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(54) Title: VOLCANO ASH SOLID GEOPOLYMER COMPOSITE AND A METHOD OF PRODUCING THE SAME

(57) Abstract: The present invention relates to a solid geopolymer composite comprising a pozzolanic mixture comprising volcano ash and an alkaline activator for the use as construction material and/or friction material. Besides, this invention relates to a method for producing a solid geopolymer composite from volcano ash.

VOLCANO ASH SOLID GEOPOLYMER COMPOSITE AND A METHOD OF PRODUCING THE SAME

FIELD OF INVENTION

The present invention relates to a solid geopolymer composite comprising a pozzolanic mixture containing volcano ash and an alkaline activator. In particular, the volcano ash is employed as artificial aggregates in preparation of construction material or friction material. Besides, the present invention also provides a method of producing the geopolymer composite.

BACKGROUND OF THE INVENTION

Geopolymer are essentially silicate and aluminosilicate materials linked with covalent bonds. Geopolymer composites are formed when silicate and aluminosilicate materials are physically or chemically blended with one or more materials to form materials with different mechanical properties. It is a new generation of material that can be used with fillers or be reinforced to suit different industrial needs. Nowadays, the common sources of silicate and aluminosilicate materials for geopolymer composites are pozzolanic materials such as fly ash, silica fume, metakaolin and rice husk ash. Pozzolanic materials are siliceous or siliceous and aluminous material that possess little or no cementitious properties. They are capable of reacting with calcium hydroxide and water to form compounds with cementitious properties. These pozzolanic materials are rendered as supplementary cementitious materials to substantially curb carbon dioxide emission by 80 to 90% from cement manufacturing processes. Besides, geopolymer composites formed from these pozzolanic materials are capable of withstanding extreme conditions such as being acid-resistant, fire-proof and heat-insulating. Hence they are widely used in automotive and aerospace industries, non-ferrous foundries and metallurgy, cements and concretes industries, ceramics and plastics industries and so forth.

Several geopolymer composites with different compositions are revealed in prior arts.

Chinese Patent No. 101844911 (A) introduces the composition of a composite that could replace cement which comprises kaolin, a mixture of sodium silicate and sodium hydroxide, and reinforcing fibers. Further, United States Patent No. 2012192765 (Al) discloses a geopolymer cement that includes metakaolin or a mixture of metakaolin and an activated aluminosilicate, an alkaline silicate solution and a superplasticizer. Nevertheless, the production of metakaolin requires high sintering temperature which increases the use of energy in manufacturing processes.

Korean Patent No. 100855686 (Bl) reveals a composite of cement replacement material consists of blast furnace slag and fly ash with alkaline inorganic material. Further, United States Patent No. 2011287198 uses slag or fly ash, and a sodium-free inorganic alkaline material as binder for mortar and concrete products. However, the sources of these materials became limited since the technology of geopolymer composite has been disclosed. Hence other sources are needed to cope with high demand of geopolymer composites.

SUMMARY OF THE INVENTION

The primary object of the present invention is to provide a solid geopolymer composite from volcano ash that is comparable to geopolymer composite formed from Class F fly ash.

Another object of the present invention is to provide a solid geopolymer composite from excessive volcano mud into a highly demanded construction material and/or friction material.

Yet another object of the present invention is to provide a solid geopolymer composite that could vary in mechanical properties by adjusting the ratio of volcano ash to an alkaline activator.

At least one of the preceding aspects is met, in whole or in part, by the present

invention, in which the embodiment of the present invention describes a composition of producing solid geopolymer composite comprising volcano ash and an alkaline activator, wherein the volcano ash is present in the range of 50-80% by weight of the pozzolanic mixture.

The present invention also provides a production method for a solid geopolymer composite comprises of reacting volcano ash with an alkaline activator, followed by curing the mixture to obtain a dried solid composite. The production method applied is simple and low in energy consumption.

Yet another object of the present invention is to provide a method that reduces carbon emission with the use of pozzolanic material to form a geopolymer composite.

DETAILED DESCRIPTION OF THE INVENTION

One skilled in the art will readily appreciate that the present invention is well adapted to carry out the objects and obtain the ends and advantages mentioned, as well as those inherent therein. The embodiment describes herein is not intended as limitations on the scope of the invention.

