COMMONWEALTH OF AUSTRALIA
THE PATENTS ACT 1952
DECLARATION IN SUPPORT OF A
CONVENTION APPLICATION FOR A PATENT
In support of the Convention Application made for a patent for an invention entitled:

"FLOCATED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MADE THEREFROM"

Title of invention

I, Wil...Claude Leo Beaudoin
Full name(s) and address(es) of
Declarant(s)
of
508 Whitby Drive
Wilmington, DE 19803
United States of America

do solemnly and sincerely declare as follows:-

1. I am/we are the applicant(s) for the patent

(or, in the case of an application by a body corporate)

1. I am/we are authorised by

ARMSTRONG WORLD INDUSTRIES, INC.
the applicant(s) for the patent to make this declaration on

their behalf:

2. The basic application(s) as defined by Section 141 of the
Act was/were made

Basic Country(ies)
in United States of America
Priority Date(s)
on October 18, 1984

Basic Applicant(s)
by Thomas Michael Tymon

3. I am/we are the actual inventor(s) of the invention referred

to in the basic application(s) as defined by Section 141 of the
Act as the first application(s) made in a Convention
country in respect of the invention

 declared at Lancaster, PA this 17th day of June, 1985

THOMAS MICHAEL TYMON

of 515 West Chestnut Street, Lancaster, Commonwealth of
Pennsylvania 17603, United States of America,

(is/are the actual inventor(s) of the invention and the facts upon
which the applicant(s) is/are entitled to make the application are

as follows:

The said Company is the assignee of the actual inventor(s).

4. The basic application(s) referred to in paragraph 2 of this
Declaration was/were the first application(s) made in a Convention
country in respect of the invention(s) the subject of the application.

Declared at Lancaster, PA this 17th day of June, 1985

ARMSTRONG WORLD INDUSTRIES, INC.

By Claude Leo Beaudoin
Assistant Secretary

SFP4
To: The Commissioner of Patents

Signature of Declarant(s)
Claude Leo Beaudoin
Assistant Secretary

FORM 10

SPRUSON & FERGUSON
COMMONWEALTH OF AUSTRALIA
PATENTS ACT 1952

CONVENTION APPLICATION FOR A STANDARD PATENT

We ARMSTRONG WORLD INDUSTRIES, INC. of Liberty and Charlotte Streets, Lancaster, Pennsylvania 17604, United States of America hereby apply for the grant of a standard patent for an invention entitled:
"FLOCCED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MADE THEREFROM"

which is described in the accompanying complete specification.

DETAILS OF BASIC APPLICATION

Number of Basic Application: 662,057.

Name of Convention Country in which Basic Application was filed: United States of America

Date of Basic application: 18 October, 1984

Our address for service is:
C/- Spruson & Ferguson Patent Attorneys Level 33 St Martins Tower 31 Market Street Sydney New South Wales Australia

DATED this TWENTY-SEVENTH day of JUNE 1985

ARMSTRONG WORLD INDUSTRIES, INC.

By: [Signature]


TO: THE COMMISSIONER OF PATENTS AUSTRALIA

SBR: ep 241U

FLOCCED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MATERIALS MADE THEREFROM
1. A method of preparing a flocked mineral material that can be utilized to form a non-asbestos high temperature article that exhibits water resistance, which method comprises contacting a swelled layered silicate gel that has an average charge per structural unit that ranges from about -0.5 to about -1 and which contains exchangeable interstitial ions with at least one species of guanidine derived cations to thereby effect an ion exchange reaction between at least some of the exchangeable interstitial ions and at least some of the guanidine derived cations.

8. A flocked mineral material which comprises a swelled layer silicate gel that has an average charge per structural unit that ranges from about -0.5 to about -1, said silicate containing at least some interstitial cations that are guanidine derivates.

