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Environmental Assessment of Recycling (EAoR) for Safe Recycling of Steelmaking Slag in the Republic of Korea: Applications, Leaching Test, and Toxicity

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Abstract: Steel slag is a major waste product of the steelmaking process. Owing to its chemical composition, it can be reused as a raw material in steel mills and a substitute for aggregates in engineering. In 2016, the Republic of Korea (ROK) introduced the Environmental Assessment of Recycling (EAoR) system for the safe recycling of waste products, which divides recycling methods into either matrix contact or non-matrix contact types; numerous challenges have been raised regarding weaknesses in the matrix contact type. Consequently, the standards for matrix contact recycling of slag (inorganic waste) have become stricter, and the recycling applications have also become more diverse. This study investigates the treatment, recycling, and management of steel slag in the ROK. In 2019, the annual generation of slag in the ROK exceeded 23.03 million tons with a recycling rate of 98.3%, showing a high level of recycling, compared with that of the United States (81.3%), the European Union (87%), Australia (100%), and Japan (98.4%). However, a comparative analysis of overseas slag recycling systems and recycling standards found that the recycling applications and recycling standards of slag in the ROK were somewhat stricter than those in the European Union and the United States. Furthermore, an investigation comparing the applications of recycled slag in different countries showed that the ROK could expand its applications of recycled slag. The limitation of the EAoR system was that the excluding products, environmentally certified from Korean Standards in the EAoR regulations, might be attributing to environmental pollution. The leaching and toxicity tests for recycling application of steelmaking slag were evaluated and showed no significant negative effects. Lastly, there is an urgent need for environmental standards relating to slag that can be recycled as a product, and the recycling and environmental label certification for products should also be amended. In addition, the applications of steelmaking slag were necessary to partially extend according to the hazardous characteristics.

Keywords: steelmaking slag; recycling; application; waste management; environmental assessment of recycling

1. Introduction

The production of global steel has increased from 2 million metric tons (Mt) in 1950 to 18.69 Mt in 2019 according to the World Steel Association [1] (Figure 1). China produces the largest quantity (53%), followed by the European Union (8.5%), India (6.0%), and Japan (5.3%) (Figure 2) [2]. In addition to consuming large amounts of raw materials and energy, the steel industry also generates large quantities of various by-products and waste types, through complex production processes such as raw material treatment, ironmaking, steelmaking, and rolling. Of these by-products, 27.68 Mt of slag were generated in 2019, which comprised 16.56 Mt of blast furnace slag and 11.12 Mt of steelmaking slag [3].

Figure 1. Global crude steel production from 1950 to 2019.

Crude steel production in

World total: 1869 million tons

Figure 2. Worldwide distribution of steel production (2019).

In the Republic of Korea (ROK), blast furnace slag, converter furnace slag, and electric arc furnace slag are recycled for various applications; however, as there is limited demand for cement raw materials, concrete admixtures, and silicate fertilizer raw materials with high proportions of added slag, there is a need to develop the applications of recycled slag.

Korean Standard (KS) regulations and guidelines are being implemented in relation to these issues, and studies are being conducted to increase utilization in areas beyond those where slag has traditionally been recycled. Such studies typically focus on using slag as an aggregate, as well as the use of not only blast furnace slag but also converter furnace and electric arc furnace slag. Aggregate demand in the ROK increased steadily from 201,424,000 m³ in 2014 to 231,770,000 m³ in 2018, and is forecast to continue to increase due to future housing construction [4]. In these social conditions, the value and demand for steel slag as an aggregate is also expected to be infinite. Hence, various studies, mainly conducted by research institutes and universities, have examined how to supply aggregate to meet this demand.

Steel slag has been utilized for a long time. Europe began using steel slag for phosphate fertilizers as early as 1880 [5]. Owing to its chemical composition, comprising calcium oxide (CaO), Fe, iron oxide (FeO), silicon dioxide (SiO₂), magnesium oxide (MgO), and manganese oxide (MnO), steel slag can be used in steel mills (in both sintering and converter processes) [6,7] and for wastewater treatment due to its porous structure and alkalinity [8,9]. Sarfo et al. [10] investigated a carbothermal reduction method capable of recycling valuable metals from steelmaking slag and using residual slag in the glass and ceramic industries. Ferreira et al. [11] and Pasetto et al. [12] confirmed that steel slag can be effectively used as aggregate for road and hydropower construction due to its advantageous physical and mechanical properties, including hardness, wear resistance, adhesion, roughness, and toughness. Previous studies have reported that since steel slag contains CaO and MgO, it can be reused to capture and store carbon dioxide (CO₂) [13,14]. Studies have reported that the components of steel slag which are beneficial for promoting plant growth, such as Si, Ca, Mg, P, and Fe, can be reused for fertilizer and improving soil [15,16].

