Recent Advances in the Reutilization of Granite Waste in Various Fields

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Abstract: Quarrying and processing of granite produce large amounts of waste residues. Besides being a loss of resources, improper disposal of these wastes results in pollution of the soil, water and air around the dumpsites. The main components of granite waste are quartz, feldspars and a small amount of biotite. Due to its hard and dense texture, high strength, corrosion resistance and wear resistance, granite waste may be recycled into building materials, composite materials and fine ceramics, effectively improving their mechanical properties and durability. By using the flotation process, high value-added products such as potash feldspar and albite may be retrieved from granite waste. Also, granite waste has the potential for application in soil remediation and sewage treatment. This review presents recent advances in granite waste reutilization, and points out the problems associated with its use, and the related countermeasures, indicating the scale of high value-added reutilization of granite waste.

Keywords: Granite, Solid waste, Reutilization, Construction material, Fine ceramics.

1. INTRODUCTION

As a typical decorative architectural material, granite is an acidic volcanic rock formed by the condensation of magma in deep underground layers. Some granites have been metamorphosed into gneiss or migmatite. The main mineral is quartz, together with small amounts of anorthite, microcline, mica, hornblende and pyroxene, and secondary minerals (*e.g.*, tourmaline, apatite, zircon, garnet and magnetite). Granite usually has a light color, but may become gray, pink or orange depending on the content of feldspar and other minerals [1]. Granite is widely distributed in large reserves around the world; the major producers of granite are China, Brazil, India, South Africa, Spain, Finland, Norway and the U.S.A. In the past decade, China's granite exports have increased steadily, and the disposal of granite processing waste has become a severe environmental challenge [2].

During the excavation of granite and subsequent mechanical processing, some cannot be used due to blasting damage, cracks and defects. Granite processing factories produce tens of millions of tonnes of waste residues, 40 percent of which is derived from the cutting and polishing processes [3]. The impact of granite waste on the environment is threefold: (1) marginally useful scrap generated during quarrying and processing, which is randomly discarded in farmlands and rivers, causing the pollutions of soils and water resources; (2) dust generated from the cutting and polishing may be inhaled, causing lung damage [4]; and (3) cutting coolants (mineral oil, alkaline additives and surfactants) to reduce the wear on cutting saws and to reduce noise pollution are not readily precipitated and are strongly adhesive. Coolant discharge containing granite sludge pollutes farmland and rivers. Contamination of soils by granite waste permanently alters its texture, pH, redox potential and conductivity, reducing moisture and organic matter content. In addition, the build-up of heavy metals from this source leads to the deterioration of soil quality [5].

At present, disposal of granite wastes is frequently treated in simple accumulation or landfill, and its reutilization is relatively low [6]. Figure **1** shows the extent of reutilization of granite waste in construction materials in various countries [7]. It is primarily used as aggregates, fillers or additives in construction materials. It does not adversely affect the product performance, but its economic value is low and utilization is limited accordingly. This review systematically introduces the status of granite waste reutilization and analyzes the problems and their countermeasures, thus, laying the foundation for sustainable development of the granite industry.

2. STATUS OF GRANITE WASTE REUTILIZATION

2.1. Green Building Materials

2.1.1. Cements

As indispensable cementitious materials, cements are extensively applied globally in modern housing and infrastructure. Cement production is one of the main

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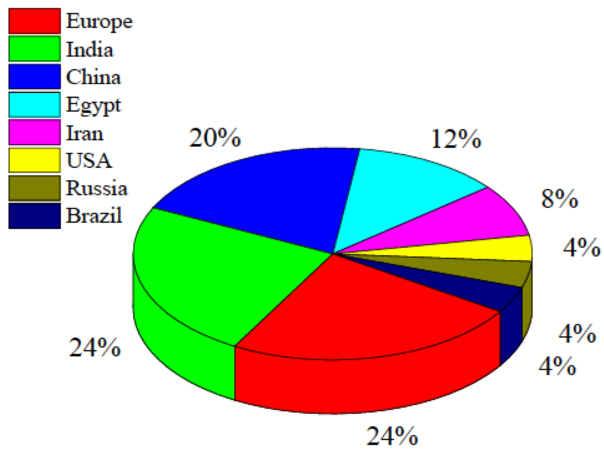

Figure 1: Reutilization of granite waste in construction materials in different countries.