15. A high temperature, water resistant article that is prepared from a swelled layered silicate that has an average charge per structural unit that ranges from about -0.5 to about -1, said silicate containing at least some interstitial cations that are guanidine derivates.
20. A method of preparing a high temperature silicate article that exhibits water resistance, which method comprises contacting an article formed from gellable layered water-swelling silicate that has a charge per structural unit that ranges from about -.5 to -1 and which contains exchangeable interstitial ions with a source of at least one species of guanidine derived cations to thereby effect an ion exchange reaction between at least some of the guanidine derived cations and at least some of the interstitial ions.
FORM 10

COMMONWEALTH OF AUSTRALIA
PATENTS ACT 1952

COMPLETE SPECIFICATION

(ORIGINAL)
FOR OFFICE USE:

Class Int. Class

Complete Specification Lodged:

Accepted:

Published:

Priority:

Related Art:

Name of Applicant: ARMSTRONG WORLD INDUSTRIES, INC.

Address of Applicant: Liberty and Charlotte Streets, Lancaster,
Pennsylvania 17604, United States of America

Actual Inventor: THOMAS MICHAEL TYMON

Address for Service: Spruson & Ferguson, Patent Attorneys,
Level 33 St Martins Tower,
31 Market Street, Sydney,
New South Wales, 2000, Australia

Complete Specification for the invention entitled:

"FLOCCED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MADE THEREFROM"

The following statement is a full description of this invention, including the best method of performing it known to us

SBR:ep 241U
FLOCCED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MATERIALS MADE THEREFROM

Abstract

Disclosed are flocced mineral materials which may be utilized to prepare high temperature resistant, water resistant articles. These materials are prepared by utilizing, as a starting material, a gellable layered swelled silicate that has an average charge per structural unit that ranges from about -0.5 to -1 and which contains interstitial cations which promote swelling with a source of at least one species of guanidine derived cations.
FLOCCED MINERAL MATERIALS AND WATER-RESISTANT ARTICLES MADE THEREFROM

Background of the Invention

It is known that non-asbestos papers and/or sheets may be produced from water-swellable inorganic materials and, in particular, swelled silicate gels. For example, United States Patent No. 4,239,519 is directed to the preparation of inorganic, crystal-containing gellable, water-swelling sheet silicates and certain articles, such as papers, fibers, films, boards, and coatings, produced therefrom. These non-asbestos papers and/or sheets exhibit good high temperature stability and good chemical resistance. Furthermore, since asbestos fibers are not utilized in their manufacture, such articles will not have the health hazards which are associated with asbestos containing articles.

U. S. Patent 4,239,519 teaches the method for making the precursor gellable silicates used to produce said papers or sheet articles, as involving three fundamental steps: (a) a fully or predominantly crystalline body is formed which contains crystals consisting essentially of a lithium and/or sodium water-swelling mica selected from the group of fluorhectorite, hydroxyl hectorite, boron fluorphlogopite fluorphlogopite, hydroxyl boron phlogopite, and solid solutions between those and other structurally compatible species.
selected from the group of talc, fluortalc, poly lithium-nite, fluoropoly lithium nite, phlogopite, and fluor phlogopite; (b) that body is contacted with a polar liquid, normally water, to cause swelling and disintegration of the body accompanied with the formation of a gel; and (c) the solid:liquid ratio of the gel is adjusted to a desired value depending upon the application thereof. Glass-ceramics are the preferred crystalline starting bodies. Those products are then contacted with a source of large cations, i.e., with an ionic radius larger than that of the lithium cation, to cause macro flocculation of the gel and an ion exchange reaction to take place between the large cations and the Li⁺ and/or Na⁺ ions from the interlayer of the crystals.

Alternatively, U. S. Patents No. 3,325,340 and 3,454,917 teach producing aqueous dispersions of vermiculite flaked crystals which have been caused to swell due to the introduction therein of interstitial ions such as (1) alkylammonium cations having between 3 and 6 carbon atoms inclusive in each carbon group such as methylbutylammonium, n-butylammonium, propylammonium and iso-amylammonium, (2) the cationic form of amino acids, such as lysine and ornithine, and/or (3) lithium.

