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METHOD OF FORMING CARBON ELECTRODES

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This invention relates to a method of making carbon electrodes for electric furnaces and for other purposes and includes the treatment of carbonaceous materials such as coal, coke, lampblack, or the like and suitable bonding agents which yield a carbonaceous bond on heating to provide bodies suitable for conducting the electric current, as for example, electrodes for electric furnaces, arc lamps, brushes and the like. This application is a continuation-in-part of my application Ser. No. 627,093, filed July 30, 1932.

Carbon articles of various types have been manufactured, principally from a suitable type of carbon such as petroleum coke, lampblack, anthracite coal, graphite and the like, which were proportioned with a binder, after having first been crushed or ground to the desired degree of fineness. Among the binders used were coal tar, soft, medium and hard coal tar pitches, and various hydrocarbons and organic binders. The mixing was performed in simple agitating machines usually in the presence of air and under atmospheric pressure. The articles were then formed and baked under suitable conditions. Such articles were produced with the concomitant production of much scrap both as formed in the so-called "green" state and later in the baked condition after considerable additional expense had been incurred in their fabrication.

As an example of the difficulties involved in the art of manufacturing carbon electrodes, more particularly those in the large sizes up to 40 inches in diameter, it has required from 30 minutes to 1 hour at the start of a day's operations before the product from the forming operation, especially in extrusion methods, is in suitable condition to be passed on to the baking department. This is exceedingly wasteful and expensive in that the product produced during this initial or starting period must be scrapped and be re-treated. The product up to this point has been unsuitable principally from inability to quickly control and correlate the temperature conditions in the mass, in the press and in the die. Much of the product made after the attainment of what might be called normal control conditions has been scrapped also due to inability to constantly maintain said normal conditions. This results in considerable loss at that point from inspection designed to send to the baking furnaces a product calculated to pass through that rather slow and expensive operation with as little further rejection after baking as possible. It has not been exceptional to scrap as much as 25% of a day's output and during times of labor shortage or other disturbances to scrap as much as 50% of the output. Frequent causes for rejection were expansion of some of the formed product beyond limits of tolerance permitted by the trade, surface blistering, tearing of the surface or edges of the product, distortion of ends due to cutting off to proper lengths, apparent specific gravity below minimum tolerance, internal checks and cracks, slippage and coring, etc. It was not unusual for the formed product to contain one or more of these defects and go undetected in the "green" or unbaked state, but such defects would finally appear in the baked product which would be rejected and which would result in substantially reduced output and increased costs.

In the prior art various expedients have been tried for improving the bond and electrical and physical properties of the finished product and for reducing the amount of scrap produced. For example, it has been attempted to increase the mixing temperature of the ingredients, to age the mixture and follow the same by a second mixing or agitation prior to forming. It has also been proposed to use various chemicals in the mixture to act as means for retaining a greater percentage of the binder carbon during the baking operation.

These and other attempts to improve the qualities of the finished articles may have resulted in some instances in a slight improvement in some of the properties of the finished carbon articles, but in substantially all cases the improved results, if any, were obtained at too great an increase in cost of processing. In other words, the cost of making the improvement was greater than the improved value of the article thus produced. It has been highly desirable to find means for reducing scrap losses and improving the qualities and particularly the bond of the finished carbon articles. This has been of the utmost importance when the carbon articles were to be subjected later to the process of graphitization in the electric furnace.

The present invention is adapted to overcome the disadvantages of the prior art and has succeeded in substantially eliminating the production of green and baked carbon scrap and in improving the properties and bond of the finished products. I thereby succeed in substantially reducing manufacturing costs and in manufacturing a carbon article of greater merchandising value.

The invention consists in the discovery of a

simple expedient and an inexpensive means for improving the carbonaceous bond in carbon articles resulting in a great improvement in the strength of the article, increase in the apparent density thereof, and a marked increase in resistance to oxidation and spalling during use.

