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METHOD OF FORMING PELLETS OF FINELY DIVIDED COKED CARBONACEOUS MATERIAL AND FINELY DIVIDED NON-FUSING MATERIAL

Carl E. Lesher, Ben Avon Heights, Pa., assignor to Lesher and Associates, Inc., Pittsburgh, Pa., a corporation of Pennsylvania

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1 Claim. (Cl. 75—4)

This invention relates to a method of forming pellets of finely divided coked carbonaceous material and finely divided non-fusing material. This invention is in the nature of an improvement over the invention of my Patent No. 2,794,728,

My said patent discloses a process of making a flowable solid ore-carbon mass according to which preheated ore is admixed with finely divided coking carbonaceous material and the admixture is agitated in a rotating retort and heated through the wall of the retort so that the carbonaceous material softens and becomes sticky while in intimate contact with the finely divided ore, the agitation causing particles of the carbonaceous material and particles of the ore to become adherently connected together as fine solids forming a flowable mass, and continuing the heating and agitation of the admixture until it attains a temperature of at least 850° F. but not substantially higher than 1000° F. and the carbonaceous material returns to solid form. The process is preferably continuous, the material being treated normally being introduced continuously at one end of the retort and advancing through the retort toward the other end thereof as the retort rotates either because of baffles in the retort or by reason of the retort being inclined to the horizontal, and the product being continuously delivered at the second mentioned end of the retort.

For certain uses it is desirable to have the agglomerate produced by the process disclosed in my said patent of such average size in subsequent treatment gases may freely flow through the voids. Such sized material may thus be desired for use in a fixed or a moving bed reactor in which it may be desired to utilize pellets of controlled average size much greater than the size of the fine solids ordinarily constituting the flowable mass of my said patent.

I have discovered that I can produce pellets of large size as compared with that of the discrete solids normally made by the process disclosed in my said patent by following the process of the patent but additionally controlling in a novel way the temperature to which the mixture of finely divided coking carbonaceous material and finely divided non-fusing material is subjected in the rotating retort. I find that to accomplish such a result the admixed materials in the retort should be heated through the wall of the retort to a temperature approaching but not substantially exceeding the temperature of maximum fluidity of the carbonaceous material. By thus limiting the maximum temperature to which the admixed finely divided coking carbonaceous material and finely divided non-fusing material are subjected in the retort I produce pellets of relatively great size and also of relatively great uniformity of size.

Also, I find it advantageous as contributing to the production of pellets of large uniform size to limit the loading or the extent to which the retort is filled with the admixture of finely divided coking carbonaceous material and finely divided non-fusing material. I find that

the retort should be filled to the extent of not over about 7% of the volume of the retort with the admixed materials and optimum results are obtained if the retort is filled with such materials to the extent of not over about 5% of the volume of the retort. I am, of course, referring to the materials in the condition in which they exist throughout their travel through the retort. Initially the finely divided coking carbonaceous material and the finely divided non-fusing material are simply admixed. As they progress through the retort they agglomerate into pellets and the carbonaceous material cokes. The volume of material other than gases within the retort at one time should not exceed about 7% of the volume of the retort and preferably is kept down to not over about 5% of the volume of the retort.

The limiting of the loading of the retort as just described results in a relatively thin layer of material in the retort which is heated through the wall of the retort, the relative thinness of the layer resulting in relatively rapid heating of the material which in turn results in the carbonaceous material in the retort being substantially more fluid at the temperature of maximum fluidity than when the material in the retort is heated more slowly as is the case when the retort is more heavily loaded. I find that such rapid heating of the material coupled with the limitation of the temperature to which the admixture is subjected in the retort as above described produces pellets of unprecedentedly large size.

Various coking carbonaceous materials may be employed as the binder for the making of pellets according to my invention. One such material is high volatile coking coal, and for the purpose of explanation and illustration the invention will be described in connection with the formation of pellets using high volatile coking coal as a binder. Other examples of coking carbonaceous materials which may be employed are coal tar pitch and petroleum as asphaltic pitch. Various non-fusing materials may be used, such, for example, as ores of minerals, non-coking coal, coke fines or anthracite. By "non-fusing" I mean non-fusing at the temperature employed in my process.

Coking coal has the property of softening or fusing as it is heated; when softened it is plastic or fluid after which with continued heating it hardens and becomes coke. The temperatures at which different coking coals soften, become fluid and harden to coke vary as does the degree of fluidity but in general the high volatile coking coals which I prefer to use have a spread of about 200° F. between the beginning and the end of the plastic range. This characteristic of coking coal has been extensively investigated and several laboratory devices are in use for measuring the relative degree of fluidity and the temperatures at which different coals soften, become fluid and harden, by which testing those skilled in the art may determine for each coal the range of its fluid property. The fluidity of coking coal may be increased substantially by rapid heating. Increasing the rate of heating increases the maximum fluidity and raises the final solidification temperature, but has little effect on the initial softening temperature. Also, prolonged heating in the preplastic range, that is, below 700° F., will reduce the fluidity of coal and at the same time raise its softening temperature.

