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PROCESS FOR MANUFACTURING DENSE SHAPED CARBON ARTICLES

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1 Claim. (Cl. 18—55)

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This invention relates to an improvement in the manufacture of shaped carbon bodies comprising calcined coke and a binder, and more particularly to producing carbon electrodes.

The methods of manufacturing carbon electrodes which are commonly employed commercially comprise mixing crushed calcined coke with a binder such as coal tar pitch or other bituminous tar or pitch, followed by molding or extruding the mixture into the desired shape using high pressures generally in excess of 500 pounds per square inch. The green electrode thus made is baked and may be graphitized. In conventional processes the coke is crushed to a desired particle size, usually smaller than ¾ inch depending on the size of the electrode.

In the initial formation, coke flour comprising finely divided calcined coke particles is mixed with relatively larger coke particles or granules in varying proportions to fill the spaces between the larger pieces; the coke aggregate is then thoroughly mixed with the pitch binder. Oil may be added to assist in the lubrication during extrusion.

The calcined coke particles and coke flour employed is generally produced by heating petroleum coke or other low ash content cokes, at a temperature in a range of about 1000 to 1600° C. to reduce the volatile content below about 2%. The coke particles are made by crushing after calcination and are used as produced in the crushing operation. This is carried out in the conventional type of grinding equipment such as jaw crushers, roller crushers, hammer mills and the like, leaving angular projections on the pieces. The coke flour is made by pulverizing the fines obtained during the preparation of particles, so that 40–60% approximately, passes a 200 mesh screen.

The present invention relates to an improvement in the manufacture of electrodes which comprises subjecting the rough crushed coke particles to attrition milling to more or less spherulize the particles but without substantially reducing their particle size. The spherulized particles are then used in the conventional manufacturing steps employing coke flour and pitch binder.

The calcining operation is carried out on lumps of the raw coke followed by crushing. The calcined coke is hard and brittle and upon being crushed in the jaw type crusher or similar apparatus breaks into particles having angular points and edges. The present operation causes these angular projections to be attrited and the operation is carried out in such a manner that little or no further crushing or reduction in desired particle size occurs.

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According to this invention the crushed coke particles are introduced into an attrition mill which may comprise a drum containing baffles or lifters and which can be rotated. The barrel of the mill may be set on an eccentric so that the particles during the rotation of the barrel are caused to be lifted and rolled against one another as the operation proceeds. In this manner the sharp edges and points on the coke particles are broken or ground down. The milling should not be so severe as to cause substantial breakage of the particles themselves, i. e., breakage through the body of the particle. If this is done, the mesh size of the particles are reduced to an undesired extent. Since these points or projections represent but a comparatively small proportion of the volume of each particle, grinding off the points reduces the weight of the particle by only a relatively small amount and the average particle size as determined by screen analysis is substantially unchanged. The packing characteristics of the milled particles as evidenced by increased bulk density, are substantially improved. Thus for example a calcined petroleum coke from a Texas crude oil crushed to 8 to 10 mesh had a gravity packed bulk density of 0.80. After tumbling the granules in a cylindrical container equipped with baffles, for a period of 360 minutes, using a cylinder speed of 100 revolutions per minute, the bulk density had increased to 0.91, or 114% of the original. Attrition of 10 to 14 mesh crushed coke particles from the same source gave a bulk density 113% of the original. 14 to 20 mesh particles of the same coke were improved 117%. A screen analysis showed no substantial change in the particle size of any of these. The minor amount of fines produced was not over 3% of the original crushed particles and could be included to replace a corresponding amount of coke flour in the final electrode mix.

In a similar manner tumbled 8 to 10 mesh particles of calcined coke from an Ohio crude oil had a bulk density 118% of that of the original crushed particles, with no substantial reduction in mesh size. Tumbled 10 to 14 mesh particles from the same coke increased in bulk density to 117% of that of the original crushed particles. The bulk density of 14 to 20 mesh particles of the same coke were improved 121%.

Carbon electrodes were prepared from the coke from these two sources, using 60 parts by weight of the 8–20 mesh coke, 40 parts of coke flour and 26 parts of coal tar pitch binder. One part of

heavy lubricating oil was added to assist in extrusion. Green electrodes extruded from these mixtures were compared as to apparent density before baking. An improvement in apparent density of the green electrodes was consistently noted when using the particles subjected to the tumbling operation of the order of 102% of that of otherwise identical samples which had not been tumbled.

These green electrodes were then subjected to baking by gradually raising the temperature to about 1000° C. over a period of 8 days. An improvement in the apparent density of the baked electrodes from Texas tumbled coke was 102% of that of the electrode containing original crushed coke. The apparent density of tumbled Ohio coke electrodes was 103% of the apparent density of the electrodes containing the nontumbled coke particles. At the same time an improvement in electrical conductivity of the baked electrodes was noted, this conductivity being 108% of that of the electrodes containing crushed unspherulized coke.

These baked electrodes containing tumbled coke were then graphitized by heating over a period of 3 days until a maximum temperature of 2800° C. was attained and showed an improvement in apparent density of 106% and an increase in electrical conductivity of 103% over electrodes containing the untumbled coke particles. An improvement of this order is of substantial commercial importance since it represents a proportional reduction in power consumption in the process in which such electrodes are used. Such savings result in lower processing costs to the user of the electrodes, which in turn can be passed on to the consumer of the steel or other alloys made by electrothermal processes, thereby increasing their field of utility.

In the production of carbon electrodes, improvements of the order shown are substantial in view of the fact that (1) the improvement is consistently obtained and the electrodes are consistently better than can be obtained by using the crushed coke particles themselves and (2) because the spherulizing operation is inexpensive and does not waste the coke. The dust and fine particles formed due to attrition can be incorporated in the mixture to be molded or extruded, as a part of the flour.

Even relatively small improvements in conductivity and apparent density of baked or graphitized carbon electrodes, or anodes, are of considerable commercial importance due to the fact that this represents a substantial decrease in power consumption because of their greater conductivity. It also means that the shut-downs due to breakage and wearing away of the electrodes in electrothermal processes is reduced because of the increased strength of the electrodes or anodes made from coke particle components subjected to attrition milling.

It should be pointed out that the spherulizing operation as it is herein referred to should not extend to a point where substantial reduction in particle size of the coke occurs but only to the extent that the projecting or sharp edges are removed and the particles become rounded.

Any calcined coke suitable for electrode manufacture can be used in the process. Any suitable binder which is known may also be employed. The processing conditions of electrode manufacture are otherwise applicable.

We claim as our invention:

A process for manufacturing hard, dense, shaped carbon articles which comprises crushing calcined coke, separating fines, subjecting the coarser particles to attrition milling to remove the sharp edges and points therefrom without substantially reducing the particle size, mixing the milled particles with a pitch binder, forming the article in the desired shape and baking it, thereby to produce a hard, dense carbon article of higher apparent density and lower resistivity than is produced from the crushed coke without the milling treatment.

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