sources of greenhouse gas. The production of 1 tonne of cement emits 1 tonne of CO_2, and accounts for about 7 percent of total global emissions. Granite waste is frequently used to replace part of the raw materials in cement production, which helps to reduce CO_2 emission [7]. Because the active silica in granite waste is very similar to the content in cement, it can be directly used in the production of ordinary Portland cement without any pretreatment [8]. The hydration reaction produces calcium silicate hydrate (C-S-H) gel and layered double hydroxides (LDH). The content of C-S-H gel increases with time, which improves the durability of cement [9] and reduces the CO_2 emission in the hydration reaction [10]. Granite powder plays an important role in forming a dense microstructure, improving the packing density of particles and reducing porosity, thus increasing the compressive strength of fly ash magnesium oxychloride cement [11].

When cement is used in oil well concreting material, granite waste is used to replace part of the silica fume, increasing the compressive and tensile strengths of the concrete by 5.7% and 39.3%, respectively, and decreasing its permeability and porosity by 64.7% and 17.9%, respectively [10]. The use of granite waste to partly replace the cement content has been found to improve the anti-corrosion performance of the concrete. Reports of controlled experiments indicate that it also has higher resistance to corrosion, frost and abrasion [12, 13]. Soaking in a sulfuric acid solution produces ettringite and precipitates gypsum in the pores, which increases the corrosion resistance of concrete components [14]. With a dense matrix in concrete, the expansion of concrete containing granite waste was reduced by 38%. The aluminate in granite waste reacts with chlorides to form chloroaluminate, increasing chloride resistance by 70% [15].

Granite waste can be used to replace fine aggregate in the cement mortar. Compared with the traditional cement mortar, the difference in mechanical properties is very small, and conforms to European standards [16]. When 30% and 40% granite waste replace fine aggregate, the water content of cement mortar is reduced by 7% and 3%, respectively. The lower water content helps to improve the mechanical properties of the mortar, and both the tensile strength and bonding strength are increased by 23% [17]. Marmol *et al*. [18] studied the role of granite waste in cement mortar and found that the addition of granite waste increases its compressive strength. The Fe_2O_3 in granite waste calcined at 700–900°C is converted to a reddish color. In this way, colored cement mortar can be prepared without affecting the compressive strength.

Refined granite waste improves the densification of the concrete and reduces its porosity. For a setting time of 7 to 28 days, the porosity and water absorption of all the tested cement mortars decreased with increasing setting time, and the bulk density increased with increased setting time [19]. The hydration product fills the pores in the mortar, thereby improving the compressive strength of the concrete after setting. The filling effect of granite waste produces a lower porosity matrix, which inhibits acid intrusion and improves its corrosion resistance. However, it was found that excessive substitution by granite waste in the mortar matrix leads to lower density and increased porosity, and reduces the compressive strength and corrosion resistance [20].

Using granite waste to simultaneously replace part of both sand and cement in mortar improves its strength and fluidity. After setting, it has a better overall performance, with a compressive strength up to 66.2 MPa [21]. In another study, nanosized granite powder was used to replace 5% of the cement and 10% of the sand in the mortar. The simultaneous replacement increased the hydration rate of the cement mortar, reduced its porosity and increased its 28-day compressive strength [22].

2.1.2. Concrete and Geopolymers

Concrete is the most important building material in civil construction works and other infrastructure. The fine aggregate in concretes accounts for about 35% of its volume. River sand is a commonly used natural fine aggregate, but is faced with increasingly depleted resources and strict restrictions in environmental protection regulations. The main minerals in granite waste are quartz and feldspars, which have characteristics of granulation, compactness and water absorption similar to river sand. The addition of granite waste into concrete has been found not to affect the concrete structure or the formation of hydration products [23]. Taji *et al*. [24] studied the mechanical properties of concretes containing granite waste and its effect on the corrosion of steel reinforcing bars. The addition of 10% granite waste distinctly improved corrosion resistance. When 20% of granite waste was added, the mechanical properties of concrete were not affected, indicating that granite waste is a high-quality aggregate in the concretes.

(1) Compressive Strength

Compared with natural fine aggregate, coarse granite powder (CGP) has a rougher surface, and the angle and geometry of the particles vary greatly. The specific particle surface produces higher friction in cement slurry, implying that it is suitable as a concrete aggregate. Figure **2** compares the compressive strength of concretes with added granite powder for different setting times. Compared with the control sample (CM), the addition of granite powder basically did not affect the compressive strength of concretes, which in fact increased with longer setting times. The optimal replacement amount was found to be 10% [25].