Although it is customary in the literature to report 500° C. (932° F.) as the lowest temperature at which low temperature carbonization of coal is conducted, high volatile coking coal may be carbonized to coke at lower temperatures. A coal that begins to soften at 700° F., when heated to 800° F., but not above that temperature, will subsequently harden to form a high volatile low temperature coke. A weakly coherent coke has been obtained in the laboratory with coal heated to a maximum tempera-

ture as low as 700° F. Carbonization is a process of destructive distillation in which if conditions favor destruction the solid residue is increased; if distillation is favored the gas and particularly the condensable products are increased.

In a rotating, externally heated retort coking coal heated to 800° F. will become plastic and, if removed immediately from the retort while at that temperature, will be soft and can be molded, but it becomes rigid and hard upon cooling. It will also evolve tarry vapors for a short period; that is, until it begins to cool. If then reheated to substantially higher temperatures it will not again soften, but will of course be further devolatilized. In this instance destruction of coking property prevailed over distillation. The results to be observed when heating is stopped at or near the temperature of maximum fluidity of the coal, which may be about 850° F., are different only in degree. Less vapors are evolved and the product coke becomes rigid much more quickly. When the temperature is increased to 900° F. or more, the solidified product evolves very little vapor when removed from the retort.

In the application of my process the important variables that control and determine the size range of pelleted product are percentage of coking coal in the charge, screen size of the ore, and techniques of operation. There is no technical basis or standard for acceptable size of pellets other than, as stated, that of void space in a reactor bed. A size range of pellets not suitable for one reactor process may be acceptable for another. I prefer to have at least 40 percent by weight of the pellet product greater than ¼ inch, with a relatively small amount greater than 1 inch, and with less than 5 percent that will pass a 100-mesh screen. The weighted average size may thus be from a quarter to one-half inch.

Using any ore of uniform size consist, operating conditions being the same, the size of pellet product will increase lineally with the percentage of coal in the charge. A series of seventeen tests with a fine sized ore gave 25 percent pellet product plus ¼ inch with 25 percent coal, increasing regularly to 66.7 percent of plus ¼ inch product with 44.5 percent coal.

The operator who may desire to make ore-carbon pellets by my process and who is not limited by conditions imposed by subsequent processing of the pellets may use increasing percentages of coal—hence increasing carbon—to obtain larger average sized product.

The finer the size of the ore the smaller the average size of pellet product, all other conditions being the same. For instance, an average of six tests in which the ore contained 79 percent that passed the 100-mesh screen, and in which the average percentage of coal charged was 27, gave a product with 10.3 percent over ¼ inch. A series of five tests in which the minus 100 mesh ore was 49 percent and the coal used 30.8 percent gave a product with 49 percent over ¼ inch. Blending coarser ore, not substantially over ¼ inch, with fine sized ore is thus a means of control of product size. Varying the ratio of coal to ore and the selection of ore size are thus means of control of pellet size.

There is, however, a large area of use for ores agglomerated with reactive carbon as in my process in which the ores to be used are of fine size, and where it is desired to maintain the proportion of carbon in the agglomerate or pellet within predetermined limits. The smelting of iron ore to pig iron in the blast furnace requires coke—that is, carbon—for chemical reduction of the oxide ore as well as for production of heat by combustion. In the electric pig iron furnace carbon is required only for reduction. For the electric pig iron furnace it is desirable to have the maximum average sized pellets containing a limited percentage of carbon. For instance, it may be desirable to limit the carbon in the pellets to 20 percent while having an average size greater than would be attained under ordinary operation with coal in the charge limited to that which would give but 20 percent carbon.

The effects of the process variables of ore particle size and percentage of coal in the charge have been discussed. The third variable in my pelleting process that affects pellet size I have designated as techniques of operation, by which I mean process control.

My preferred process for making pellets is to rapidly heat the mixture of coal and preheated ore in a rotating, externally heated retort; to continue the rapid heating until the mixture of ore and coal reaches but does not exceed the temperature of maximum fluidity of the coal, which for most high volatile coking coals lies in the range of 820 to 870° F.; meanwhile causing the plastic mixture to form pellets in the retort rotating at a rate to give a peripheral speed of not less than 80 feet per minute and preferably more than 100 feet per minute; continuing the pellets in the retort to compact, round and harden them; then discharging the pellets continuously from the retort in solid form.

I heat the coal rapidly during the pelleting process to increase fluidity and thereby to promote the bonding of ore pellets. The rates of heating that I have utilized in my process have ranged from 60° to over 100° F. per minute. Preferably the coal at atmospheric temperature, 70° F., is elevated to approximately 850° F. and discharged in pellet form from the retort in about 10 minutes. The average rate of heating of fourteen tests in my pilot retort was 80.5° F. per minute, with an average residence in the retort and discharge mechanism of 10 minutes.

The manner in which I heat material in my process has been generally described in my said patent and in my copending application Serial No. 460,769, filed October 6, 1954. My said patent and application describe the heating of the coal with hot ore in the pre-mixer. The improvement in the process covered by the present invention is concerned with the preferred conditions for rapid heating at controlled temperature and for pelleting within the rotary externally heated retort.