The present invention is a solid geopolymer composite for the use as construction and friction material. A solid geopolymer composite comprising a pozzolanic mixture containing volcano ash and an alkaline activator is disclosed. In the preferred embodiment, the pozzolanic material employed is volcano ash. The volcano ash used is proven to be comparable to Class F fly ash in silica, alumina and iron oxide composition. Hence similar to the use of Class F fly ash, volcano ash can be used as artificial aggregates in construction materials and/or friction materials.

The volcano ash used is derived from volcano mud acquired in vicinity of volcano. The volcano mud is dried, grinded and sieved to obtain volcano ash in preferred particle size. Volcano ash with finer particle size has higher surface area of reaction

during geopolymerization, hence it is able to form a geopolymer composite with greater strength stability. Preferably, the particle size of the volcano ash used is in a range of 10 - 500 μm. Further, sintered volcano mud can be utilized as volcano ash in the geopolymer composite. The content of silica and alumina in the sintered volcano mud is higher than the original volcano mud, therefore rendering it suitable for geopolymer composite. Nevertheless, the use of sintered volcano mud increases the energy usage during manufacturing process.

Preferably, the volcano ash is presented in the range of 50-80% by weight of the pozzolanic mixture to ensure a homogeneous mixture is acquired. In a condition where the composition of volcano ash in the pozzolanic mixture exceeds the preferred range, the pozzolanic mixture would be too saturated to cause a loss in workability of the geopolymer composite. On the contrary, a solid geopolymer composite would not be formed.

The present invention is a solid geopolymer composite from volcano ash that contains silica, alumina, metal oxides or a combination thereof. The existence of polymeric Si-O-Al sialate bonds in the employed volcano ash renders the volcano ash as an excellent pozzolanic material in the production of geopolymer composite.

In the preferred embodiment, the process of geopolymerizing is conducted by mixing the volcano ash with an alkali activator. The alkaline activator is a mixture of sodium silicate and an alkaline hydroxide, having a weight ratio in the range of 0.1 to 1.5: 1.5 to 3.0. The alkaline hydroxide is preferably sodium hydroxide. However, potassium hydroxide can also be used.

Further, the present invention discloses a method of producing a solid geopolymer composite comprised of reacting volcano ash with an alkaline activator to obtain a pozzolanic mixture and curing the mixture to obtain a solid composite. Preferably, the composition of volcano ash is presented in a range of 50 to 80% by weight of the

pozzolanic mixture. The mixture is cured at temperature preferably ranged from 150 to 1500 °C for 24 to 48 hours to obtain a solid composite for construction materials and/or friction materials.

The method further comprises a step of drying the volcano mud at 60 to 110 °C to obtain volcano ash before reacting the volcano ash with the alkaline activator. The method further comprises a step of grinding and sieving the volcano ash to obtain a particle size in a range of 10 to 500 μm. In particular, dried volcano mud is grinded into volcano ash of the preferred particle size by using a ball mill. Moreover, volcano ash with finer particle size is able to form a relatively more compact geopolymer composite for the fabrication of strong construction or friction material.

The following example is intended to further illustrate the invention, without my intent for the invention to be limited to the specific embodiments described therein.

Example 1

A solid geopolymer composite for a brake pad mixture is produced according to the amount of materials listed in Table 1. The mechanical properties of solid geopolymer composite for brake pad mixture is depicted in Table 2. Volcano mud is dried at 60 °C to obtain dried volcano mud. Dried volcano mud is then grinded and sieved to obtain volcano ash with a particle size of lower than 425 μm. A sodium hydroxide solution with concentration of 10 to 12M is prepared and mixed with sodium silicate to form an alkaline activator. The ratio of the sodium silicate to the sodium hydroxide used is 0.6. The alkaline activator is then mixed and reacted with volcano ash to obtain a homogeneous paste mixture. The mixture is then left to be cured at 150 °C for 24 hours.