Due to the large surface area of fine granite particles, an excessive addition of granite powder increases the amount of cement binder, resulting in poor density and reduced compressive strength [26]. After the addition of granite powder, the C-S-H phase in the concrete increases, while the calcium hydroxide (CH) phase decreases. The increased density and reduced porosity of the concrete are favorable for increasing the compressive strength and reducing water absorption [27].

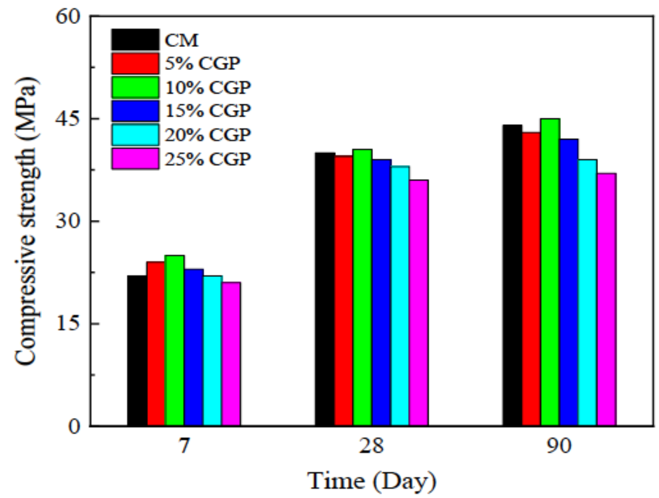

Figure 2: Variation of compressive strength of granite-added concretes vs. setting time.

When marble and granite powders are used as the concrete mineral admixture, the fine particles fill the pores and effectively disperse the cement, producing concrete with stronger cohesion and denser microstructure, which improves its compressive strength [28]. The granite sludge generated by the gang saws contains worn steel particles. The dense mixture of steel particles and granite particles contains 15% Fe_2O_3 and up to 5% CaO, which can be used to prepare ultra-high-performance concrete [29]. It has been found [30] that the addition of steel fibers to self-compacting concrete with granite significantly improves its compressive strength, exceeding 55 MPa with 0.2% added steel fibers. Likewise, increased compressive strength of concrete with granite powder was achieved by partially replacing cement and fine aggregate with 10% wollastonite fibers [31].

(2) Flexural Strength

Granite waste has been used to replace fine sand in the production of self-compacting concrete [32], increasing the flexural strength by 5.65% after 28 days setting time. This is due to the rough surface of the granite particles, which causes rigid accumulation between cement and aggregate. The granite particles provide a hard filling in the concrete, thereby improving the flexural strength. For a water: cement ratio of 0.30–0.40, the 28-day flexural strength of the mixture with 25% granite waste was the largest. The increase in flexural strength is due to the enhanced adhesion between the aggregate and the cement paste [33]. Due to high content of C-S-H in the hydration products, the flexural strength of the modified concrete was 20–50 MPa, which is 15%–60% higher than that of ordinary concrete [27].

Patil *et al*. [34] used granite waste and copper slag to replace 30% of the river sand in concrete. After 28-days the flexural strength of the concrete increased by 5%, but as the replacement increased, too much water remained in the concrete and increased the porosity, and both the compressive strength and flexural strength were reduced. When the fine aggregate was composed of river sand and granite waste, the early flexural strength of the concrete was relatively low, but it increased by 3.6, 9.5 and 13.4% with the setting times of 28 days, 56 days and 90 days, respectively [35].

(3) Durability

Concretes using granite waste as fine aggregate were subjected to chloride ion penetration experiments [25]. Higher aggregate replacement resulted in greater chloride permeability. This was attributed to poor compaction, resulting in a high-porosity microstructure and discontinuous pore system. Nevertheless, the permeability of concrete containing less than 15% of granite waste was substantially the same as for the control group. After setting for 180 days and 365 days, the concretes were soaked in Na_2SO_4 and $MgSO_4$ solutions for the acid resistance experiments. The compressive strength of the concrete with granite waste was largely lost; this became more serious with increase of replacement ratio. The use of petroleum ether (hydrocarbon) to chemically bleach the granite waste to remove the organic matter improved the sulfate resistance of concrete.