Drissen et al. [17] reported that the greatest issue when internally reusing slag in steel mills is the enrichment of P and S. The amount of free CaO and MgO is an important factor to consider when using steel slag in construction, as it affects volume stability. Nakase et al. [18] reported that the total Fe content and the leaching potential of heavy elements such as As, Cr, and V are key factors influencing the use of steel slag in cement production. In particular, Riley et al. [19] monitored the soil and rivers around steel mills and nearby stockpiles for over 36 years and reported that toxic slag-derived V had leaked into the groundwater and nearby rivers, thereby causing environmental damage. To the best of our knowledge, no study has been conducted on the hazardous assessment of steelmaking slag for diverse recycling. The estimation for various recycling and application of steelmaking slag such as content, leaching test, and toxicity evaluation, were performed. To understand the leaching characteristics for various environmental conditions, the characteristics and toxicity according to the use of distilled water and seawater were identified.

Regarding waste recycling management policy in the ROK, the Environmental Assessment of Recycling (EAoR) system for the safe recycling of waste has been reflected in the Wastes Control Act since July 2016. It includes a method for managing 11 types of hazardous substances, distinguishing between existing specified waste and general waste, and designating recyclable types to identify the impacts of waste recycling on humans and the environment. Moreover, evaluation items and methods have been improved and expanded for new recycling types, or recycling types for which no recycling standard had been designated. The EAoR system is divided into matrix contact types and non-matrix contact types. The former corresponds to waste recycled as filling and cover materials which can affect soil, groundwater, and the atmosphere, while the latter corresponds to recycled products that do not directly or indirectly affect the soil or water environment.

Since the introduction of the EAoR system, environmental pollution and disputes related to matrix contact recycling have increased; one possible cause is the application of unified standards without the pre-inspection and confirmation of waste types. Environmentally certified products (EL: Eco-Label, GR: Good Recycled, KS: Korean Standards) such as filling material, cover material, and aggregates, which incorporate waste as specified in the Environmental Technology and Industry Support Act, can be used as recycled

products. However, some certified products do not satisfy the certification conditions that ensure environmental stability, and because matrix contact recycling sites that are less than 30,000 m² in area or produce 120,000 tons of material are excluded from the Wastes Control Act, there is no way to check for potential environmental pollution after recycling.

According to Article 25 of the Act on the Promotion of Saving and Recycling of Resources and the Guidelines for Recycling Steel Slag and Coal Ash Discharging Business in the provisions of Article 35 under the same act (Ministry of Environment Notice No. 2009-285, Ministry of Knowledge Economy Notice No. 2009-326, 31 December 2009), the efficient use of resources relies on recycling of by-products generated in the production process. Relevant standards, design and construction guidelines, and target rates for recycling steel slag and coal ash (in relation to Articles 9 and 11) are said to enable recycling of these materials for various applications [20]. Such details include slag aging methods, categorized according to the application of recycled slag, and Korean industrial standards categorized by usage [20]. Accordingly, approximately 100% of steel slag is currently recycled as a recycled raw material, thus playing a major role in conserving resources and preserving the environment. Owing to the recent completion and operation of additional steel mills in the Republic of Korea, steel slag generation is forecasted to increase by more than 6 Mt by 2025. Therefore, there is an urgent need to expand recycling applications and methods in line with this rapid increase in steel slag generation.

This study investigates the factors contributing to steel slag treatment, recycling, and management in the ROK. First, a general overview of steel slag is provided, and the chemical composition and utilization of steel slag in several countries are described. A historical overview of steel slag management and usage in the ROK is provided, and the laws, regulations, policies, and standards related to steel slag are explained. This study describes challenges that the ROK faces in terms of steel slag recycling. Finally, to promote the rational recycling of resources, specifically, by-products discharged by industrial processes, this study proposes a plan to improve the designated by-product (steel slag) system and promote recycling.

2. Materials and Methods

2.1. Materials and Samples

Deionized (DI) water was used from a Super-Genie E water purification stations (Rephile Bioscience Ltd., Philadelphia, Pennsylvania, USA). The pH was measured using a pH meter (Orion star A215, Thermo Fisher SCIENTIFIC, Waltham, MA, USA). The chemical composition of steel making slag was identified using X-ray fluorescence spectrometry (Panalytical Epsilon 3). The measurement of effective grain size was conducted using particle size analyzer (Laser scattering particle size distribution analyzer—LA960, HORIBA, Japan). The analysis of chemical composition was conducted using X-ray fluorescence (XRF) (Epsilon 4, Malvern Panalytical Ltd, United Kingdom). The sample of steel making slag was collected from 16 steel companies in ROK. The sample was transported to the laboratory and stored at 4 °C.