My invention resides in the discovery that when carbonaceous materials and a suitable binder are "worked" prior to the forming, molding or extruding operation, either in a partial vacuum or in as nearly perfect a vacuum as can be obtained with the ordinary commercial vacuum apparatus, a very marked improvement is obtained in the above named qualities of the finished extruded or molded carbon article over that produced by ordinary methods.

By the term "worked" I mean more particularly the application of what might be called a "rub and twist" movement for want of a better descriptive term, as applied to the carbon aggregates and binder at a temperature above the melting point of the binder. This action subjects the materials or mass being worked to a mechanical pressure with relative motion between the carbon particles and binder or between the mass and the apparatus in which the mass is being worked, whereby there results a shearing off of air or other gas adsorbed on the surface of the particles. "Working" may include compacting, compressing or combination of treatments which produce the rubbing and twisting or smearing action.

In order to effect a sufficient improvement in bond necessary to give the desired results, I find it highly desirable to "work" the mass under vacuum and when "working" of the ingredients is performed under a vacuum or at a pressure substantially below atmospheric, the gas which is adsorbed on the surface is removed permitting a very substantially increased amount of surface to be firmly and adherently coated and this later becomes bonded by the baking or carbonizing process thereby resulting in carbon articles, such as electrodes, being substantially free from blisters, cracks, checks, laminations and other imperfections which by the old method of manufacture would call for the rejection or scrapping of substantial quantities of the baked product.

By my process, there will be removed during the "working under vacuum" adsorbed gas films by "work" and this allows the vacuum or reduced pressure to effect the removal of such gas from the system. My process results in the replacement of said gas film by a film of binder and the working of said binder into the available or exposed pores, which characterize the carbonaceous materials to be treated.

The use of vacuum alone does not remove adsorbed air or gas. It is the combination of "work" with vacuum that is the essence of this invention in the making of improved and cheaper electrodes and like articles. It is to be observed that my process is directed to the treatment of my materials prior to forming but the working under vacuum may be continued throughout the forming operation, if desired.

The present process improves the behavior of the material during extruding where a high temperature may now be used without danger of distortion or surface blistering as the article leaves the extrusion press or die. This is very important as it permits a higher ratio of press to die diameter without danger of clogging the die. And a fixed diameter press may be used to form various smaller sizes of electrode, decreasing the number of presses required. The transverse strength of the extruded material is further increased. By this process there is secured the same results by extrusion at low pressures which were hitherto considered obtainable only by molding processes under extremely high pressures. This materially reduces the amount of wear and tear and upkeep expense on pumps, valves, and other hydraulic apparatus and accessories.

Another advantage of my process is that the binder is not only more effectively applied but is applied in a much shorter time and as a result thereof, I have found an unexpected advantage. I find that I can now supply the extrusion or forming presses at double the former rate and by so doing I am enabled when starting up each day to reach that condition of temperature stability or equilibrium between the mass, press and die, which is necessary to make a good product, in about fifteen minutes which formerly required from 30 minutes to one hour. The greater rate of throughput of the press adds greatly to the temperature control at the press and die in that the mass itself apparently becomes a greater temperature regulating factor than the press and die heating means with the result that I find the daily output can now be very substantially increased and the unit cost thereby reduced.

As a specific example of the operation of my process I may take 100 pounds of calcined petroleum coke flour and mix the same with 25 pounds of medium pitch with sufficient oil for lubrication. To this mixture I may, if I so desire, add up to one pound of creosote oil although this is not essential. The material is maintained at about 170° C. or higher. In the preferred method of working the carbonaceous material and binder I cause heavy mulling rolls to pass over the mass generally in a thickness of at least several inches. Heavy rolls mounted on a horizontal axis and disposed vertically are caused to travel in a circular path so that there is produced by the face of the roll in contact with the mass a rubbing and twisting motion which under the heavy weight of the rolls shears off the adsorbed air and smears the binder onto the surface from which the adsorbed air has been removed.

The container for the mass and rolls is covered with a housing to make it air-tight and a vacuum pump connection is made thereto.