Granite waste was substituted for river sand to prepare autoclaved aerated concrete. An acid erosion experiment was carried out in 5% HCl and 5% H_2SO_4 solutions. The compressive strength loss in HCl solution was 19.44% and 13.14% for the control and 20% for the replacement samples. The compressive strength losses in H_2SO_4 solution were 27.88% and 12.82%, respectively, indicating that granite waste aggregate effectively improves the acid resistance of concretes [36,37]. The water absorption of concretes was tested using magnetized water with higher water molecular activity, and it was found that concrete with granite waste had a reduced water absorption and significantly improved compressive strength and acid erosion resistance [38]. The addition of soda lime glass powder increases the concrete density, reduces its water permeability and water absorption, and further improves its acid resistance [39].

As a cementitious material, geopolymers have excellent mechanical strength, heat resistance and favorable stability in acidic and alkaline environments. The main component of granite waste is silicon-aluminum oxide, suitable for the production of geopolymers by the alkali activation method. Granite waste reacts with the alkaline activator (sodium silicate, or water glass) to form a geopolymer binder mainly composed of sodium aluminosilicate hydrate (N-A-S-H) gel. Depending on the amount of Na_2O and curing time in the alkali activation process, the compressive strength of geopolymer mortar reaches up to 40.5 MPa [40]. When the activator solution consists of 18% Na_2SiO_3, 7% NaOH and 75% distilled water, the silicon-based geopolymer from granite waste has a maximum compressive strength of 22 MPa after curing at 220°C for 2 h [41].

Extending the curing time improves the flexural strength and crack resistance of granite waste geopolymer. The content of granite waste and water glass has the greatest impact on the flexural strength; the alkali content has little effect on the flexural strength of the geopolymer mortar [42]. When metakaolin and blast furnace slag were used as the starting materials for geopolymer synthesis, the bonding strength between the geopolymer and the granite aggregate decreased with increase in the activator modulus and the liquid/solid ratio, but it was much higher than between cement paste and granite. A liquid/solid ratio of 0.35 gives a maximum bonding strength of 1.53 MPa [43].

When nanosized alumina (Al-0450) and liquid hydroxyl functionalized nanotubes (MWCNT-OH) were added to granite waste-based geopolymers (GP), they were uniformly distributed within the geopolymers to form a dense and ductile structure, resulting in enhanced mechanical properties and delayed setting times for the slurries, as shown in Figure **3** [44].

2.1.3. Unfired and Fired Bricks

Granite powder has been used as part of the fine aggregate in the preparation of hollow concrete blocks. With a liquid/cement ratio of 0.55 and a fine/coarse aggregate ratio of 15%, the prepared blocks meet the strength requirements and have good frost resistance [45]. For sintered floor tiles and clay bricks, the addition of granite waste increases the bulk density and mechanical properties [46-48]. This is attributable to

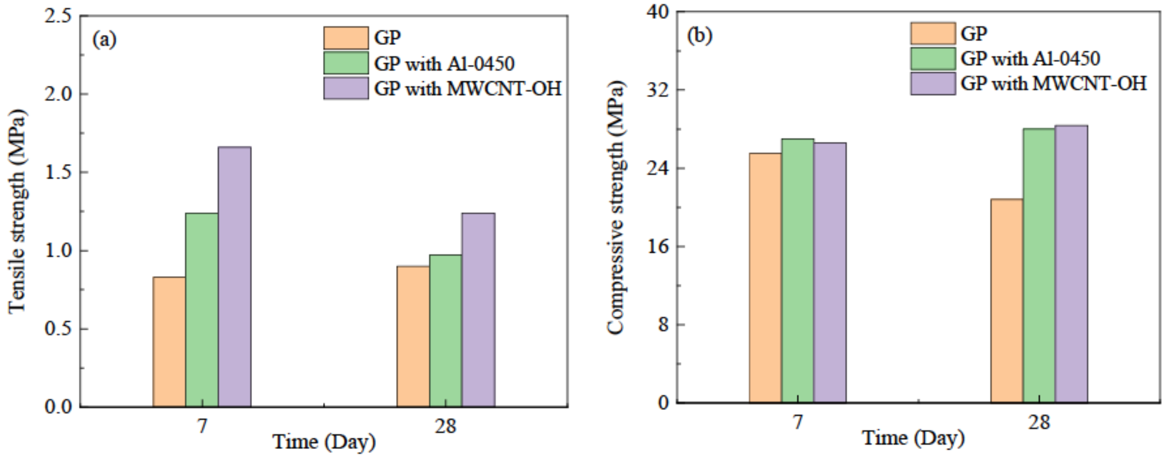

Figure 3: Mechanical strength of granite-based geopolymers with different setting times. (**a**) tensile strength, (**b**) compressive strength.

the densification by liquid-phase sintering and is derived from the low-temperature melting of alkali feldspars in the granite waste.

