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2,956,868

METHOD OF MAKING CARBONIZED
BRIQUETTES

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This invention is a new and useful method of making carbonized briquettes consisting of particles of an inorganic oxide disseminated in a supporting structure of coke carbon. In certain reduction processes, the charge is made up of masses of coke carbon containing, in disseminated form, the particles of oxide to be reduced. One is the process of making elemental phosphorus described in my copending application, Serial No. 530,507, filed August 25, 1955, now United States Patent 2,897,057. Another is the continuous process for the production of certain carbides, for example the carbides of aluminum, calcium, titanium, zirconium, boron and silicon, by reduction of certain oxides with carbon described in my copending application, Serial No. 575,567, filed April 2, 1956, now United States Patent 2,869,990. Another in which such a charge is desirable is the St. Clair de Ville process for the production of certain metal chlorides by contacting the respective oxides with carbon and chlorine at a red heat.

Such charges may be made by mixing the oxide in subdivided form with coal or coke and a cokeable substance, such as tar or pitch, and heating the mixture in a coke oven or broad oven to from 700–1000° C., followed by ejecting, quenching and subdividing the coke so produced. This involves a waste of the specific heat of the hot coke and the production of fines which must be rerun. It is therefore desirable to produce the coke as briquettes of definite form, preferably by a continuous process, such that they can be supplied to the reduction stage at an elevated temperature.

The tars and pitches are not well adapted to this type of carbonization since they soften before reaching the temperature of pyrolysis and unless the green briquettes are heated in shallow layers, they deform and stick together. In most cases the oxides are relatively fine, i.e., predominantly passing 100 mesh, either because the material is friable or to facilitate reduction, and in such case bituminous coal in amount substantially sufficient to effect the desired reduction will not alone produce a satisfactory bond. Some success has been achieved by the use of a mixture of oxide, bituminous coal and a carbonizable substance, such as lignin or sulphite waste liquor, which effects a temporary bond until a temperature is reached at which the coal particles become plastic and unite to form a monolith or lattice which determines the structure of the eventual coke. Even with this expedient, however, the carbonizing must be carried out in shallow layers with very careful temperature control, otherwise the briquettes will deform and stick together.

I have now discovered, however, that it is possible to produce a highly satisfactory bond with the use of bituminous coal alone by grinding the materials, i.e. the oxide and bituminous coal, to certain relatively fine particle sizes and by the application of pressures of at least a certain magnitude. I have discovered if the oxide and bituminous coal are ground to predominantly pass a

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300 mesh screen (Standard Screen Scale, A.C.S. Year Book 1921–1922) it is possible by the application of pressures which are relatively high but still within the range of practical application, to cause the particles of bituminous coal to flow with respect to the oxide particles and to fill the voids and establish marginal contact, thereby forming a continuous structure of coal in which the oxide particles are disseminated. With particles predominantly passing 300 mesh, the pressure required to effect this result may be of the order of 25 tons per square inch. However, by grinding the particles so that at least 75% passes 300 mesh, the same result is obtainable by pressures of the order of 15 tons per square inch, and in some cases as low as about 8 tons. The material is preferably pre-compressed in a continuous briquetting machine to smaller sizes than ultimately required to minimize the compression ratio in the final briquetting. Preferably the materials are ground together, which insures uniform particle size of both the oxide and the bituminous coal. The grinding may be carried out in any suitable manner, for example in a jet grinder, a ring roller mill or a ball mill.

At this stage the mixture, theretofore gray, becomes brown to black and the particles of coal and oxide are no longer separately distinguishable by macroscopic examination or even low power (about 10×) magnification. Briquettes prepared in this way require no other binder. They are much denser and stronger than briquettes prepared with the binders heretofore mentioned. They have a strength in compression of about 3000 lbs. per square inch.

Inasmuch as the binder is the bituminous coal itself the briquettes are much less plastic at elevated temperatures than briquettes bonded with tar or pitch and their plastic range is narrower. At this stage they behave very much like masses of bituminous coal of the same kind and form. If the coal is highly plastic, deformation may occur on heating. If the coal is a swelling coal, some swelling of the briquettes will occur on carbonization. I find, however, that by substituting a non-coking, carbonaceous material, such as anthracite or coke, for part of the bituminous coal, I can produce a briquette that will retain its form exactly during carbonization and its strength in compression to a greater extent during carbonization than briquettes prepared by any method heretofore available. The amount of coke or anthracite required is easily determinable by a few simple preliminary tests in which the proportion of coke or anthracite is progressively increased until the briquettes show no swelling or deformation or sticking on carbonization.