2.2. Experimental Procedures

2.2.1. Leaching Test

For the analysis of leaching characteristics, the method of leaching test in Waste Process Test Criteria (ES 06150.C) was used in this study. According to ES 06150.C, an eluate was prepared by shaking for 6 h at a high liquid ratio of 1:10 according to the dissolution test method, and then acid treated according to the conditions of the sample. The pretreated eluate was assessed for heavy metals using ICP-OES.

2.2.2. Toxicity Test Using *Aliivibrio fischeri*

The luminescent bacteria toxicity test was applied according to the acute toxicity test method using luminescent bacteria of the “Water Pollution Process Test Standards (ES 04706.1)” in the ROK, to evaluate the luminescence inhibition by exposing freeze-dried

products or luminescent bacteria in culture (*Aliivibrio fischeri*) to control. After the sample was exposed for 30 min, the luminescent was calculated using Equation (1). The sample was analyzed in quadruplicate.

$$f_{kt} = I_{kt} / I_0 \quad (t = 30 \text{ min}) \quad (1)$$

where f_{kt} is a correction factor for 30 min exposure; I_{kt} is a luminous intensity in the control sample after 30 min exposure (Relative luminous unit); I_0 is a luminous intensity of the control test suspension immediately before the addition of the diluent (relative luminous unit).

The results were reported as the luminescent inhibition value, $1 - L_t/L_c$, where L_t is the luminescent intensity of the sample and L_c is the luminescent intensity of *Aliivibrio fischeri* with 3% sodium chloride.

The pretreatment was conducted following this procedure. The salinity of the sample was adjusted into 20 psu. The statistical analysis of the luminescent bacteria toxicity was conducted using the ToxCalc 5.0 program (Tidepool Scientific Software, McKinleyville, CA, USA).

3. Results and Discussion

3.1. Historic Trends in the Republic of Korea

Figure 3a shows the production of steel slag in the ROK over the last 10 years, classified into blast furnace slag and steelmaking slag. The domestic steel industry demonstrated stable growth in the 2010s; however, from the late 2010s, it entered an overall stagnation phase amid the sluggish global steel industry (Figure 3b). After the 2008 financial crisis, the strong growth of the Chinese economy fueled steel demand, which has shown steady growth since the early 2010s. From the late 2010s, both demand and production entered a stagnation phase because of the slowing growth of the Chinese economy, and the growth trend in the ROK has therefore slowed.

The steel industry is facing mounting difficulties, such as reduced domestic demand and exports due to the recent COVID-19 pandemic, lower sales, and profitability due to sluggish upstream industries, and a blow to exports caused by the spread of global protectionism. In the first half of 2020, domestic demand and exports of steel products decreased by 8.7% and 7.6% year-on-year, respectively. In the first half of 2020, domestic automobile production decreased by 19.8% and orders from shipbuilders decreased by 68.6% year-on-year. Consequently, the sales and operating performance of major steelmakers has decreased. Export difficulties are becoming more severe due to growing protectionism in major countries such as the United States.

Inorganic waste in the ROK is classified into 13 types and separately classified into 10 recycling types (R-1 to R-10), listed in Table 1. Figure 4 shows the production of the 13 types of inorganic waste and the amount of R-7 recycled in the ROK in 2019. A total of 59.93 Mt of the 13 types of inorganic waste were generated, of which slag (23.03 Mt), inorganic sludge (12.17 Mt), and combustion residue (11.13 Mt) comprised 77%. Figure 5 shows the amount of slag recycled by R-type. R-3, which can be made into a recoverable state or used for recycling, comprises 70.1% (1583.5 Mt); R-7, which is recycled as filling and cover material, road base material, banking, etc., in soil and public waters, accounted for 22.7% (513.7 Mt); while R-4, which is directly recovered, comprised 6.4% of the total recycled slag (143.9 Mt).

Figure 3. The production trend of (a) steel slag and (b) the trend of domestic demand and export in the Republic of Korea.

Table 1. Recycling types for inorganic waste in the Republic of Korea.

Type	Regulation and Standard
R-1	Original form or simply repair and reuse
R-2	Simply repair or wash and reuse
R-3	Can be made into a recoverable state or used for recycling
R-4	Directly recoverable
R-5	Recycling for agriculture or soil improvement
R-6	Recycling of organic materials to improve soil quality
R-7	Recycling as embankment materials, cover material, road layer material, and filling material in soil or public water
R-8	Directly recovered as energy, or processed to be recoverable
R-9	Can be recovered as energy
R-10	A type of intermediate processed waste for product manufacturing

Figure 4. The production and recycling of inorganic waste in the Republic of Korea (2019).

