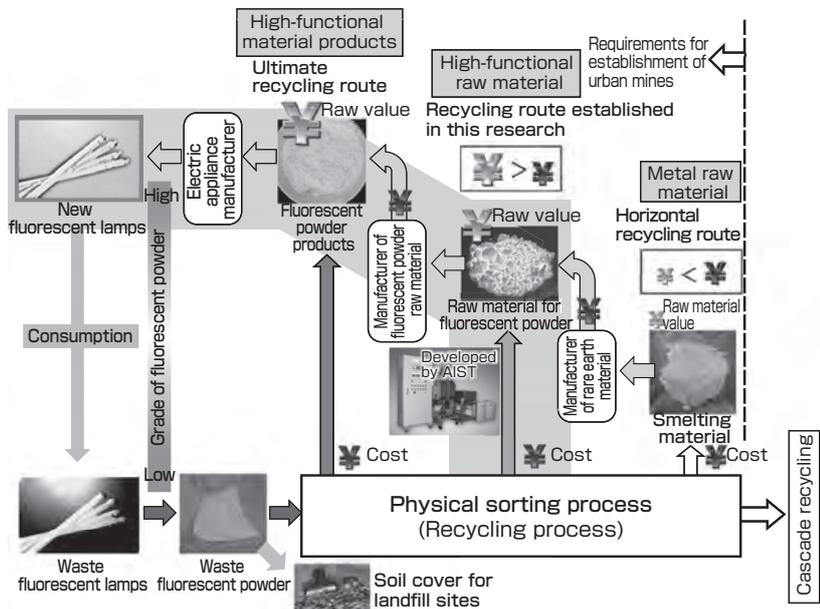


Fig. 14 Horizontal recycling and advanced material recycling loop

place as many waste products on horizontal recycling. The cost is a major issue, but for cost reduction of recycling, the social system must change into one with a “circulating” loop in which products follow the artery–vein like cycle where products are made from raw materials and distributed (like the artery that carries oxygenated blood from the heart), and then products after use are collected as waste (like the vein which carries deoxygenated blood back to the heart), and reused and recycled like through the “artery.” The authors established the Strategic Urban Mining Research (SURE) base at AIST in 2012, and created the SURE Consortium in 2013 with the collaboration of government and private companies. Currently, the members include 35 AIST researchers, 61 private companies, and 26 public research institutions, and they are engaging in activities for the construction of near future urban mines operated in a “circulating” loop. We hope the results of this research will provide hints as a leading system of near-future resource circulation allowing Japan to lead the world in urban mining development.

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Discussions with Reviewers

1 Overall

Comment (Hiroki Yotsumoto, AIST)

This research succeeded in developing a recycling technology of rare earth fluorescent powder by applying a high-gradient magnetic separator used in the resource engineering field. I think it is significant that the researchers of different fields cooperated, set a clear goal, advanced the research in steps, and developed a new recycling technology.

Comment (Chikao Kurimoto, AIST)

In modern society, recycling materials is one of the most important issues. This paper presents a success story on the

advanced integration of knowledge through the collaboration of researchers specializing in materials and those specializing in sorting devices, toward the practical realization of recycling for rare earth fluorescent powder. The structure of the paper is clear, with presentation of detailed figures and tables. It appropriately describes the scenario from R&D to practical realization, and also discusses future prospect. I think the paper fulfills the objective of *Synthesiology*, and is appropriate for publication in this journal.

2 Scenario

Comment (Hiroki Yotsumoto)

I understand that it is important that the researchers of different fields cooperate, but you seem to emphasize the cooperation of different divisions inside AIST. This may not be necessary information for the readers.

I rather think it will be more valuable and interesting to the readers if you present how common knowledge and way of thinking of life science researchers clashed or blended with those of inorganic materials researchers and resource technology researchers. Can you provide us such stories?

Answer (Tatsuya Oki)

I understood that this journal is different from usual journals where the results are presented matter-of-factly, and it asks for descriptions of researchers' roles, configuring of research, as well as the way of thinking and strategy that lead to a certain result. I revised the whole paper according to your comments.

On the other hand, pertaining to your latter indication, we did not seek optimal solution by integrating the opinions of all researchers. Rather, we passed on the issues that a researcher could not handle, in the order of life science → inorganic materials → resource technology, and the investigations for practical realization were fed back from resource technology → inorganic materials. Therefore, there weren't any clashes. I think integration occurs when a problem is not left to an individual to solve, and the roles are played by the right people in the right places.

Comment (Chikao Kurimoto)

To show continuation from the research strategy released earlier (Oki, *Synthesiology* 2013), I think the argument will become clearer if you discuss by citing Oki (2013) in "Chapter 1

Introduction."

Answer (Tatsuya Oki)

I added "At the time, the authors were conducting research and development of physical sorting of magnets and capacitors,^[1]"

Comment (Chikao Kurimoto)

Figure 12 is important as it shows the flow of the scenario of this paper. In relation to the comment above, why don't you add the establishment of SURE as a movement that took place in Japan (or AIST)?

Answer (Tatsuya Oki)

I added SURE and the SURE Consortium that are described in the text to the figure.

Comment (Hiroki Yotsumoto)

In the abstract, you use the expression, "there are few cases worldwide in which materials circulation from post-consumer waste has been established," but doesn't this contradict the fact that gold and platinum are recycled regularly? If you are saying the recycling described in this paper is of a different category from the recycling of precious metals, you must explain or else the readers will be confused.

Answer (Tatsuya Oki)

The recycling of copper and precious metals falls into the category of "horizontal recycling" in which the recycled substance is reused at the same value as the original "metal" (as ingots). This research handles the "case in which metal circulation is established in a loop further inside horizontal recycling" (refer to Fig. 14). That is, waste fluorescent powder is not reused as "rare earth metal," but we made it usable as "fluorescent powder raw material" that has higher value. In the sense that it is returned to make products again, it seems to be "horizontal recycling," but normally, the highest concept of recycling is to return to the original metal form. This time, recycling was done to create a valuable material that surpasses that stage, and I think this is a rare case worldwide.

The point that you indicated was changed to "a case in which metal circulation is established not for metal, but for high-functional material with higher value in the inner loop of horizontal recycling."