After carbonization the briquettes have a greater density and a greater strength, both in tension and compression, than those prepared by any method heretofore available.

Whether at any particle size within the range of fineness specified the applied pressure is sufficient to produce flow of the type described, is easily determined by inspection. Satisfactory briquettes have a smooth exterior surface resembling pitch. They are strong and not easily shattered, even by dropping from a height of several feet onto a concrete floor. If fractured the fracture appears black. Marginal cases are occasionally encountered in which the surface shows an approach to the smooth, pitch-like surface. Such cases are easily distinguished, however, by the fact that they are easily shattered; although consolidated their strength in compression is much lower, ordinarily not over 1000 lbs. per square inch. After shattering the fracture is gray and the individual particles in the fracture are easily

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distinguishable and the pieces are easily crumbled by hand.

The following are practical examples of the application of my invention:

Example 1

Three parts of Florida phosphate rock were mixed with one part of silica flour and one part of bituminous coal. The phosphate rock contained the equivalent of 70% tricalcium phosphate and was about 49% through 200 mesh. The silica flour was through 100 mesh. The bituminous coal was ground in a hammer mill to pass 100 mesh. It was from the Empire Seam of the Alabama Coal Field and analyzed as follows:

	Percent
Volatile	37.40
Fixed carbon	60.30
Ash	2.30
Sulfur	0.9

The mix was ground in a ball mill to about 81% through 300 mesh. It was then subjected to various pressures to ascertain the minimum pressure at which the bituminous coal present could be caused to flow in relation to the oxide particles, to form a continuous monolith of bituminous coal in which the oxide particles are disseminated. This was found to occur at an applied pressure of about 8 tons per square inch. The remainder of the mix was then briquetted with similar results under an applied pressure of about 15 tons per square inch. The green briquettes so prepared had a strength in compression of 3100 lbs. per square inch. During carbonization they developed no sticking or deformation but retained their original dimensions and sharpness of outline. After carbonization they had a strength in compression of about 6680 lbs. per square inch.

A similar test was run on the same mix ground to approximately the same particle sizes in a so-called jet grinder, in which the particles are projected by means of a gas at high velocity. The results were similar.

Example 2

As a further practical example of the application of my invention, one part of calcined bauxite was mixed with one part of bituminous coal. The bauxite referred to was metal grade, from a deposit in Arkansas, running about 70% Al_2O_3 , balance principally silica with minor amounts of titania and ferric oxide. The coal was that referred to in the preceding example. The charge so prepared was ground in a ball mill to about 81% through 300 mesh. It was then subjected to various pressures to ascertain the minimum pressure at which the bituminous coal present could be caused to flow in relation to the oxide particles, forming a continuous monolith of bituminous coal in which the oxide particles were disseminated. This was found to occur at an applied pressure of about 7 tons per square inch. The remainder of the mix was then briquetted with similar results under an applied pressure of about 15 tons per square inch. During carbonization the briquettes so prepared developed no sticking or deformation but retained their original dimensions and sharpness of outline. After carbonization they had a strength in compression of 1860 lbs. per square inch.

Example 3

As a further practical example of the application of my invention, five parts of quicklime were mixed with 635 parts of bituminous coal. The coal was from Elkhorn #3 Seam, Weeksbury Mine, Floyd County, Kentucky, and analyzed as follows:

	Percent
Volatile	36.9
Fixed carbon	57.8
Ash	5.3
Sulfur	0.5

The mix was ground in a ball mill to about 82%

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through 300 mesh. It was then briquetted under an applied pressure of 15 tons per square inch. During carbonization the briquettes developed no sticking or deformation but retained their original dimensions and sharpness of outline. After carbonization they had a strength in compression of 2810 lbs. per square inch.