Figure 5. The amount of recycled slag, by R-type, in the Republic of Korea (2019).

Figure 6 shows the changes in usage of recycled for blast furnace and steelmaking slag over 10 years in the ROK. The usage categories are categorized into cement, road, banking, civil construction, brick/concrete, fertilizer, process reuse, and others. The blast furnace slag recycled as raw materials for cement showed the greatest change in usage, increasing from 60% in 2010 to 80% in 2019 (Figure 6a). From 2010, the amount of blast furnace slag that was used for road construction (21%) and banking material (18%), gradually decreased and was instead used recycled as raw materials for cement. This reflects the global trend to resolve environmental disputes and eliminate pollution by safely recycling slag, through a kiln process (1450 °C), as cement raw materials rather than through direct recycling. Figure 6b shows that steelmaking slag was recycled in all seven categories. The proportion recycled as road, banking, and process reuse varied from 1.9% to 22.5%, 26.5% to 46.3%, and 11.9% to 20.5%, respectively.

Figure 6. Changes in usage of (a) blast furnace slag and (b) steelmaking slag in the Republic of Korea between 2010 and 2019.

Table 2 summarizes the applications of recycled slag in different countries. In Japan, Europe, the United States, and China, slag is used in road construction, cement products, internal recycling, construction, and agriculture (Figure S4). The Japan Steel Association produced 137,000 tons in 2017, with a reuse rate of 100% [21]. Of this, 35% was reused for road construction and 32% for construction, with applications categorized into cement raw materials, soil improvement, internal and external recycling, and other uses. In Japan, steel slag is used to reduce H_2S in the sea; thus, effective use of steel slag can also improve the marine environment. According to a survey by the European Steel Association, 28 countries in Europe together generated 478,000 tons of steel slag in 2018 [22]. Based on 2018 data from the European Steel Association, 69.9% of this slag was recycled for road construction, 13.4% for metallurgical use, 5.5% for cement and concrete, and 5.3% for other uses [22]. According to a United States Geological Survey report, the United States produced 170,000 tons of steel slag in 2019; cement production (49.7%) was the largest application, while road construction, final disposal, and other uses each accounted for 15% [23]. Steel slag production in China reached 100 Mt in 2013. As the reuse rate of steel slag in China is very low, an accumulated 3 Mt of steel slag has been discarded; as such, the proportion of steel slag being used for roads or agriculture is small.

Table 2. The application of slag in different countries.

Area	Usage	Republic of Korea	Japan	USA	EU	Australia	China	Remark
Raw material	Cement raw material	○	○	○	○	○	○	
	Concrete admixture	○	○	○	○	○	○	
	Silicate fertilizer	○	○	○	○	○	-	
Aggregate	Brick clay	○	-	○	-	○	○	
	Concrete aggregate	○	○	○	○	○	○	
	Banking	○	○	○	-	○	○	
	Soil covering	○	-	○	-	○	○	
	Dike construction	○	-	-	○	-	-	
	Public water surface backfilling	○	○	○	-	○	○	
	Road aggregate	○	○	○	○	○	○	
	Chemical construction	○	-	-	-	-	-	
	Dewatering aggregate	○	○	-	○	○	-	
	Non-skid aggregate	○	-	○	-	○	-	
Other	Retaining wall and backfilling	○	-	○	○	○	○	
	Rubble stone	○	-	○	-	○	○	Road construction
	Rockwool	○	-	○	○	○	-	
	Pigment	○	○	-	-	-	-	
	Soil amendment	○	○	○	○	-	-	
	Chemical additive	○	-	○	-	-	-	
	Processing material	○	○	-	-	-	-	Magnetic separation
	Railway material	-	○	○	○	-	○	
	Wastewater treatment	-	-	○	○	-	-	Remove nitrous and phosphorus
	Filter	-	-	○	-	○	-	Trout farm
	Land fill material	○	-	○	-	-	-	
	Mining wastewater treatment	-	-	○	○	-	-	
	Building roof materials	-	-	○	○	-	-	

3.2. Steel Slag Management Regulations and Legal Position in the Republic of Korea

Table 3 shows the regulations and guidelines for the aging period of slag in different countries. The ROK determines the required aging period of slag by size. Slag with a particle size less than 100 mm must undergo a natural aging period of at least 1 month, while slag greater than 100 mm in size must undergo a natural aging period of at least 3 months. The aging period and method are not determined by its slag type.

Table 3. Regulations for the recycling of slag in different countries.