2.2. Composite Materials

Composite materials are filled with granite powder to improve their mechanical properties and durability. For example, in wind turbine blade manufacture, composite materials with a density of 1.108 to 1.321 g·cm^{-3} were prepared by adjusting the addition of granite powder without changing the contents of polyamide fibers and polyester resin. In air jet erosion experiments, the addition of less than 10% granite dust had a minor effect on the erosion rate of composites. Conversely, the Rockwell hardness increased with increased granite powder content: a composite with 5% granite dust withstands up to 60 ℃ [49]. In the case of decorative construction materials, marble and granite powder were added to high-density polyethylene to prepare a new type of composite material [50]. The flexural strength of the composite increased up to 23.5 MPa with increasing marble and granite powder content. Similarly, a composite material prepared with epoxy resin as the matrix and granite powder as the reinforcement [51] gave a Vickers hardness and impact strength of 22.93 and 42 J·m^{-2}, respectively. These composites are typical of simple, low-cost manufacturing processes.

Environmentally friendly composites have successfully been prepared using acrylonitrile butadiene styrene (ABS) and granite powder as raw materials [52]. The stiffness and thermal conductivity of the composites increased and the elastic modulus more than doubled with increasing content of granite powder; the flexural strength and fracture toughness decreased with the addition of more than 50 wt% granite powder.

To investigate the deterioration of mechanical properties, granite powder was modified by stearic acid to disperse granite particles in polystyrene composites [53]. Although the bending and impact strength of composites decreased with increased granite powder content, their surface hardness was increased by 130%.

Incorporation of granite powder into natural-fiber-reinforced polyester composites [54] continuously increased the tensile, flexural and impact strengths of the composites with increase of granite filler content up to 15%, then deteriorated due to the agglomeration of granite particles.

Polypropylene has excellent properties such as dimensional stability, thermal stability, optical properties, flame retardancy and high deformation temperature. The addition of granite powder to polypropylene was found to reduce the thermal expansion coefficient and improve heat resistance of the composite material [55].

2.3. Sintered Ceramics and Glass-Ceramics

Sintering in ceramic manufacture usually needs the addition of sintering additives. The liquid phase formed at the sintering temperature, viscosity and surface tension are important factors for selecting sintering additives. The chemical composition of granite waste contains alkaline oxides and alkaline earth oxides, which are suitable as sintering additives [56]. In the production of red ceramics, the alkaline oxides in granite waste reacts with silica and alumina to form a liquid phase, which promotes the densification of the ceramic and reduces its porosity [57]. Naga *et al*. [58] reported using granodiorite as a sintering additive. The sintered ceramics consist of primary and secondary mullites, glass phase and pores. With the addition of 35% granodiorite, the flexural strength was 41.1 MPa. In addition, periclase-forsterite ceramics have been sintered from magnesia and granite sludge by a temperature-induced forming method [59]. When the granite sludge increased from 10 to 40 wt%, the forsterite phase increased by up to 78%, and the microhardness increased to 7.9 GPa. The forsterite was formed by the diffusion of the granite sludge into the periclase phase.

Granite powder has been directly used to prepare sintered glass-ceramics. The effects of sintering additives on the densification, crystallization and flexural strength of glass-ceramics have been thoroughly investigated [60]. Boehmite is more suitable than silica sol and glass powder for the densification of glass-ceramics. At a sintering temperature of 1075°C, the bulk density of glass-ceramic is 2.49 $g{\cdot}cm^{-3}$ and the flexural strength is as high as 125 MPa. With increasing rate of sintering heating, the densification, crystallinity and mechanical properties were all completely improved [61]. Figure **4** shows the effect of sintering heating rate on flexural strength, Vickers hardness and fracture toughness of the sintered glass-ceramics. When sintered to 1085°C at a heating rate of 50°C$\cdot min^{-1}$, the main crystalline phase was anorthite with a flexural strength of 143 MPa and fracture toughness of 2.1 MPa$\cdot M^{1/2}$.

