

論文の要旨

Reuse of stone powder in slag or cement-treated clay

(スラグまたはセメント処理土における石粉の再利用)

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To achieve Goal 12 of the United Nations Sustainable development goals (SDGs), the construction industry emphasizes reducing, reusing, and recycling waste materials. Also, to meet the Paris Agreement on Climate Change and the Goal 13 of the SDGs, the industry needs to find innovative ways to reduce its carbon emissions. The manufacturing process of cement contributes about 8% to global carbon emissions; hence alternative materials should be found to reduce the reliance on cement. Waste materials (stone powders) produced from crushing rocks such as limestone and granite are often discarded in an environmentally unfriendly manner. These materials account for up to 25% of the quantity of crushed rock aggregates produced, but due to their small particle size ($<1-850\mu\text{m}$) they cannot be used in concrete hence they are disposed of. In Japan, India, and the United States, the quantity of crushed rock aggregates produced annually is approximately 390, 200, and 175 million tonnes, respectively. This means that the quarries are stocking or discarding large quantities of stone powder; hence innovative ways to reuse this stone powder should be investigated. Steel slag, a by-product of steel manufacturing, is increasingly utilized in constructing infrastructure worldwide. However, steel slag is produced in vast amounts and about 40% is used in China, while 1% of the over forty million tons produced in Japan is disposed of. This leaves the industry with financial and environmental costs associated with steel slags' abundant production, which necessitates further enhancement of its reuse.

Stone powder is increasingly being reused to improve the geotechnical properties of soils. They are known for increasing the maximum dry density, friction angle, California bearing ratio (CBR), shear strength, unconfined compressive strength (UCS) and coefficient of permeability as well as reducing the Atterberg limits, optimum moisture content, swelling, and compressibility. However, the mechanisms governing the strength development of treated soils using stone powder are unclear. Steel slag is used to treat clays where it improves their unconfined compressive strength while the clays reduce their expansive properties, which promotes its reuse in solving geotechnical engineering problems. However, there are limited studies on the reuse of stone powder in steel slag-treated clays. Our research aimed to promote the massive reuse of stone powder and steel slag in improving the geotechnical properties of clay dredged from the sea. We mixed 0% and 30% of limestone or 0%, 15%, 30%, 50%, and 70% of granite powder with cement (8%, 15%, and 10%) or slag (8%) and clay (0, 30%, 50 and 85%) to form a cement-treated clay-granite powder, cement-treated clay-limestone, and slag-treated clay-granite composites. We investigated the mixing characteristics, and physical, chemical, thermal, and microstructure properties governing the unconfined compressive strength development of the cement-treated clay composites. Further, the durability of the cement-treated clay-granite powder and slag-treated clay-granite powder composites under seawater exposure was determined. In our study, interdisciplinary collaborations with civil engineers, geologists, and chemical engineers were implemented to explain the mechanisms of UCS development in composites effectively. The investigated mixing characteristics included flow value and liquid limit of composites while the physical properties included stone powder particle's size (A, B, C, D and E: >75 , $40-75$, $20-40$, <20 and

< 1–106 μm , respectively), specific surface area and content. The chemical properties included chemical compositions, pozzolanic reactivity, amorphous silica as determined using X-ray fluorescence (XRF), electrical conductivity and NaOH digestion, respectively. The thermal properties determined by thermogravimetry-differential thermal analysis (TG-DTA) were calcium hydroxide and bound water content. The Ca-k edge Synchrotron X-ray absorption near-edge structure spectroscopy (XANES) of cement, clay, stone powder, and cement-treated clay-stone powder composites was determined. The microstructure of the cement-treated clay-stone powder composites was also observed using a Scanning Electron Microscope (SEM). The durability parameters evaluated included the calcium ion leachate and magnesium ion absorption determined from sea water concentrations measured using a photometer, XRF analysis on composites, the UCS, and stress-strain curves of the composites.

The particle size of granite powder influenced the UCS of cement-treated clay in the order of $D > E > A > C > B$, and D and E exhibited higher UCS than the control sample that did not contain granite powder. The increase in the UCS was due to granite powder D's large surface area that promoted the nucleation of cementitious products and amorphous silica that promoted its pozzolanic reactivity with calcium hydroxide, forming more cementitious products. This was proved by the reduction in calcium hydroxide at 28 days of curing and chemical-bound water at 3 days. The microstructure of the composites was also altered by granite powder, where D improved the cement-clay paste and increased cementation, while interfacial transition zones were observed around granite powder A and B, which led to their reduced UCS. The reduced UCS caused by granite powder A, B and C was caused by their low surface area and their non-pozzolanic reactivity due to low concentration of amorphous silica. It was confirmed through regression equations that 9.4 mg/g of amorphous silica is the minimum concentration for a reactivity that leads to UCS increment.

However, particle size of limestone powder did not influence the cement-treated clay as the cement-treated clay-limestone composites obtained similar UCS to their control samples. This is because limestone does not contain amorphous silica and hence it is not reactive. However, due to its large surface area, limestone powder can increase the composite's 1 and 3 days' UCS, just like granite powder. The XANES spectra of the cement-treated clay-granite powder and cement-treated clay-limestone composites differed at the energy levels 4076 and 4084 eV where they displayed a calcite-like structure at 1 day of curing that progressed to a calcium hydroxide-like structure at 3 days. The slag/cement-treated clay-granite powder composites displayed lower leachates of calcium ions, although their magnesium absorption was higher than their respective control samples at 91 days. This implied that the slag/cement-treated clay-stone powder composites are more durable under seawater exposure; hence granite powder can be used to construct sustainable infrastructure at sea.

The reuse of stone powder at contents of 30%, 50%, and 70%, results in a 6.9%, 14.7%, and 28.6% reduction in cement used per cubic meter of cement-treated clay, respectively. This leads the construction industry to the realization of target 5 of goal 12 of the SDGs, which emphasizes the substantial reduction of waste generation through prevention, reduction, recycling, and reuse. Goal 13 and Paris agreement on climate change are impacted through the reduction of cement, which contributes 8% of the global carbon emissions.