Country	Slag Type	Regulation and Guideline About Slag Aging
Republic of Korea	Steel slag	- Size $\leq$ 100 mm, stockpiled for at least 1 month - Size $\geq$ 100 mm, stockpiled for at least 3 months
USA (DOT)	Air-cooled slag or blast furnace slag	- Stockpiled for a minimum of 1 month - Must pass the test of leachate determination (bucket test)
	Steelmaking slag	- Stockpiled for 6 months to allow hydration and expansion - ASTM D4792 [00(2006) Standard Test Method for Potential Expansion of Aggregates from Hydration Reactions]
Belgium	Steelmaking slag	- Stockpiled for a minimum of 12 months - Free CaO: $\leq$ 4.5%
Netherlands	Steelmaking slag	- Stockpiled for a minimum of 12 months - Free CaO: $\leq$ 4.5%
Australia	Steelmaking slag	- Stockpiled for 1~3 months
New Zealand	Steelmaking slag	- Stockpiled for 1~3 months
Brazil	Steelmaking slag	- Stockpiled for a minimum of 6 months

In the United States, aging period guidelines are different for blast furnace slag and steelmaking slag. Blast furnace slag must be stockpiled for at least one month and must also pass a leaching test. Steelmaking slag must be stockpiled for six months, with consideration given to hydration and expansion, and must pass the “Standard Test Method for Potential Expansion of Aggregates from Hydration Reactions,” an American Society for Testing and Materials (ASTM) D4792 test method.

In Belgium and the Netherlands, steelmaking slag must be stockpiled for at least 12 months; in particular, the free CaO content must be 4.5% or less, a requirement different from other countries. This is to alleviate concerns of hydration expansion in steelmaking slag. Australia and New Zealand require steelmaking slag to be stockpiled for one to three months for aging, while Brazil requires it to be stockpiled for at least six months.

Based on the steel slag aging regulations or guidelines of other countries, the slag aging period in the ROK should be extended, to a more long-term period. Furthermore, alternative indicators should be used, such as pH and the amount of free CaO, to ensure ideal aging of steel slag.

Table 4 shows the legal position of slag in different countries. In the ROK, slag is managed as general waste and designated as a by-product in the Wastes Control Act. In Japan, slag is not considered as waste if used as a beneficial substance, as stipulated in the Law of Waste and Clean in the Waste Management Act and the Law for Promotion of Effective Utilization of Resources. In the United States, the federal government designates slag as a co-product, Pennsylvania designates it as a by-product rather than waste, Michigan defines only non-ferrous metal slag, generated during the smelting or melting processes of steel, as slag, and Ohio states that blast furnace slag generally does not cause environmental damage when reused for construction. Additionally, in Indiana and Illinois, steel slag is not subject to sanctions under the Wastes Control Act. The United Kingdom, the European Union, and Australia consider slag a by-product rather than waste.

Table 4. The legal position of slag in different countries.

Country	Main Content	Remark
Republic of Korea	Slag is classified as a “by-product” in the Resource Conservation and Recycling Promotion Act	Article 3 under Waste Management Act in Republic of Korea
Japan	- Law promoting effective use of resources: slag material is a co-product of the steel industry and is defined as a special resource - Act on the Promotion of Effective Utilization of Resources: slag is not waste if used as a beneficial material	
US	Slag is a co-product rather than a by-product or waste, according to federal government and in certain states	United States
	Department of Environmental Protection (DEP): slag is a co-product rather than a by-product or waste	Pennsylvania state
	Compo Laws § 324.11506 (I)(f). “Slag” is defined as “the nonmetallic product resulting from melting or smelting operations for iron or steel”	Michigan state
	The policy also notes that blast furnace slag generally does not have negative environmental effects when used as a construction material	Ohio state
	Steelmaking slag is not covered by solid waste regulations in Indiana or other states	Indiana, Illinois state
UK	Blast furnace slag defined as a by-product. BFS produced in the UK could be classified as a by-product	www.environment-agency.gov.uk (accessed on 6 July 2021)
EU	- Granulated slag from the production of iron and steel is considered to be a product in Austria and in many Organizations for Economic Co-operation and Development member countries - In Europe, steel slag and blast furnace slag products have been eliminated from the European Waste Catalogue - Slag treated with granulation, palletization, foaming, separation, crushing, sieving, and milling are excluded	European works council 10 02 01 waste the processing of slag 10 02 02 unprocessed slag
Australia	Slag is a by-product of the iron and steel manufacturing process	