While the articles, such as papers, sheets and films, prepared via the prior art processes set forth above exhibit excellent heat resistance and are very useful in a wide variety of applications, it has been discovered that they exhibit a certain amount of water sensitivity which is generally exhibited by the articles having a considerable loss of strength and general deterioration of mechanical and electrical properties when exposed to high humidity environments or submerged in water or other polar liquids. This sensitivity to water correspondingly curtails the utility of these articles in certain applications, such as, for example, head gaskets, electrical insulators, environmental protective coatings, and washable and environmentally stable building materials.

12. The material of claim 10 wherein the polar liquid is water.
Summary of the Invention

It has now been unexpectedly discovered that high temperature, fire-resistant, non-asbestos, water-resistant articles, such as sheet, paper, board, film, fiber and coating articles, can be made from a swelled, layered flocced silicate gel material that is prepared by utilizing an exchange cation that is selected from guanidine derivatives. Such articles surprisingly have been found to exhibit, in general, much improved results in tensile strength and puncture resistant tests that are conducted when the articles are wet than do materials that are prepared utilizing prior art exchange cations. Furthermore, the articles made according to the present invention generally display superior electrical and mechanical properties over those materials made by prior art methods.

With reference to heat resistance, the articles that are produced according to the present invention are completely stable to temperatures of approximately 350-400°C and maintain their structural stability to approximately 800°C.

Detailed Description of the Invention

The articles and the flocced mineral suspensions of the present invention are, in one embodiment of the invention, prepared by utilizing, as a starting material, a water-swelling silicate that has an average charge per structural unit of from about 5 to about -1 and which contains interstitial exchangeable cations that promote swelling. The specific exchange cations in the starting material will depend on the silicate being utilized. For example, if a synthetically derived gellable silicate, which is made according to the procedures of U.S. Patent 4,239,519, is utilized as a starting material, the exchange cations will generally be Li⁺ and/or Na⁺ ions. If a natural vermiculite dispersion, such as made according to U.S. Patent 3,325,340, is utilized, the exchange cations will generally include alkylammonium cations and the other
cations specified in U. S. Patent 3,325,340. The silicate, whether synthetic or natural in origin, will generally have morphologies that are represented by thin flakes which are generally disc, strip, and/or ribbons. The flakes will have typical measurements which are from about 500 Å to 100,000 Å, and preferably 5,000 Å to 100,000 Å in length, 500 Å to 100,000 Å in width, and less than 100 Å thick. The term "charge per structural unit" as used in the specification and claims refers to an average charge density as specified by G. Lagaly and A. Weiss, "Determination of Layer Charge in Mica-Type Layer Silicates," Proceedings of International Clay Conference, 61-80 (1969) and G. Lagaly, "Characterization of Clays by Organic Compounds," Clay Minerals, 16, 1-21 (1981).

The starting silicate can be made according to the afore-mentioned procedures of U. S. Patent 4,239,519; 3,325,340; or 3,434,917 or other methods which result in dissociated layer materials with charge densities in the desired ranges.

The silicate is then contacted with a source of at least one species of guanadine derived cations to thereby effect an ion exchange reaction to occur between the cations and the interstitial ions. This ion exchange reaction may be carried out between the cations and the silicate material to thereby form a floc which is then utilized to form the articles of the present invention. In another embodiment of this invention, the starting silicate can be directly formed into a product, such as a lithium fluorhectorite fiber or film by using the procedures fo U. S. Patent 4,239,519, and a cationic exchange reaction utilizing the guanadine derived cations can be carried out with the product, such as by immersing the product into a solution of guanadine derived cations. Thus, the ion exchange reaction may be carried out in situ during the actual forming process for the product.

Exchange cations that may be utilized in the
present invention will be derived from compounds that correspond to the Formula

\[ R_1 \]
\[ R - C - R_2, \text{ wherein } R, R_1 \text{ and } R_2 \]

5 \( R_2 \) are individually selected from \(-\text{NH}_2\) and \(-\text{CH}_3\), with the proviso that at least two of \( R, R_1 \) and \( R_2 \) are \(-\text{NH}_2\) and, furthermore, wherein one or more of the hydrogens on \( R, R_1 \), and/or \( R_2 \) can be replaced by substituents such as \( \text{C}_1-\text{C}_5 \) alkyl, \( \text{C}_1-\text{C}_5 \) alkenyl, \( \text{C}_1-\text{C}_5 \) alkynyl and, wherein further one or more groupings of two of such substituents can join to form rings which may be optionally aromatic.