Example 4

As a further practical example of the application of my process I mixed 8 parts of zircon with 3 parts of metallurgical coke in the form of breeze and 3 parts of bituminous coal. The coal was from Pittsburgh Seam, Federal #1 Mine, northern West Virginia near Fairmont, analyzing as follows:

	Percent
Volatile	40.3
Fixed carbon	51.8
Ash	7.9
Sulfur	3.2

The zircon analyzed as follows:

	Percent
ZrO_2	66.06
SiO_2	32.62
TiO_2	0.30
Al_2O_3	0.79
CaO	0.02
MgO	0.02

The coal referred to is a swelling coal which if used without the addition of the coke, would produce a briquet subject to deformation in carbonization.

The mix was ground in a ball mill to 100% through 300 mesh. It was then subjected to various pressures to ascertain the minimum pressure at which the bituminous coal present could be caused to flow in relation to the oxide particles, forming a continuous monolith of bituminous coal in which the oxide particles were disseminated. This was found to occur at an applied pressure of about 6 tons per square inch.

The remainder of the mix was then briquetted with the production of a somewhat more dense briquet, but with otherwise similar results, under an applied pressure of about 15 tons per square inch. The briquettes so prepared had a strength in compression of 1888 lbs. per square inch. During carbonization they developed no sticking or deformation but retained their original dimensions and sharpness of outline. After carbonization they had a strength in compression of 3900 lbs. per square inch.

Although all bituminous coals tested have proven suitable, I do not assume that there may be bituminous coals, possibly of lower volatile content, which may not function similarly. In any case, no difficulty is involved in determining if a particular bituminous coal is satisfactory by application of the tests hereinabove described.

I claim:

1. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of such oxide and a substance selected from the group consisting of bituminous coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated, said oxide and said coal being of a particle size predominantly passing 300 mesh, subjecting said mixture to pressure at which the particles of bituminous coal in said mixture flow with respect to said oxide particles, forming a continuous monolith of bituminous coal in which said oxide particles are disseminated.

2. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke

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carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of a substance selected from the group consisting of bituminous coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated and said oxide, grinding said mixture to a particle size predominantly passing 300 mesh, subjecting said mixture to pressure at which the particles of bituminous coal in said mixture flow with respect to said oxide particles, forming a continuous monolith of bituminous coal in which said oxide particles are disseminated.

3. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of such oxide and a substance selected from the group consisting of bituminous coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated, said oxide and said coal being of a particle size at which at least 75% will pass through 300 mesh, subjecting said mixture to pressure at which the particles of bituminous coal in said mixture flow with respect to said oxide particles, forming a continuous monolith of bituminous coal in which said oxide particles are disseminated.

4. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of a substance selected from the group consisting of bituminous coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated and said oxide, grinding said mixture to a particle size which at least 75% will pass through 300 mesh, subjecting said mixture to pressure at which the particles of bituminous coal in said mixture flow with respect to said oxide particles, forming a continuous monolith of bituminous coal in which said oxide particles are disseminated.

5. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of such an oxide with a substance selected from the group consisting of coke and anthracite and a substance selected from the group consisting of bituminous

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coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated, each of said components being of a particle size predominantly passing 300 mesh, subjecting said mixture to pressure at which the particles of bituminous coal in said mixture flow with respect to the other particles present, forming a continuous monolith of bituminous coal in which said other particles are disseminated.

6. The method of preparing carbonizable briquettes to be used in reduction processes adapted to yield on carbonization a substantially continuous structure of coke carbon with particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof disseminated therein, which comprises forming a mixture of such an oxide with a substance selected from the group consisting of coke and anthracite and a substance selected from the group consisting of bituminous coals and mixtures thereof in amount sufficient to form a continuous phase of bituminous coal in which said particles are disseminated, grinding said mixture to a particle size predominantly passing 300 mesh, subjecting said mixture to pressures at which said particles of bituminous coal flow with respect to the other particles present, forming a continuous monolith of bituminous coal in which said other particles are disseminated.

7. As a new article of manufacture, a green briquet to be used in a reduction process consisting of a continuous monolith of about 20-56% by weight of bituminous coal with about 80-46% by weight of particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof uniformly disseminated therein.

8. As a new article of manufacture, a green briquette consisting of a continuous monolith of about 20-56% by weight of bituminous coal with a mixture of about 80-46% by weight of particles of a substance selected from coke and anthracite and particles of an oxide selected from the group consisting of metal and metalloid oxides and mixtures thereof uniformly disseminated therein.

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