Patented Jan. 11, 1938 2,104,841

UNITED STATES PATENT OFFICE

2,104,841

REFRACTORY ARTICLES

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No Drawing. Application July 11, 1936, Serial No. 90,258

10 Claims. (Cl. 106—9)

This invention relates to refractory articles, such as crucibles, melting pots, and the like articles, more particularly to those composed of graphite, or of graphite or silicon carbide, and similar refractory materials, and it is among its objects to provide improved working qualities in the batches used for such purposes; to provide for greater resistance to oxidation at elevated temperatures; and to provide improved high temperature strength in the finished articles. Other objects will appear from the following description.

The invention is predicated on my discovery that material and unexpected advantages attend the use of talc, steatite, and other hydrous magnesium silicates similar to them, in the production of refractory articles from batches containing flaky or granular materials, such as flake graphite and silicon carbide, both of which may be considered to be granular, at least as far as concerns this invention.

A major advantage of the invention is that by the use of talc, steatite, or the like in graphite or graphite-silicon carbide refractory batches the strength of the finished article at high temperatures is materially improved as compared with a similar batch not containing talc or steatite, and at the same time the resistance of the article to oxidation at elevated temperatures is increased. I do not now understand the action of these materials in affording improved strength, which is a result contrary to what would have been expected because talc and steatite, being hydrous magnesium silicates, are not especially refractory. That is, they tend to melt, or to form substantial amounts of a liquid phase, at relatively low temperatures, and in the ceramic art such a phenomenon is usually not accompanied by particularly high strength. I now believe that the action in protecting the articles against elevated temperatures results from the formation at the surface of the article of a type of glaze arising from the use of the talc or steatite.

A further advantage which is particularly important from the manufacturing standpoint is that with these batches containing granular or flaky materials the use of talc or steatite confers a real and marked improvement in the working characteristics of the batch. Owing, apparently, to the granular character of materials such as flake graphite and silicon carbide, batches containing them are normally difficult to work and shape. The practice up to the present time in the manufacture of carbon-bonded crucibles and the like from such batches has been to use expensive molds and hydraulic presses, or special spinning machines, rather than the types of machine equipment ordinarily used in the manufacture of similar articles from, for example, clay compositions. The use of such special and expensive apparatus has been necessitated by the aforesaid difficulty of satisfactorily working and forming the batches.

I have discovered, however, that the use of talc or steatite, even in rather small amounts, so improves the working characteristics of these difficultly workable mixtures that they can be worked and shaped with ordinary machines, thus rendering unnecessary the special molds, hydraulic presses, and spinning machines that have heretofore been necessary.

Although the invention is not limited thereto, it is applicable particularly to the manufacture of carbon-bonded articles, both those of a temporary nature and also those which provide a residual carbon bond after firing, and it will be further described with particular reference thereto.

Since talc and steatite are equally applicable for the purposes of the invention, reference hereinafter to either will be understood to comprehend both of them as well as their equivalents.

I have discovered further that additional benefits, in so far as strength is concerned, are had by the conjoint use of talc and metals, such as ferrosilicon, silicon, ferromanganese, ferromanganese silicon, and the like. Thereby very substantial increase in strength results, even up to almost double that of a batch containing the same amounts of graphite, silicon carbide and binder but no talc and metal. This beneficial effect is attained without depreciating the effect of talc in improving the working characteristics and in providing a protective glaze which reduces oxidizing attack of the graphite at high temperatures.

As evidencing the improvements obtainable in the practice of the invention, reference may be made to tests in which various graphite-silicon carbide refractory compositions adapted to the manufacture of crucibles, pots, and the like were made up and formed into test bars which after the customary finishing operations were subjected to test to determine their modulus of rupture at 2200° F. The compositions were as follows:

Body No.	1	2	3	4
Flake graphite	35	35	35	35
Silicon carbide (16 mesh and finer)	20	20	20	20
Silicon carbide (150 mesh and finer)	10	10	10	10
Pulverized pitch	15	15	15	15
Talc (150 mesh and finer)	15	0	20	10
Ferrosilicon (100 mesh and finer)	5	5	0	5
Feldspar (150 mesh and finer)	0	15	0	5
Modulus of rupture #/in² (2200° F.)	802	488	593	728

Composition 1 of the foregoing table is indicative of the strength characteristic obtainable in the preferred practice of the invention using both

talc and ferrosilicon, and the batch containing no feldspar. Composition 2 represents the same material in which the talc has been replaced by feldspar, which latter has been used in some types of batch used for these purposes. Comparison of the two shows that the strength has been greatly increased by the use of talc in place of feldspar in accordance with this invention.

Composition 3 is comparable to Composition 1 except that the ferrosilicon has been replaced by talc. Its strength is substantially greater than that of Composition 2, not containing talc, and while not as great as that of Composition 1, containing both talc and ferrosilicon, it shows that talc alone is capable of substantially increasing the strength, and a comparison of the three shows the benefits derivable from both talc and ferrosilicon.

Composition 4 represents a batch containing both talc and feldspar, the talc being less than that of Compositions 1 and 3. Its strength approaches that of Composition 1 and is much greater than that of Compositions 2 and 3. It shows in comparison with Composition 2 how through the use of talc feldspar can be used while still attaining the high strengths which characterize the invention. The use of feldspar may in some instances be desirable to confer refractory qualities of some particular degree. And as compared with Composition 3 it again shows the value of the combination of talc and ferrosilicon.

