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F. J. TONE

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LOW PERMEABLE REFRACTORY AND METHOD OF MAKING THE SAME

Original Filed Aug. 10. 1927

*Fig. 1.*

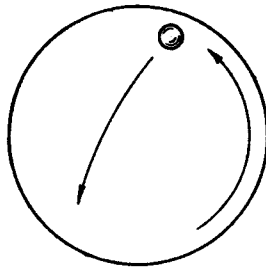


*Fig. 2.*

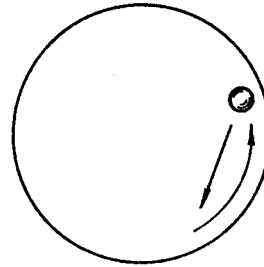


*Fig. 3.*

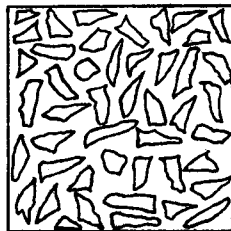
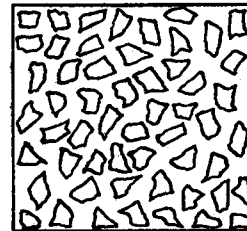


*Fig. 4.*



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## UNITED STATES PATENT OFFICE

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## LOW PERMEABLE REFRACTORY AND METHOD OF MAKING THE SAME

Continuation of application Serial No. 212,139, filed August 10, 1927. This application filed May 26, 1930. Serial No. 455,716.

This invention relates to improved refractory articles, particularly those composed of silicon carbide, and to a method of producing such articles of minimum permeability. This application is a continuation of my copending application, Serial No. 212,139, filed August 10, 1927.

The presence of permeable pores in a refractory is usually undesirable, and in many cases is very harmful. The permeability resulting from pore space is especially injurious in the case of refractories such as silicon carbide, where the grain is oxidized at high temperatures. The penetration of both gases and slags into the pores of the material greatly accelerates its destruction.

While the closing of the pores at the surface of the brick by various means, such as those described in United States Patent No. 1,473,286 to E. B. Forse, offers some relief in this respect, the relief is temporary and the advantage is lost as soon as the original facing of the brick has been destroyed to a very slight depth. My improved process involves the production of a non-permeable structure extending throughout the body of the refractory and independent of any surface glazing, although the presence of the latter may also be desirable.

In the manufacture of a non-permeable refractory, I take a suitable mixture of grits and bond in accordance with any of the various practices well known in the art, such as described in the United States Patents, Nos. 772,262 and 1,204,211 issued to Tone and 1,439,286 issued to Beecher. However, I employ a type of grain which has been submitted to a process which I shall designate as "mulling". I define "mulling" as the process of removing the rough corners from the grain and breaking up the needle shaped particles or splinters, without greatly reducing the average grain size of the material.

The "mulling" operation as used in my process of making a refractory is distinct from the usual operations of crushing and grinding. In grinding a substance in a tube mill, for example, the mill is revolved at such a speed that the pebbles are carried to the top of the mill by centrifugal force,

whereupon they drop by the action of gravity to produce the grinding or crushing effect without contact with the material on the sides of the mill until they strike at the bottom of their fall. The degree to which particles are crushed in a mill such as a tube mill or ball mill depends primarily upon the energy which is possessed by the balls or pebbles at the time of impact. If this energy is sufficiently great, the particle may be completely crushed, but if the energy is somewhat less, the particle may merely fracture along a plane of mechanical weakness, or chip at the edges or at sharp projecting corners. In "mulling", the mill is rotated at a slower speed in order that the particles may be smoothed and the splinters broken by the action usually known as attrition.

In the preferred method of carrying out my process, the material is first ground or pulverized to the desired grain size. The grain is then "mulled" in a suitable mill where the particles can be smoothed and brought to a more regular shape by attrition. As an example, a tube mill may be used, the mill being rotated at such a rate that the material slides down the sides of the mill in contact with the pebbles without cascading. The corners of the particles are thus worn off and the splinters broken without appreciably affecting the average grain size.

If a tube mill is used for the mulling operation, the mill is rotated at approximately two thirds the speed of that ordinarily used for crushing. As an example, a speed of 15 to 20 revolutions per minute is sufficient for mulling in the case of a mill that would normally operate at 25 to 30 revolutions per minute for crushing or grinding. The feed is preferably somewhat faster than in the usual crushing or grinding operations, and the balls or pebbles may be somewhat smaller than those used for crushing. By adjusting the conditions as described above, it is possible to secure a grain in which the corners and sharp edges are broken off, and in which the splinters are broken up and the number of irregular shaped particles reduced to a minimum.

In the drawing,

Figure 1 shows the approximate path of the balls or pebbles in the usual crushing operation in a ball or tube mill,

6 Figure 2 illustrates the path of the pebbles during the "mulling" operation,

10 Figure 3 shows the particles of silicon carbide grain after they have been crushed and screened to a particular grain size or mesh, and

Figure 4 shows the particles of silicon carbide grain after they have been mulled and screened to the same mesh as the particles shown in Figure 3.

15 In the "mulled" grain the dimensions are nearly equal in all directions, while the grain which has not been mulled contains grain with irregular corners and projections, as well as flat or plate-like pieces. In screening, 20 the particles of irregular shape often pass through the meshes of the screen even when their dimensions in certain directions considerably exceed the mesh size. For this reason a "mulled" grain usually gives the appearance of a somewhat smaller mesh grain 25 than one which has not been "mulled", even when both are carefully graded to the same mesh.

