



Chemical behavior of sand alternatives in the marine environment



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HIGHLIGHTS

- This study clarified Ca elution of 6 sand alternatives in the sea.
- Calcium elution was evaluated as a main chemical behavior causing pH increase, etc.
- Basic parameters for the Ca elution were obtained in lab experiment and in the sea.
- The maximum amount of Ca eluted was almost zero in clinker ash and molten slag.
- That of 2 steel slag and 2 granulated materials were estimated as 19–114 mg g⁻¹.

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ABSTRACT

Steel slag and granulated agglomerates from the steelmaking process, clinker ash and granulated agglomerates from thermal power stations, and molten slag from municipal solid waste incineration plants are some of the promising alternatives to marine sand for restoration of coastal marine habitats. In this study, the characteristics of elution of Ca from the six sand alternatives during aging in the marine environment was determined. The maximum calcium elution potentials of decarburized slag, dephosphorized slag, granulated slag, clinker ash, granulated ash, and molten slag were 204, 75, 26, 6, 19, and 5 mg g⁻¹, respectively. However, the elution of Ca from decarburized and dephosphorized slags was limited to depths of about 140 and 300 μm from the surface of the slag, whereas there was no limitation in calcium elution in 3 mm from granulated agglomerates and ash. The maximum amount of calcium eluted into seawater during 500 d was estimated to be almost the same for the four alternatives if the particle diameters were about 4.75 mm because of the shallow maximum calcium elution depth of the decarburized and dephosphorized slag, even though their maximum elution potentials were 3–10 times those of granulated slag and granulated ash.

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1. Introduction

Coastal zones are important ecologically and socioeconomically, but they have been greatly modified to accommodate urban and industrial development. The population within 100 km of a shoreline and 100 m above sea level has been estimated to be 1.2×10^9 people, the average density being nearly three times the global average (Small and Nicholls, 2003). Thirty-five percent of the total population of Japan lives within 10 km of the shoreline and 30 m

above sea level, a coastal zone that accounts for only 10% of the total land area (Ministry of Land, Infrastructure, Transport and Tourism, 2011). Coastal development and increasing pollution have led to the degradation of coastal ecosystems such as intertidal flats and seagrass beds. In Japan, 40% of intertidal flats (33 120 ha) disappeared within the last 60 years (Inoue and Suito, 2002; Suito and Inoue, 2002). There has recently been an attempt to restore and create seagrass beds and tidal flats to maintain healthy marine ecosystems. Similar restoration projects and studies of the recovery of seagrass beds and tidal flats have been conducted all over the world (Terawaki et al., 2005). However, a large amount of sand is required to rehabilitate these areas, and the extraction of sand

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from the sea or riverbeds may disturb the associated aquatic ecosystems. Hence there is a need for some sea sand alternatives (hereinafter referred to as “SAs”).

Iron and steel slag from the steelmaking processes, coal ash from thermal power stations, and molten ash from domestic waste incineration plants are all industrial byproducts, and in Japan are produced at rates of approximately 36, 10, and 0.8 million tons per year, respectively (Nippon Slag Association, 2010). These byproducts have been used as roadbed construction material, the coarse aggregate in concrete, and a component of the raw material for cement (Martin, 1990). However, the output of these byproducts in excess of demand is still enough to provide substrates for the creation and restoration of coastal ecosystems such as tidal flats and eelgrass beds.

Iron and steel slag is classified into two main categories, blast furnace slag and steelmaking slag (steel slag, including converter slag). The former is produced from the process of converting iron ore into pig iron, and the latter is produced when CaO is added in the purification process when pig iron is converted into steel. Steel slags are further subdivided into desiliconized slag, dephosphorized slag, decarburized slag, and others, the category depending on the specific process used to purify the pig iron. The value of these slags for recycling may be augmented by making granulated agglomerates with cement and the fly ash from coal ash (Utagawa and Matsunaga, 2008). The coal ash from thermal power stations can be classified mainly into clinker ash and fly ash. The fly ash contains CaO from the original coal (Murayama et al., 2001). The material used to make granulated agglomerates contains about 10% cement, which also contains Ca (Asaoka et al., 2008). Molten slag is the fused ash from incineration plants and is produced to decrease the elution of added Ca during incineration and the release of heavy metals during the gas purification process so as to promote their recycling.