The flocced mineral suspensions of the present invention are prepared, for example, by reacting, generally with agitation, a suitable silicate gel with a source of exchange cations derived from the guanidine compounds set forth in the Formula above in order to effect an ion exchange between the guanidine derived cations and the interstitial cations in the silicate gel to form exchanged macro flocculated particles. For example, if the exchange cation of choice is guanidinium or melaminium, the silicate will be reacted with the corresponding hydrochloride.

As stated above, one or more exchange cations that are derived from the Formula above can be utilized in the cationic exchange reaction. Since the various cations will give floc, and eventually end products, with differing physical properties, the specific cation or combination of cations will be chosen by the practitioner of this invention based on the desired end use.

The flocced mineral suspension will be used to form the desired end products. The specific treatment steps applied to the floc will depend on the particular article being formed. For example, if the articles of
the present invention are to be formed into sheet materials, the resultant exchanged floc will be agitated with sufficient shear to produce a particle size distribution which leads to suitable particle packing in the sheet forming operation. Following this process the floc is optionally washed to remove any excess salt solution and the consistency of the flocced slurry is adjusted to from about 0.75% to about 2% solids. To promote better drainage rates on a fourdrinier wire, polyelectrolyte flocculating agents can then be added to the slurry at a level of from about 0.1% to about 1%, and preferably 0.2%-0.3% of floc solids. One example of a suitable polyelectrolyte flocculating agent is Polymin P, which is a trademark of BASF Corporation for a polyethylene imine.

This slurry is then fed to a papermaking apparatus where it is dewatered by free drainage and/or vacuum drainage followed by pressing and drying on drum dryers. The thus formed sheet material can be used in applications such as gaskets and the like.

If desired, and depending on the intended end use of the product, additional inert materials may be added to the flocced mineral suspension. For example, if desired one or more fibrous materials from the group of natural or synthetic organic fibers or inorganic fibers may be added to the floc to improve its drainage rate and to provide an end product that has improved strength and/or handleability. For example, when the desired end products are gaskets, the fibers of choice are cellulose fibers, glass fibers, and/or Kevlar fibers (Kevlar is a trademark of DuPont Corporation for an aromatic polyamide fiber). In addition, latex or other binders may be added to the floc to provide for a product with improved strength characteristics.

If the cationic exchange reaction is conducted directly on a product formed from the silicate starting material, any desired additional inert materials would be added to the slurry of the silicate starting material
prior to the formation of the product and, of course, the subsequent cationic exchange reaction.

The term "water resistant" as used in the specification and claims is not meant to imply that the articles of the present invention are waterproof or are completely impervious to water. By contrast, the term is used to indicate that the materials do not substantially degrade, at least in their tensile strength and puncture resistant properties, when exposed to water.

In these following Examples, unless otherwise specified, the starting material utilized was a lithium fluorhectorite made according to procedures taught in U. S. Patent No. 4,239,519.

Example 1

This example illustrates a method of producing both a guanidinium exchanged fluorhectorite flocced silicate and a formed sheet that was prepared therefrom.

A slurry of guanidinium fluorhectorite was prepared by adding 475 grams of a 10% dispersion of lithium fluorhectorite to 1.4 liters of 1M guanidine hydrochloride solution. The slurry was then agitated with a high shear mixer to reduce the particle size of the resultant floc, was washed and then was analyzed for water content and diluted to result in a 2% solids slurry. The slurry was transferred to a 11.5" x 11.5" hand sheet mold (manufactured by Williams Apparatus Co.) and dewatered. The resultant formed sheet was then wet pressed and dried on a drum drier. The sheet had good flexibility and performed well in the gasket sealing test.

Example 2

This example illustrates a method of producing films of the present invention wherein the cationic exchange is made in situ.