As exemplifying a further aspect of the invention, another composition was made up containing 35 per cent of flake graphite, 20 per cent of silicon carbide (16 mesh and finer), 10 per cent of silicon carbide (150 mesh and finer), 20 per cent of pulverized pitch, 10 per cent of talc (150 mesh and finer), 2.5 per cent of ferrosilicon (100 mesh and finer), and 2.5 per cent of 78 per cent ferromanganese (100 mesh and finer). This composition exhibited a modulus of rupture at 2200° F. of 588 pounds per square inch, showing how other metals than ferrosilicon combine with the use of talc to give high strengths.

The invention is applicable generally to refractory batches for making crucibles and similar articles from graphite, preferably graphite-silicon carbide compositions, and the exact contents and proportions of these and other constituents of the batch, such as binder and the like, will be dependent upon various characteristics desired in the final product, as will be understood by those skilled in the art. However, I now prefer for most purposes to use flake graphite, such as Madagascar graphite or its equivalent, between about 25 to 55 per cent, and from 10 to 55 per cent of silicon carbide which may, for many purposes, comprise about 5 to 30 per cent of silicon carbide grain approximately 16 mesh and finer, with from 5 to 25 per cent of silicon carbide fines 150 mesh and finer.

For most purposes in achieving the benefits of the invention talc or steatite, or the like, is used in an amount constituting from about 3 to about 20 per cent. A talc which has given satisfactory results is of the following composition:

	Percent
SiO_2	56.54
R_2O_3	1.04
MgO	30.74
CaO	6.25
Ignition loss	4.60
Combined water	2.27
CO_2	0.33
MnO	Trace

A steatite suitable for use in the practice of the invention is one having the following analysis:

	Percent
SiO_2	63.29
MgO	27.13
FeO	4.68
Fe_2O_3	1.25
Al_2O_3	1.24
CaO	Trace
Water	4.40

While various bonding agents may be used, it is preferred for most purposes to use a carbon bond such as coal tar pitch, coal tar, goulac (sulphite pitch), dextrine, or other carbonaceous binders affording a temporary binder and productive of a residual bond when the article is burned. Such agents may be used in amounts from about 5 to 30 per cent, or such as to provide about 3 per cent of residual carbon bond. Another binder that may be used is sodium silicate, say in amount up to 4 per cent.

The ferrosilicon may be present in an amount between 0.5 and 6 per cent. Elemental silicon may also be used in its place, and other metals exerting an action similar to that of ferrosilicon, such as ferromanganese, may be used with it or in place of it. If ferromanganese or ferromanganese silicon is added it may be used in an amount, say, up to about 6 per cent.

As indicated hereinabove, the refractoriness of the article may be varied, and to this end feldspar may be added to the batch but for most purposes it is preferred that this constituent shall not exceed about 10 per cent.

The graphite may suitably pass a 14-mesh sieve and be equally divided on 20, 30, 40 and 60 mesh, with the fines removed through 80 mesh.

According to the provisions of the patent statutes, I have explained the principle and manner of practicing my invention, and have described what I now consider to represent its best embodiment. However, I desire to have it understood that, within the scope of the appended claims, the invention may be practiced otherwise than as specifically described.

I claim:

1. A batch for forming refractory articles comprising talc, flake graphite, silicon carbide, and binder.

2. A batch for forming refractory articles comprising talc, flake graphite, silicon carbide, and binder providing residual carbon bond upon firing.

3. A refractory article, such as a crucible, pot, or the like, comprising talc, graphite, silicon carbide, and residual carbon bond.

4. A batch for forming refractory articles comprising about 25 to 55 per cent of graphite, about 10 to 55 per cent of silicon carbide, about 3 to 20 per cent of talc, and about 5 to 30 per cent of binder.

5. A batch for forming refractory articles comprising about 25 to 55 per cent of graphite, about 10 to 55 per cent of silicon carbide, about 3 to 20 per cent of talc, and about 5 to 30 per cent of carbonaceous binder providing residual carbon bond upon firing.

6. A refractory article, such as a crucible, pot, or the like, comprising about 25 to 55 per cent of graphite, about 10 to 55 per cent of silicon carbide, about 3 to 20 per cent of talc, and residual carbon bond.

7. A batch for forming refractory articles comprising granular refractory selected from the

group flake graphite and silicon carbide, talc, and a strength-improving metal.

8. A batch for forming refractory articles comprising graphite, silicon carbide, talc, and a strength-improving metal.

9. A batch for forming refractory articles comprising about 25 to 55 per cent of graphite, about 10 to 55 per cent of silicon carbide, about 3 to 20 per cent of talc, and a metal of the group ferrosilicon, silicon, ferromanganese, and ferromanganese silicon in a substantial and effective amount not exceeding about 6 per cent.

10. A batch for forming refractory articles comprising about 25 to 55 per cent of graphite, about 10 to 55 per cent of silicon carbide, about 3 to 20 per cent of talc, about 5 to 30 per cent of carbonaceous binder, and a metal of the group ferrosilicon, silicon, ferromanganese, and ferromanganese silicon in a substantial and effective amount not exceeding about 6 per cent.

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