I have found that maintaining the preheated charge material in the retort in a thin layer and passing it over the externally heated shell of the retort at a relatively fast rate along the length of the retort while rotating the retort at a peripheral speed of 100 feet per minute, more or less (a typical retort may be 5 to 10 feet in diameter and 15 to 50 feet in length), gives a rate of heating to the charge not previously attained. Desirably the peripheral speed of the retort should be of the order of at least 100 feet per minute. By external heat the steel shell is maintained at a temperature above that desired in the charge and as the charge is spread in a thin layer on the steel, with fresh particles continuously coming in contact with the hot metal, heat transfer is accelerated.

If the heating of the coal is continuous to and beyond the "setting" temperature, as, for instance, from the initial atmospheric 70° to 900° F. or more, the interval of time during which the coal is plastic or "sticky" is a function of the rate of heating. The more rapid the coal is passed through the stages of change to solid coke, the shorter the time it is capable of picking up the ore particles. It is only while in the plastic condition that the coal serves as a binder to make pellets. Even when rapidly heated to increase the degree of fluidity and extend the time between softening and solidification, the time interval during which the coal is sufficiently sticky to act as a binder is limited. For instance, a coal that begins to soften at about 700° F., if progressively heated may reach its maximum degree of fluidity at 840–850° F. and thereafter will rapidly harden and at about 900° F. become solid coke. Such a coal, when progressively heated at a rate to raise its temperature 75° F. to 80° F. per minute, is fluid for but two or three minutes.

In my process the formation of pellets begins as the coal softens and proceeds until the plastic binding ma-

terial hardens. It is desirable therefore that the coal be fluid and that it remain in that state for a sufficient time to form a plastic mass or "dough" that may be rolled into pellets before changing to the rigid state of coke. In other words, it is desired to have the coal become plastic and sticky, to have it pick up the ore, and then be rolled into pellets before decomposition of the plastic binder to solid coke.

I have found that by rapid heating to but not substantially above a temperature approximately that of the maximum fluidity of the coal the product will contain a much higher percentage of pellets of the desired size than if the temperature in the retort had been substantially higher.

The effectiveness of this technique of operation will be illustrated by an example. Two pilot plant tests were made using identical ores of identical size consist, with the same coal, the same rate of charging of both coal and ore and the same temperatures of preheat of the ore. All materials and operating conditions were alike except that in one case the carbonizing temperature in the retort was 855° F. and in the other the carbonizing temperature was 105° higher, or 960° F.

Heating rates from start of coal feed to mix screw to discharge of product were about 100° F. per minute; the average retention in the pelleting retort was almost the same. The difference in carbonizing temperature of 105° F. was the only variable. The lower temperature gave a product containing 51 percent of pellets above ¼ inch, the higher temperature gave pellet product having but 14.1 percent above ¼ inch. Following are the test data:

Test Number	1	2
Rate of charging:		
Ore, lb. per minute	2.250	2.210
Coal, lb. per minute	.815	.835
Temperature of ore preheat ° F.	825	828
Carbonizing temperature ° F.	855	960
Peripheral speed of retort, ft. per min.	94	94
Average time in process from initial charge of coal to discharge of product, minutes	8	7.5
Average retention in retort, pounds	21.7	20.2

Size Consist of Product	Weight percent	
Plus 1 inch	8.3	0.6
½ x 1 inch	19.2	3.3
¼ x ½ inch	23.5	10.2
Total plus ¼ inch	51.0	14.1
Passing ¼ inch	49.0	85.9
	100.0	100.0

While I have described a present preferred method of practicing the invention it is to be distinctly understood that the invention is not limited thereto but may be otherwise variously practiced within the scope of the following claim.

I claim:

A method of forming pellets of finely divided non-fusing material and coked carbonaceous material in which the coked carbonaceous material acts as a binder to bind together the finely divided non-fusing material comprising mixing finely divided non-fusing material with finely divided carbonaceous material which when heated initially softens, subsequently attains maximum fluidity and finally solidifies, rapidly heating said mixture to but not substantially above the softening temperature of said carbonaceous material, tumbling the admixed materials, while said carbonaceous material is at or not substantially above its softening temperature, on the inner surface of a rotating retort, heating through the wall of the rotating retort said admixed materials to a temperature approaching but not substantially exceeding the temperature of maximum fluidity of said carbonaceous material whereby the tumbling admixture forms into pellets and continuing the tumbling of the pellets in the retort to compact, round and harden them.

References Cited in the file of this patent

UNITED STATES PATENTS

2,477,412 Lohse ---------- July 26, 1949
2,812,249 Culberson et al. ---------- Nov. 5, 1957

UNITED STATES PATENT OFFICE

CERTIFICATE OF CORRECTION

Patent No. 2,918,364 December 22, 1959

Carl E. Lesher

It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 1, line 44, after "size" insert -- that --; column 2, line 40, for "temperature" read -- temperatures --.

Signed and sealed this 31st day of May 1960.

(SEAL)
Attest:

KARL H. AXLINE
Attesting Officer

ROBERT C. WATSON
Commissioner of Patents