A 10% solids lithium fluorhectorite gelled dispersion was prepared according to procedures taught in U. S. Patent No. 4,239,519. A film was made of this material by using a 4.5 mil Byrd applicator, which was 5
inches wide, to draw down a 4½ mil thick wet film of the dispersion on a glass plate. The glass plate, with the film attached, was then immersed in a .25M guanidinium hydrochloride solution to cause a cation exchange between the guanidinium cations and the fluorhectorite's interlayer cations. A skin was formed, seemingly instantaneously, on the film which indicated such an exchange was taking place. In 10 minutes the film was removed from the plate, washed in deionized water to remove residual salts, and dried. The film had good flexibility and strength retention when wet.

Examples 3-9

For each of these examples, the procedure of Example 2 was substantially repeated with the exchange cation as specified to form the corresponding film. In Example 7, a 0.1N solution of melamine hydrochloride was employed. In all the other examples, a .25N solution of the respective exchange source was employed:

<table>
<thead>
<tr>
<th>Example</th>
<th>Exchange Cation</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>Diaminoguanidine hydrochloride</td>
</tr>
<tr>
<td>4</td>
<td>Aminoguanidine hydrochloride</td>
</tr>
<tr>
<td>5</td>
<td>Tetramethylguanidine hydrochloride</td>
</tr>
<tr>
<td>6</td>
<td>Methylguanidine hydrochloride</td>
</tr>
<tr>
<td>7</td>
<td>Melamine hydrochloride</td>
</tr>
<tr>
<td>8</td>
<td>2,6-diaminopyridine hydrochloride</td>
</tr>
<tr>
<td>9</td>
<td>2-aminopyridine hydrochloride</td>
</tr>
</tbody>
</table>

Comparative Examples 1-3

These comparative examples illustrate fluorhectorite films that are made with various prior art exchange cations. Four and one half mil thick films of potassium fluorhectorite (KPH) and ammonium fluorhectorite (NH₄FH) were separately prepared by the process specified in U. S. Patent No. 4,239,519. A film was then cast of both the KPH and a NH₄FH slurry. A Kymene (a trademark of Hercules, Inc. for a cationic,
polyamide–epichlorohydrin resin) fluorhectorite film was also prepared by the procedure of Example 2, except that (1) a 3.0% Kymene solution was used and (2) the lithium fluorhectorite film had to be immersed in the Kymene solution for 2 hours until the resultant exchanged film was sufficiently self-supporting to be removed from the glass plate. These films, along with the films made in Examples 2–9, were then subjected to tensile strength and puncture resistance tests which were conducted as follows:

**Tensile Strength Measurements**

Dry tensile strength measurements were determined using an Instron at 1½" jaw separation and 0.2"/min. crosshead speed. Wet strength measurements were made by bringing water-saturated sponges in contact with both sides of the film sample for 10 seconds while the sample was positioned in the Instron clamps just before the strength test was conducted.

**Puncture Resistance Measurements**

Sample of film was secured in a retaining device which held the film securely. A stylus which could be loaded was impinged on the film in the direction normal to the surface of the film and loaded with increasing weight until the stylus penetrated the film. In the wet test the film in the retaining device was submerged in deionized water for 10 seconds immediately proceeding the puncture resistance test. The data from these tests is shown in the table below.