From the standpoint of environmental safety, the acceptability of these environmentally regulated substances as SAs has been verified on the basis of soil pollution standards set by laws that concern the prevention of marine pollution and maritime disasters (Hizon-Fradejas et al., 2009). For the target SAs, the concentrations of environmentally regulated substances such as heavy metals must obviously be lower than these environmental criteria. However, it has been reported that the six candidate materials (i.e., dephosphorized slag, decarburized slag, granulated agglomerates of steel slag, molten slag, clinker ash and granulated agglomerates of fly ash) for the SAs commonly contain Ca and would be expected to have CaO in their structure. CaO is called ‘free lime’, and it increases the pH of interstitial water by reacting with water to produce Ca and hydroxide ions. The pH increase triggers various chemical reactions between the SAs and seawater or sediment (Futatsuka et al., 2004; Miki et al., 2004). Understanding the elution behavior of Ca and alkalis in marine environments is important. Their behavior depends on conditions in the liquid phase, namely, seawater in the marine environment and its flow characteristics. Therefore, it is important to understand the chemical changes that occur after submergence and to apply that information when SAs are used to rehabilitate seagrass beds and tidal flats. To evaluate environmental impacts, it is also particularly important to quantify the maximum elutable amounts of Ca in situ, taking into consideration the depth of elution and the elution kinetics of each SA. However, there is no information on the aging behavior of each SA in the sea.

In this study, we quantified the in situ chemical changes of the six SAs containing reactive CaO, namely, decarburized slag and dephosphorized slag produced from the steelmaking process, granulated slag, clinker ash, granulated fly ash, and molten slag. We quantified the maximum elutable amount of Ca from the SAs and characterized the kinetics of calcium elution in seawater.

2. Materials and methods

2.1. Target materials

Furnace slag and steelmaking slag and their granulated agglomerates were provided by a steelmaking plant in Japan. The two slags were generated and discharged through decarburization and dephosphorization processes from pig iron into steel, and were designated decarburized slag and dephosphorized slag. Clinker ash and the granulated agglomerates of coal ash were obtained from a thermal power station in Japan. Molten slag was obtained from a municipal solid waste incineration plant in the Shikoku region.

We used an electron probe micro-analyzer (EPMA; JXA-8200, JEOL) to determine the chemical composition of each SA and their mapping. The SAs were sliced through at the center, and the cross-sections were ground. The analysis for the chemical composition of elements having atomic masses higher than 22 (Na) was semi-quantitative. The values are averages of four points on the surface. The large standard deviations (SDs) reflect the non-uniformity of the sample composition.

For this analysis, each SA was pulverized with a zirconium ball in a high-energy planetary ball-mill (Fritsch Japan Co., Pulverisette-7) and sieved to obtain particles less than 250 μm in diameter. Phosphate, Mn, Al, and Ti. Calcium and silicon were the main components of these six materials (Table in supplementary material). The average Ca content were 61, 44, 31, 3, 17, 29%-atom for decarburized slag, dephosphorized slag, granulated slag, clinker ash, granulated, fly ash, molten slag, and the value of decarburized and dephosphorized slag were higher than reported elsewhere (Murayama et al., 2001; Utagawa and Matsunaga, 2008; Sugawara et al., 2010).

2.2. Continuous extraction to determine the maximum elution potential of calcium

The maximum calcium elution potential of the SAs was determined in batch elution experiments by successive replacement of the artificial seawater medium. A powdered SA consisting of particles with diameters less than 250 μm was used for each elution test on the basis of JIS K 0058-1 (Test method for chemicals in slags Part 1: Leaching test method). A 20-g portion of the powdered sample was stirred in 200 mL of artificial seawater with a salinity of 30‰ (SEALIFE, Marinetech, Japan) at 25 ± 1 °C in a 700-mL vessel. Thereafter, it was mixed at 100 rpm in a rotary shaker. The artificial seawater was replaced every 6 h. The particles were separated from the old seawater by filtration through a 0.47- μm pore size membrane filter (Millipore, HAWPO470). The amount of eluted calcium in each fraction was determined by inductively coupled plasma atomic emission spectroscopy (VARIAN, 720-ES). Theoretically, this method can detect $1 \mu\text{g L}^{-1}$ of Ca, however Ca in seawater (about 260 mg L^{-1}) limits the detection of eluted Ca. Its standard variation of replicate analysis was 25.0 mg L^{-1} , therefore detection limit of eluted Ca, which determined on the basis of the significant difference from the concentration in seawater, was 50 mg L^{-1} . Detection limit and analytical error for the analysis of other elements by the semi quantitative mode in seawater for Mg and other elements were around 10 and 1 mg L^{-1} by the ICP-AES, respectively.

2.3. Aging experiments in the sea to determine maximum elution depth

The depth of maximum elution of Ca from each SA was determined by aging in the sea for over 500 d. The aging site was located in the Iwakuni coastal area of the Seto Inland Sea, Japan. The depth of submergence ranged from 1.0 to 3.0 m. The aging experiment

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