3.3. Environmental Risk Assessment of Steelmaking Slag in the Republic of Korea

3.3.1. General Physico-Chemical Estimation of Steelmaking Slag

The pH of steel making slag for ROK ranged between 9.3 and 12.4, due to the high enough composition of free CaO which reacts with H₂O to generate hydroxide ion (HO[−]) [24]. The composition of steel making slag ($n = 16$) was analyzed using XRF. Table 5 shows the result of comparative composition of steelmaking slag. The CaO (46.1%) and SiO₂ (41.8%) were the main chemical composition of steelmaking slag in the ROK. Compared with other countries, CaO in the ROK was a main composition of steelmaking slag, which was higher than 35%. Although total iron (Fe_t) was not detected in ROK, the content of Fe_t ranged between 17.4 and 24.36% in other countries. A previous study reported that CaO, MnO, and MgO, which generate a considerable portion of steelmaking slag, can be used as alternatives for limestone, dolomite, and manganese ore to effectively reduce iron content and costs [25].

Figure 7 shows the contents of steelmaking slag for 18 samples in ROK. The standard of EAoR for media-contact recycling in ROK has been regulated for heavy metal such as Cd, Cu, As, Pb, Zn, and Ni. The results showed that heavy metals such as Cd (22%, $n = 32$), Cu (28%, $n = 32$), As (28%, $n = 32$), Pb (1%, $n = 32$), and Zn (22%, $n = 32$) exceeded the Standards for soil environmental pollution. In the unregulated heavy metals of KS, the concentration of the heavy metals range was Cr (58.1–1032 mg/kg; Avg. 1345 mg/kg), Ba (25.7–16,103; Avg. 17.2 mg/kg), Pb (2.2–1578 mg/kg; Avg. 192 mg/kg), Zn (2–123 mg/kg; Avg. 22 mg/kg), V (2–1000 mg/kg; Avg. 388 mg/kg), and Mo (0.1–20,062 mg/kg; Avg. 1509 mg/kg). Previous studies reported that the content concentration of heavy

metal investigated was Cd (0.1–128 mg/kg; Avg. 128 mg/kg), Cu (3–540 mg/kg; Avg. 540 mg/kg), As (0.15–244 mg/kg; Avg. 244 mg/kg), Pb (2–1040 mg/kg; Avg. 126 mg/kg), Zn (1–11,000 mg/kg; Avg. 11,000 mg/kg), Cr (4–32,700 mg/kg; Avg. 4798 mg/kg), Ba (24–1800 mg/kg; Avg. 366 mg/kg) [26]. The concentration of Zn and Pb in ROK was 8–9 times and 1.5–2 times higher than that other countries; conversely, the concentration of heavy metals (Cr, Cd, Ba, Pb, and Cu) was lower. Previous studies reported that V in steelmaking slag had a concentration of 690 mg/kg [5,27], and was hazardous with potential toxicity [28]. The steelmaking slag contains higher concentration of heavy metals than that of the soil as shown in this section. Heavy metals may have a negative effect on humans and the environment due to the characteristic of elution when recycled as media-contact recycling. Therefore, the leaching test proposed two ways, such as distilled water and seawater, when it comes to media-contact recycling.

Table 5. Chemical compositions of slag in different countries.

Component	Composition (%)								
	Republic of Korea	USA	Australia	Turkey	Japan	Brazil	Sweden	France	China
CaO	46.1	40–52	35	38.62	45.8	45.2	45.0	47.71	42.92
SiO ₂	41.8	2.1	14	19.28	11.10	12.2	11.1	13.25	11.51
Al ₂ O ₃	-	1–3	5.5	2.71	1.9	0.8	1.9	3.04	1.4
MgO	6.3	5–10	7.7	8.05	6.5	5.5	9.6	6.37	4.36
MnO	5.4	5–8	5.7	7.52	5.3	7.1	3.1	2.64	4.04
P ₂ O ₅	-	0.5–1	-	-	1.7	-	0.23	1.47	0.83
Total iron (Fe _t)	-	-	-	22.61	17.4	18.8	23.9	24.36	23.74
S	0.08	-	0.1	0.28	0.06	0.07	-	-	0.07
K ₂ O	-	-	0.1	-	-	-	-	-	-
Cr ₂ O ₃	-	-	1	-	-	-	-	-	-
V ₂ O ₅	-	-	0.3	-	-	-	-	-	-
Fe ₂ O ₃	-	10–40	29	-	-	-	-	-	-

Figure 7. Heavy metal content in steelmaking slag.