Table 1 Amount of Materials for Producing a Brake Pad Mixture

Material	Volcano Ash	Sodium Silicate	Sodium Hydroxide
Weight (g)	600-700	100-200	150-250

Table 2 Properties of Materials for Producing a Brake Pad Mixture

Properties	Value
Density (bulk)	1800-2300 kg/m3
Water Absorption (%)	0-5
Specific Gravity	5-12
Compressive Strength (MPa)	5-35

Example 2

A solid geopolymer composite to be used as artificial aggregates is produced according to the amount of materials listed in Table 3. The mechanical properties of solid geopolymer composite for artificial aggregate mixture is depicted in Table 4. Volcano mud is dried at 60 °C for 48 hours to obtain dried volcano mud. Dried volcano mud is then grinded and sieved to obtain volcano ash with a particle size of lower than 425 μm. A sodium hydroxide solution with concentration of 12M is prepared and mixed with sodium silicate to form an alkaline activator. The ratio of the sodium silicate to the sodium hydroxide used is 0.6. The alkaline activator is then mixed and reacted with volcano ash to obtain a homogeneous paste mixture. The pellet is then left to be cured at 105 °C for 2 hours, followed by sintering at 400 to 1800 °C for 1 hour to form the artificial aggregate.

Table 3 Amount of Materials for Producing an Artificial Aggregate

Material	Volcano Ash	Sodium Silicate	Sodium Hydroxide
Weight (g)	1000	180 - 250	300 - 400

Table 4 Properties of Materials for Producing an Artificial Aggregate

Properties	Value
Density (Bulk)	400 - 2400 kg/m^3
Water Absorption (%)	0.1 - 45
Specific Gravity	0.5 - 3.5
Compressive Strength (MPa)	0.5 - 60

CLAIMS

1. A solid geopolymer composite comprising a pozzolanic mixture containing volcano ash and an alkaline activator.

2. A solid geopolymer composite according to claim 1, wherein the volcano ash used is derived from volcano mud.

3. A solid geopolymer composite according to claim 1, wherein particle size of the volcano ash used is in a range of 10 - 500 μm.

4. A solid geopolymer composite according to claim 1, wherein the volcano ash is present in the range of 50-80% by weight of the pozzolanic mixture.

5. A solid geopolymer composite according to claim 1, wherein the volcano ash contains silica, alumina, metal oxides or a combination thereof.

6. A solid geopolymer composite according to claim 1, further comprising the alkaline activator which is a mixture of sodium silicate and an alkaline hydroxide, having a weight ratio in the range of 0.1 to 1.5: 1.5 to 3.0.

7. A method for producing a solid geopolymer composite comprised of
reacting volcano ash with an alkaline activator to obtain a pozzolanic mixture; and curing the mixture to obtain a solid composite.

8. A method for producing a solid geopolymer composite according to claim 7, wherein the composition of volcano ash is present in a range of 50 to 80% by weight of the pozzolanic mixture.

9. A method for producing a solid geopolymer composite according to claim 7,

further comprising a step of drying the volcano mud at 60 to 110 °C to obtain volcano ash.

10. A method for producing a solid geopolymer composite according to claim 7, further comprising a step of grinding and sieving the volcano ash to obtain a particle size in a range of 10 to 500 μm.

INTERNATIONAL SEARCH REPORT

international application No.
PCT/MY2013/000201

A. CLASSIFICATION OF SUBJECT MATTER

C04B 7/12 (2006.01)

According to international Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and. where practicable, search terms used)

WPJ/EPODOC, STN CAPLUS. Search terms: geopolymer, silicate, volcano, activator, alkali and like terms.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	

[X] Further documents are listed in the continuation of Box C [X] See patent family annex

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Dare of mailing of the international search report
5 March 2014	05 March 2014
Name and mailing address of the ISA/AU	**Authorised officer**
AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct(<¾ipausrralia.gov.au Facsimile No.: +61 2 6283 7999	Andrew Matthews AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. 0262S32693

INTERNATIONAL SEARCH REPORT C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		International application No. PCT/MY2013/000201
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	LEMOUGNA et al "Synthesis and iheraial properties of inorganic polymers (geopolymers) for structural and refractory applications from volcanic ash." Ceramics International, 2011, Vol 37 pages 3011-3018. See whole of the document but particularly the Abstract, Fig. 1, Table 1.	1-10
X	KAMSEU et al " Investigation of volcanic ash based geopolymers as potential building materials." Interceram, 2009 Vol. 58 (2-3) pages 136-140. See whole of the document but particularly the Abstract, and 2.2 and Table 1.	1-10

INTERNATIONAL. SEARCH REPORT information on patent family members	International application No. PCT/MY2013/000201

This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose ofinformation.

Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	**Publication Date**	**Publication Number**	**Publication Date**

End of Annex

Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.