Pickling asbestos and mullite fiber have been used as the reinforcement in the sintering of anorthite glass-ceramics from granite powder [62]. With increasing fiber addition, the bulk density, flexural strength and fracture toughness of glass-ceramic first increased and then decreased, but the Vickers hardness continued to decrease. When 3% pickling asbestos is added, the flexural strength reached 144 MPa, with fracture toughness of 3.0 MPa$\cdot M^{1/2}$. In addition, granite powder was physically pretreated by magnetic separation to remove the iron-containing impurities for the decorative aesthetics of sintered glass-ceramics.

The crystal composition of glazed glass-ceramics consists of anorthite, albite and quartz. The surface glossiness of glass-ceramics is 82 gloss units (GU), and the flexural strength is up to 108.4 MPa [63]. As an architectural decorative material, copper-red glass-ceramics has successfully been prepared from granite waste with the incorporation of CuO [64]. The rod-like richterite endows excellent mechanical properties to glass-ceramics, with a flexural strength of 167.8 MPa and Vickers hardness of 7.62 GPa.

2.4. Recovery of Feldspar Minerals

Feldspars are important raw materials for the ceramic and glass industries, and the non-renewable mineral resources are increasingly being consumed. Granite waste contains a considerable amount of K-feldspar and Na-plagioclase, with an average content of 28.5% and 39.7%, respectively, making it a potential source of feldspar minerals [65, 66]. Beneficiation methods recover quartz, albite and potash feldspar from granite waste, and remove the colored impurities such as mica, iron oxides and ferro-titanium gangue. Using a combination of gravity separation and magnetic separation to reduce the content of colored impurities, the mechanical properties of sintered granite

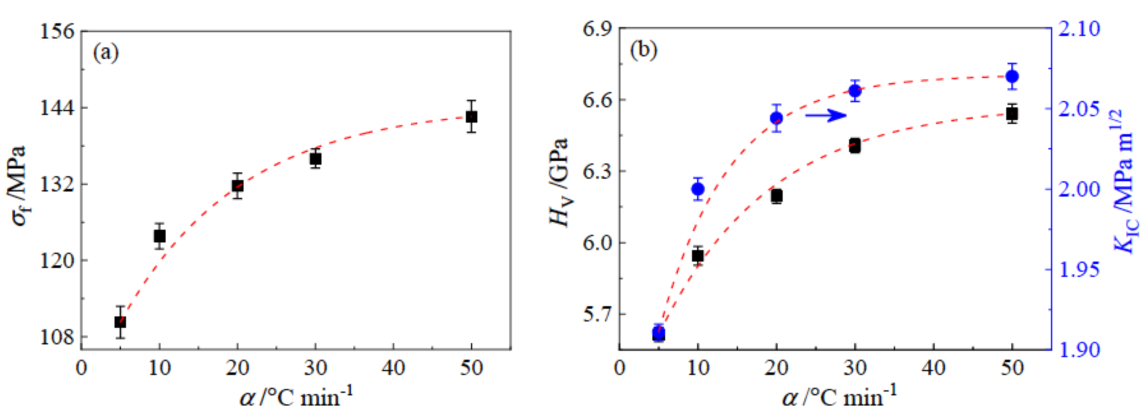

Figure 4: Effect of heating rate on the mechanical properties of sintered glass-ceramics. (**a**) flexural strength, (**b**) Vickers hardness and fracture toughness.

products are considerably better than natural granite [67].

Magnetic separation reduces the iron oxide content in granites to 0.2%. Froth flotation further removes mica and iron-titanium gangue, obtaining a feldspar/quartz concentrate with an iron content of 0.08% and with fewer impurities than magnetic separation. The concentrates obtained by both methods meet the requirements of the ceramics industry [68].

Cationic flotation technology has been used to separate Na- and K-feldspars from granite, with NaCl as the depressing agent to increase the K-feldspar grade in the concentrate [69], with recovery rates for orthoclase and albite at 70.18% and 28.27%, respectively. Figure **5** is a schematic of feldspar extraction from granite waste. It is notable that some critical raw materials, especially Li-mica and Nb-Ta-Ti mineral phases, can be recycled from the hydrothermally altered granite using a combination of gravity, magnetic and heavy liquid separation [70].