3.3.2. Estimation of Leaching Test for Steelmaking Slag

The leaching test was performed following with Waste Control Act. The results of leaching test are shown in Figure 8. Within 6 h, some heavy metals such as Cu, Zn, Cr, Ba, V, and Mo were liquated under the leaching test using distilled water and seawater. Especially, Zn, Ba, and Mo showed an extensive range of 0.25–1.2, 0.4–2.07, and 0.1–31.8 mg/L, respectively. The high concentration of Mo released from steelmaking slag was not ignored although their concentration was detected one time. The leaching characteristic of heavy metals was observed according to different leachants such as distilled water and seawater. This suggests that the transport and transformation of hazardous heavy metals is possible based on their coordination environment, molecular geometry, and oxidation state when the steelmaking slag was reused or landfilled. In addition, the different leachability of heavy metals was shown according to the different eluent.

Figure 8. Concentrations of heavy metals based on the distilled water and seawater leaching tests.

Spanka M. [29] reported that V and Mo are hazardous heavy metals that are easy to leachate to alkali conditions, and have been shown to cause environmental and ecosystem damage [30]. Chaurand et al. [31] demonstrated that the toxicity of Cr and V was significantly different according to the oxidation state. The model of heavy metal transport model can estimate the environmental impact of heavy metals [30].

The concentration of Ca and Mg ranged between 1.442–398 and 0.049–0.8 mg/L, respectively. These heavy metals also have a negative effect on the environment and humans. Mayes et al. [32] reported that Mg and Ca can increase hazardous sediment, which highlights the long-term damaging effect due to the storage and leaching of slags. Gomes et al. [33] suggested caution when reusing the steelmaking slag according to their monitoring data. The authors investigated the heavy metals of surface and ground water near steel companies and found high levels of V (1.65–1.8 µg/L) and Mo (2.8–23.4 µg/L), which contributed to environmental damage. Hull et al. [34] proposed V as potentially toxic under alkaline conditions, as previously shown [31,35], keeping mobility under neutral and alkaline pH conditions [36]. Seawater releases more heavy metals than distilled water, which can be explained as follows: (i) the pH of leachant in distilled water is more alkaline (pH 9.3–12.5) than that of seawater (pH 7.5–11.9). A previous study reported high heavy metal leachability when salt concentration is increased [37]. (ii) Substances travel with different speeds based on the different solutes such as salts, metals, and organics, which influences heavy metal release [38]. (iii) Effect of time frame affect to extrapolate between scales and conditions of percolation and leaching behavior [39,40]. (iv) Background matrices such as DOM, chloride, and sulfate impact the leaching behavior [41]. The concentrations of the heavy metals leached from steelmaking slag were generally much lower than the total concentrations, which was a solid pH dependency.

3.3.3. Evaluation of Toxicity with *Aliivibrio fischeri*

The leaching solution obtained from the leaching test of steelmaking slag was used to estimate the potential toxicity using luminescent bacteria when the distilled water and seawater were used as the leachants (Figure 9). The inhibition value of toxicity using distilled water ranged between 0.8 and 1.0, meanwhile, the inhibition value of toxicity using seawater ranged between 0 and 0.4. pH and heavy metals might have contributed to these differences. The pH for leaching solution using distilled water and seawater was 9.1–12.0 and 7.4–11.2, respectively. Toxicity was increased with increasing pH. In addition, the leaching of heavy metals was observed in high alkaline conditions [32], which increased toxicity. A previous study reported an increase in Cr and V toxicity while in oxidation state [31]. pH not only influences toxicity but also releases leachability of heavy metals. Fang et al. [42] reported that the hazardous wastes reused in soil or landfill can change the characterization of soil, thus prompting heavy metal leachability. Moreover, this toxicity may be influenced by other unknown substances generated during leaching test such as dissolved organic, inorganic, and transformation substances, and dissolved organic carbon-based ion different leachants.

Figure 9. Toxicity of leaching test on *Aliivibrio fischeri* luminescent bacteria.

3.4. Future of Steelmaking Slag Recycling

New applications for recycling slag in the Republic of Korea were examined, based on those investigated in different countries (Table 6). The following new recycling applications were identified as feasible: iron recovery, railroad ballast aggregate, wastewater treatment filtering, building roof materials, marine forests, artificial reefs, tetrapod blocks, in situ capping, soil (ground) conditioning, and materials for red tide prevention (Table 6). Of these new applications, iron recovery was excluded from review because redox slag is not separated and discharged from blast furnace slag, while marine forests, artificial reefs, and tetrapod blocks were excluded from review as they are already designated for recycling by the Ministry of Food, Agriculture, Forestry and Fisheries.

Table 6. New feasible usage for slag recycling.