<table>
<thead>
<tr>
<th>Example #</th>
<th>Cation</th>
<th>Tensile Strength (psi)</th>
<th>Puncture Resistance (g/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Dry</td>
<td>Wet</td>
</tr>
<tr>
<td>2</td>
<td>Guanidinium</td>
<td>14,000</td>
<td>9,000</td>
</tr>
<tr>
<td>3</td>
<td>Diaminoguanidinium</td>
<td>13,000</td>
<td>11,000</td>
</tr>
<tr>
<td>4</td>
<td>Aminoguanidinium</td>
<td>13,000</td>
<td>11,000</td>
</tr>
<tr>
<td>Film of Example</td>
<td>Exchange Cation</td>
<td>Tensile Strength (psi) Dry</td>
<td>Wet</td>
</tr>
<tr>
<td>-----------------</td>
<td>-----------------</td>
<td>--------------------------</td>
<td>-----</td>
</tr>
<tr>
<td>5</td>
<td>Tetramethylguanidinium</td>
<td>11,000 11,000</td>
<td>13,000</td>
</tr>
<tr>
<td>6</td>
<td>Methylguanidinium</td>
<td>5,200 2,800</td>
<td>6,600</td>
</tr>
<tr>
<td>7</td>
<td>Melaminium</td>
<td>19,000 20,000</td>
<td>10,000</td>
</tr>
<tr>
<td>8</td>
<td>2,6-Diaminopyridine (protonated)</td>
<td>13,000 5,300</td>
<td>7,900</td>
</tr>
<tr>
<td>10</td>
<td>2-Aminopyridine (protonated)</td>
<td>11,000 7,000</td>
<td>7,800</td>
</tr>
<tr>
<td>15</td>
<td>Kymene (protonated)</td>
<td>7,000 2,700</td>
<td>900</td>
</tr>
<tr>
<td>1</td>
<td>Ammonium</td>
<td>3,300 1,400</td>
<td>3,500</td>
</tr>
<tr>
<td>2</td>
<td>Potassium</td>
<td>1,100 200</td>
<td>3,300</td>
</tr>
</tbody>
</table>

The data indicates that the films made according to the procedures of the present invention have markedly superior wet tensile strength and/or superior wet puncture resistance when compared to prior art compositions.

**Fire and Smoke Resistance**

A film prepared according to Example 2 was, after being dried, subject to fire and smoke resistant tests in accordance to the procedures specified in ASTM-E-662-79. Three separate tests were made and the results are set forth below. The numerical values correspond to the maximum specified optical density as per N.B.S. Technical Note #708.
Test #  |  Flaming DM Corr |  Smoldering DM Corr |
--------|-----------------|---------------------|
1       | 2               | 0                   |
2       | 1               | 0                   |
3       | 1               | 0                   |

Electrical Properties

Films of Examples 2 and 7 and Comparative Example 3 were, when dried, tested for dielectric strength using the procedures of ASTM D149. The results are set forth below.

<table>
<thead>
<tr>
<th>Films of</th>
<th>Dielectric Strength (v/mil)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Example 2</td>
<td>5,000</td>
</tr>
<tr>
<td>Example 7</td>
<td>9,000</td>
</tr>
<tr>
<td>Comparative Example 3</td>
<td>2,920</td>
</tr>
</tbody>
</table>

Comparative Examples 4 and 5

These examples illustrate using, as a starting material, silicate materials which fall outside the scope of the present invention in their charge per structural unit and their physical measurements.

For comparative Example 4, a 10% aqueous dispersion was made from a natural hectorite obtained from the source clay minerals depository of the Clay Minerals Society, Bloomington, Indiana. For Comparative Example 5, a 10% aqueous dispersion utilizing sodium montmorillonite, which was obtained from the same source. In each example, a film was drawn down using the procedures set forth in Example 2. The glass plates were then immersed for 10 minutes in a 0.25 V guanidine hydrochloride solution. In both instances, a coherent film was not produced.

Example 10

This example illustrates a method of preparing a film of the present invention utilizing a vermiculite starting material:

A 10% solids suspension of n-butylammonium vermiculite, which was prepared according to the
procedures specified in U. S. Patent 3,325,340, was cast as a film on a glass plate according to the procedure set forth in Example 2. The glass plate, with the film attached, was immersed for 10 minutes in a 0.25 M guanidinium hydrochloride solution. The resulting film was removed from the plate, washed, and dried. The film displayed wet strength in the tensile strength and puncture resistance tests that a comparable unexchanged vermiculite film does not display.

Example 11

This example illustrates preparing fibers utilizing the method of the invention. A 15% solids suspension of lithium fluorhectorite (prepared as above) was extruded through an 11 mil opening needle into a 2N solution of guanidine hydrochloride. The extruded fiber was carried by a porous belt and delivered to a second bath of 2N guanidine hydrochloride. The fiber so produced was washed via submersion in deionized water and dried. The resultant fiber was strong and flexible.
The following statement is a full description of this invention, including the best method of performing it known to us.