30 The conditions necessary for "mulling" as here defined will depend upon the toughness of the individual particles or grains. Conditions which would "mull" a material such as silicon carbide or aluminum oxide might result in an appreciable reduction in grain 35 size with certain materials having a lesser degree of toughness or resistance to impact. These conditions can be controlled to a great extent by variations in the speed of the mill, rate of feed, and size or weight of the pebbles or other means used for crushing. 40

I have found that by "mulling" the grain previous to the bonding together of the particles to form a refractory, the permeability to gases of the resulting product is greatly 45 reduced. The particles, owing to their regular shape, can be consolidated to a more dense material than is the case when grain that has not been mulled is used, even though the grit sizes are the same in both cases. The 50 process is especially applicable to the manufacture of refractory articles made from silicon carbide, although it may also be used in the case of crystalline aluminum oxide and other similar grain.

55 Care should be taken that the bond used is itself not permeable. With silicon carbide refractories this condition is approached with a close firing clay of the ball type, or with numerous other combinations of ceramic 60 bonds and fluxes which do not react with one another or with silicon carbide in such a way as to form a vesicular structure. The desired structure is most closely approached, however, by using a bond of about 10% fine silicon carbide. 65

The following examples from test records are given to show the effect of "mulling" the grain on the permeability of the refractory. The figures in each case represent an average 70 of a number of brick made in the same way, all of which closely approach the average value.

| Sample No. | Grain            | Bond | Permeability |               |
|------------|------------------|------|--------------|---------------|
|            |                  |      | Glazed       | Without glaze |
| (1) -----  | Not mulled ----- | A    | 35           | 350           |
| (2) -----  | Not mulled ----- | A    | 30           | 335           |
| (3) -----  | Mulled -----     | A    | 18           | 38            |
| (4) -----  | Mulled -----     | A    | 18           | 35            |
| (5) -----  | Not mulled ----- | B    | 305          | 725           |
| (6) -----  | Not mulled ----- | B    | 365          | 860           |
| (7) -----  | Mulled -----     | B    | 140          | 185           |
| (8) -----  | Mulled -----     | B    | 95           | 175           |

75 These data show clearly the value of mulled grain in reducing permeability, and that this effect is independent of the surface glaze. The additional value of using a refractory made from mulled grain in combination with a glaze is also shown. In the examples above 80 noted, it will be seen that the permeability of bricks made from mulled grain, even when not glazed, is in each case less than 250 units, as compared with a much higher permeability in unglazed brick where the grain has not been 85 mulled.

The unit of permeability above referred to is defined by the following method. For evaluating the permeability of a brick, I use a device whereby the brick to be tested is supported 100 broad face down on an annular felt gasket supporting its outer edge in an iron container, and the space between the sides of the brick and walls of the container is filled with mercury, while the brick is held in position by a 105 weight supported on it in such a way as not to prevent the passage of gas through the face of the brick.

With the brick in this apparatus, air is passed through it from the gasket outward at 110 such a rate that the drop in pressure across the brick amounts to 2" of water. When this condition has been established, the rate of flow is determined by a suitable capillary tube flow-meter inserted in the air line ahead of 115 the brick. The value secured is converted to a value for standard thickness by multiplying by

$$\frac{t}{2.5}$$

120 where  $t$  is the thickness of the brick in inches. The unit of permeability is defined as the number of cubic centimeters per minute flowing through the 9" x 4½" face of a standard 125 size brick under a difference of pressure of 2 inches of water. This method of measurement gives a readily duplicable value with a precision of ± 5% or better. By use of this apparatus the permeabilities of bricks, even 130

those containing different amounts and different types of bond, may be readily compared.

It will be understood that the permeability unit used throughout this specification and the annexed claims is a unit determined as herein described.

Although the invention has a special advantage when used with an oxidizable grain, such as silicon carbide, it is also advantageous in the case of refractories made from crystalline aluminum oxide, sillimanite or other similar materials. Because of the needle-like crystals present in sillimanite, the "mulling" process for producing a dense refractory is especially applicable.

I claim:

1. A silicon carbide refractory made principally of mulled silicon carbide grain.

2. The process of manufacturing a silicon carbide refractory which comprises mulling a substantial proportion of the silicon carbide grain, mixing the mulled grain with a binder, forming and burning the mix.

3. A furnace lining formed of silicon carbide made of mulled silicon carbide grain and a binder.

4. A burned refractory silicon carbide article containing in its composition, silicon carbide grains which have been ground down and rounded to the extent that the mix packs together to form a body, which, when burned has a permeability of less than 250 units.

5. The process of making a refractory article of low permeability which comprises grinding a refractory material to the desired grit size, dry mulling the refractory material after it has been ground to wear off the sharp corners produced in the grinding operation by attrition without further material reduction in the grain size, thereafter packing and shaping the mix containing the mulled grains, and burning the chafed article.

6. As a new article of manufacture, a silicon carbide refractory having a low permeability comprised principally of silicon carbide grains and grains which have been ground from a mass of silicon carbide to the desired grit size, and have thereafter been mulled, and having a higher density than an article of similar dimension and composition formed of grains which have not been mulled.

In testimony whereof I affix my signature.

FRANK JEROME TONE.