During the working operation a vacuum of about 22" or more of mercury is applied for about 15 minutes. When the vacuum is about 26" of mercury a working time of 10 minutes is sufficient. Thereafter, the vacuum may be released and ordinary or superatmospheric pressure may be applied and the working operation continued without detriment to the product. The resulting worked mass is then formed into articles of suitable shape, which are placed in an oven and baked at about 1000° C. or higher.

I am enabled by this method to incorporate a greater amount of binder into the carbon particles to be bonded than would otherwise be the case, and to obtain a more effective bonding of the carbon particle surfaces, with the result that the formed green carbon article has an improved apparent density. The bond under my process is a greatly improved surface bond which results in part from the extension of the bond to within the interior pores of said particles. The resulting baked carbon articles show improved

strength, greater apparent density, higher electrical conductivity and increased resistance to oxidation when made in accordance with my method of severely working the mass under the simultaneous influence of vacuum or substantially reduced pressure.

The improved results obtained by the present invention may be found in various properties of the electrodes both in the green and in the baked states. The present treatment gives green articles in which the deformation thereof, on cutting the same to the required lengths after extruding the mass, is greatly decreased. The surface blistering of the product which was found in electrodes made by prior processes on release from the extrusion die is completely eliminated. Also the radial expansion of the product as it is released from the end of the die is eliminated so that all pieces are of the exact diameter or cross section desired which eliminates the necessity for machining the finished product to give perfect electrical connections in the holders.

The improvement in the products may also be observed in the increased electrical conductivity, the increased apparent density, the decreased porosity of the finished product and the elimination of interior cracks and checks. The product also shows increased strength in tension and in compression.

In the baking of the electrode it now becomes feasible to utilize an increased firing range and an increased speed of firing. There also results a decreased shrinkage and it decreases the amount of scrap in the final baked product. The invention eliminates spalling under conditions of large temperature changes during use as when the operation is closed down at the end of the day, when the electrode is cooled from the temperature of the arc to room temperature and when resuming operations at the temperature of the arc. The improvement resulting from my method is of great value in amorphous carbon electrodes which are later to be subjected to the strains and stresses of graphitizing temperatures of over 2000° C.

While I have described my invention setting forth a single embodiment thereof, it will be apparent to those skilled in the art that my invention is not limited thereto. For example, the quantities and the character of the materials used, the temperatures, time and degrees of vacuum, may be varied within wide limits. Instead of using a single carbonaceous material I may use mixtures of aggregates and flour. Instead of pitch I may use tar, and this is included in the term "pitch" as used in the claims. These and other changes may be made within the spirit of my invention, which is to be broadly construed and is to be limited only by the character of the claims appended hereto.

What I claim is:

1. A method of forming carbon electrodes which comprises providing a carbonaceous material and a binder, placing the same in a closed container, subjecting the same to an intimate working by a rub and twist movement to disrupt and separate the film of adsorbed air on said material and at the same time subjecting said material to vacuum to remove the thus separated air from said container, thereby permitting the binder to replace the film of air on said material and coat the same, then forming an electrode from the resulting mixture and baking the same.

2. A method of forming carbon electrodes which comprises providing a carbonaceous material and a binder, placing the same in a closed container, subjecting the same to a rubbing action under mechanical pressure sufficient to disrupt and separate the film of adsorbed air on said material and at the same time subjecting said material to vacuum to remove the thus separated air from said container, thereby permitting the binder to replace the film of air on said material and coat the same, then forming an electrode from the resulting mixture and baking the same.

3. A method of forming carbon electrodes which comprises providing a carbonaceous material and a binder, placing the same in a closed container, subjecting the same to a rubbing action under mechanical pressure sufficient to disrupt and separate the film of adsorbed air on said material and at the same time subjecting said material to vacuum to remove the thus separated air from said container, thereby permitting the binder to replace the film of air on said material and coat the same, continuing said working under at least atmospheric pressure, then forming an electrode from the resulting mixture and baking the same.

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