	Usage	Remark
	Raw materials (cement, admixture, fertilizer, rockwool, ceramic)	
	Aggregate (brick, concrete, road, ascon, rubble)	Use is specified in current recycling guidelines
	Filling caissons	
	Anti-slip aggregate	
	Iron recovery	Not yet possible (lack of technology)
	Railroad ballast	Excluded from review (sold according to road aggregate standards)
	Building roof materials	Excluded from review (ceramic material)
Water treatment	Wastewater treatment, filter	Excluded from review (patent application from a specific company, involving atomizing a fine powder) Sold as a product
	Mine wastewater	Excluded from review
Ocean	Artificial reef	Excluded from review
	Tetrapod block	Designated by other law (Ministry of Food, Agriculture, Forestry and Fisheries)
	In situ capping	
	Materials for red tide prevention	Reviewing
	Soil conditioner	Reviewing
	Fine aggregate substituted for sand in sewage pipe installation	Reviewing

The applications that require close review are in situ capping, red tide prevention, and soil (ground) conditioning. For in situ capping (which involves the installation of a non-polluted layer on top of a polluted sediment layer), the use of steelmaking slag reduces the quantity of material required to form a cap thickness of 100 mm or less, while also reducing facility costs by at least half (compared to other materials). Moreover, suitable particle sizes can be selected for each application environment during the production process, thereby facilitating quality control and expanding the applicability of steelmaking slag, relative to general sand. In situ capping with steelmaking slag has four main advantages. First, steelmaking slag has an excellent adsorption and removal capacity for phosphate, hydrogen sulfide, and heavy metals. Second, in actual experiments applying it in sea areas, in situ capping had an excellent inhibiting effect on leaching of pollutants from contaminated seabed sediments. Third, studies have reported inhibiting effects on leaching (50%), adsorption (40%), and chemical reactions (10%). Fourth, in situ capping has been applied in the United States (rivers, lakes, ports, etc.), Japan, and Europe.

Using slag as a material for red tide prevention has the following advantages over using loess: first, the particle size is easily adjusted (during production). Second, there is reduced risk of pollution due to impurities (suspended solids) in the loess. Third, it can improve the habitat of Manila clams and blood clams inhabiting the surface layer of sediment. Fourth, it can improve the habitat of oysters and sea squirts farmed in the water layer. Fifth, it is easily sprayed over an area. The loess spraying method, which is commonly used for red tide prevention, involves dissolving and spraying natural loess in the seawater. This method is unscientific, has not been proven effective, and uses more loess than necessary, thus causing damage to nature and secondary marine pollution.

There are five reasons to use slag as a soil (ground) conditioner. First, it can be used to create a firm surface on ultra-soft ground comprising dredged soil or marine clay along the coast. Second, it can replace natural sand used in the sand compaction pile method, where it acts like crushed stone or gravel. Third, it is applicable in various methods, such as the

preloading, replacement, and surface drainage methods. Fourth, it can be used as loading embankment and pavement material. Lastly, there are numerous documented cases where slag was applied as a ground conditioner [15,16].

4. Conclusions

In the ROK, the recycling of steel slag has progressed steadily until 2016. Recycling methods for safer and more diverse recycling applications were analyzed from 2016, after the introduction of the EAoR system. Current data on steel slag recycling in the ROK were analyzed and compared with corresponding data from several other countries. We proposed a solution through the determination of the extensive applications for steelmaking slag, which is an indicator of the proportion of recycled resources used, and determined the factors that hamper recycling. The key findings of our study are as follows:

1. The ROK recycled as much as 98.3% of their generated slag, which is relatively high compared to other countries. However, the need to minimize environmental pollution while maintaining media-contact recycling was recognized. The ROK does not use much slag in road construction, agriculture, or offshore structures, compared to some advanced countries.

2. We identified a need for environmental recycling standards. Methods for improving the aging period of steelmaking slag must also be considered. The environmental effects of different aging periods must be determined, and further research on CaO content and indirect aging evaluation methods is needed.

3. In the EAoR system, which has been utilized in the ROK since 2016, products that are environmentally certified are exempt from EAoR regulations. Moreover, there is an urgent need for environmental standards relating to slag that is recycled as a product, and the recycling and environmental label certification for products should also be amended.

4. To effectively recycle steelmaking slag, an application plan is proposed, which involves in situ capping, materials for red tide prevention, soil conditioning, and fine aggregate substituted for sand in sewage pipe installation

Based on these results, sustainable resource management can reuse resources and effective recycling within a safe range for human health and the environment by extending the applications for steelmaking slag in the ROK. The suggested applications are promising, however, they should be monitored and periodically tested over a longer period.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/su13168805/s1>, Figure S1: Schematic of percolation test, Figure S2: Procedure of toxicity test, Figure S3: Schematic of estimation toxicity using with *Daphnia magna*, Figure S4: Steel slag utilization in Japan, Europe, the United States, and China, Table S1: Standard guidelines for recycling of slag in different countries, Table S2: Comparison of leaching methods in different countries.

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