SBR:ep 241U.
WHAT IS CLAIMED IS:

1. A method of preparing a flocced mineral material that can be utilized to form a non-asbestos high temperature article that exhibits water resistance, which method comprises contacting a swelled layered silicate gel that has an average charge per structural unit that ranges from about -.5 to about -1 and which contains exchangeable interstitial ions with at least one species of guanidine derived cations to thereby effect an ion exchange reaction between at least some of the exchangeable interstitial ions and at least some of the guanidine-derived cations.

2. The method of claim 1 wherein the gelled layered silicate is a synthetic gellable silicate and the interstitial ions are Li⁺ and/or Na⁺.

3. The method of claim 2 wherein said synthetic silicate is prepared by contacting a body consisting essentially of crystals of a water-swelling mica selected from the group of fluorhectorite, hydroxyl hectorite, boron fluorphlogopite, hydroxyl boron phlogopite, and solid solutions among those and between those and other structurally compatible species selected from the group of (alc, fluortalc, polythionite, fluopolythionite, phlogopite and fluorphlogopite, with a polar liquid for a time sufficient to cause swelling of the crystals accompanied with the formation of a gel.

4. The method of claim 3 wherein the crystals are fluorhectorite.

5. The method of claim 3 wherein the polar liquid is water.

6. The method of claim 1 wherein the silicate
is vermiculite and the interstitial ions are alkylammonium cations, the cationic form of amino-acids and/or Li+.

7. The method of claims 2 or 6 wherein the guanidine derived cations are selected from the group of diaminoguanidine, tetramethyl guanidine, guanidine, aminoguanidine, methyl guanidine and melamine derivatives.

8. A flocced mineral material which comprises a swelled layer silicate gel that has an average charge per structural unit that ranges from about -.5 to about -1, said silicate containing at least some interstitial cations that are guanidine derivates.

9. The material of claim 8 wherein the silicate is synthetically derived.

10. The material of claim 9 wherein said silicate is prepared by (1) contacting a body consisting essentially of crystals of water-swelling mica containing interstitial lithium and/or sodium cations, said mica selected from the group of fluorhectorite hydroxyl hectorite, boron fluorphlogopite, hydroxyl boron phlogopite, and solid solutions among those and between those and other structurally compatible species selected from the group of talc, fluortalc, polyliithionite, fluorphylithionite, phlogopite and fluorphlogopite, with a polar liquid for a time sufficient to cause swelling of the crystals accompanied with the formation of a gel, and (2) contacting the thus formed gel with at least one species of a cationic guanidine derivative to thereby effect an ion exchange reaction between at least some of the lithium and/or sodium cations and at least some of the guanidine derived cations.

11. The material of claim 10 wherein the crystals are fluorhectorite.
12. The material of claim 10 wherein the polar liquid is water.

13. The material of claim 8 wherein the silicate is vermiculite.

14. The material of claims 9 or 13 wherein the interstitial cations that are guanidine derivatives are selected from the group of diaminoguanidine, tetramethyl guanidine, guanidine, aminoguanidine, methyl guanidine, and melamine derivatives.

15. A high temperature, water resistant article that is prepared from a swelled layered silicate that has an average charge per structural unit that ranges from about .5 to about -1, said silicate containing at least some interstitial cations that are guanidine derivatives.

16. The article of claim 15 that is prepared from a silicate floc.

17. The article of claims 15 or 16 which further is a sheet material.

18. The article of claims 15 or 16 which further is a fiber.

19. The article of claims 15 or 16 which further is a film.

20. A method of preparing a high temperature silicate article that exhibits water resistance, which method comprises contacting an article formed from a swellable layered water-swelling silicate that has a charge per structural unit that ranges from about -.5 to -1 and which contains exchangeable interstitial ions with a source of at least one species of guanidine.
8 derived cations to thereby effect an ion exchange
9 reaction between at least some of the guanidine derived
10 cations and at least some of the interstitial ions.

DATED this TWENTY-FIFTH day of JUNE 1985

ARMSTRONG WORLD INDUSTRIES, INC.

Patent Attorneys for the Applicant
SPRUSON & FERGUSON