2.5. Environmental Protection

Urbanization leads to extensive changes in the use of agricultural land in urban areas, and diazonium fertilizers for vegetable plants has accelerated soil acidification [71]. The acidic wastewater discharged from mining operations is another cause of soil acidification [72]. The pH of acidic sandy soil is usually less than 4.5, while granite waste is alkaline (pH > 9) and can be used as an effective acid soil amendment [73]. The acidic environment increases the solubility of granite, so that the alkaline cations (Ca, Mg, Na and K) on the surface of the silicates are quickly released. It can be used as a source of plant nutrients in acidic soils. For example, K-feldspar can be used as a high-efficiency fertilizer following hydrothermal alteration or high-temperature calcination [74, 75].

Black cotton soil is decomposed black lava, but due to its high content of montmorillonite and large soil expansion, it cannot be directly used as a construction material. The addition of granite waste increases the plastic index and maximum dry density of black cotton soil; also, the California bearing ratio is increased, making it suitable as a road subgrade material [76]. Granite waste has also been used as a stabilizer in expansive, highly plastic soils. With 70% added granite waste, the swelling index of such soils has been found to decrease from 58.3% to 11% after curing for seven days [77].

Granite waste is used in wastewater treatment to remove specific pollutants. Adding the fungus *Aspergillus Niger* to granite waste removes phosphate ions from wastewater, suggesting a cheap and ecofriendly material for phosphate removal [78].

Iron-rich granite waste in the presence of hydrogen peroxide catalytically degrades organic orange dye by the solar photo-Fenton process [79]. The dye solution is completely decolorized and effectively mineralized, achieving 68.7% total organic carbon removal at pH 3.0. This indicates that granite waste is an efficient heterogeneous photo-Fenton catalyst. A mixture of granite powder and pine bark compost has a high hexavalent chromium (Cr(VI)) adsorption capacity and is used in sewage treatment as a permeable reactive barrier in groundwater remediation [80].

The addition of 15% granite sludge has been found to be sufficient to stabilize heavy metals in hazardous industrial sludge. The aluminosilicates or silicate matrix within the granite sludge transforms heavy metals in their insoluble hydroxides or adsorbed in the stabilized matrix [81].

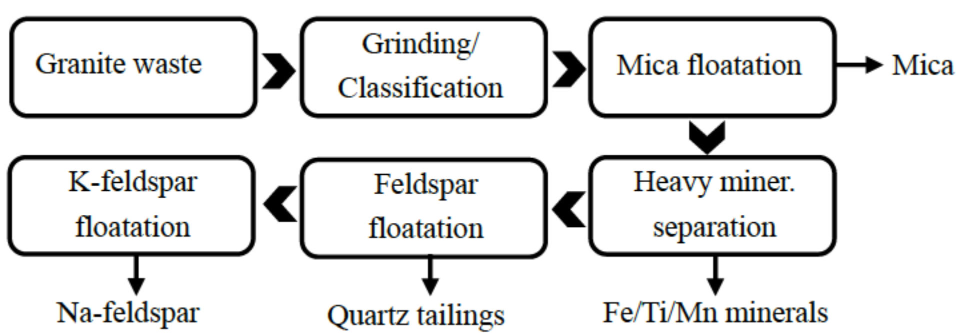

Figure 5: Flowchart of feldspar flotation from granite waste.

3. FUTURE OUTLOOK

Numerous studies have been conducted on the reutilization of granite waste. Its reutilization in building materials could recycle granite waste in large quantities. From the viewpoint of economic value, granite wastes are distributed at different processing sites, and the transportation cost is not acceptable for industrial-scale utilization. Because local governments attach much importance to environmental protection and mineral sustainability, corresponding policies and financial support should be put forward to promote cooperation between research institutes and related enterprises in exploring novel technologies for the scalable and value-added utilization of granite waste. In building materials, architectural glass-ceramics is a high-grade decorative material that has been developed in the past two decades. After colored impurities are removed, granite waste can be used to produce architectural glass-ceramics in order to improve its added value. To explore the utilization routes, granite waste can be used as the substitute for feldspar minerals, marble processing abrasives [82, 83], and soda lime glass raw materials [84]; and so on. Such lines of research favor the scalable and value-added utilization of granite waste.

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