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COMPOSITION OF MATTER PRODUCED BY AGGLOMERATING OR CEMENTING GRANULAR MATERIALS.

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To all whom it may concern:

Be it known that I, THOMAS M. CHANCE, a citizen of the United States, residing at Philadelphia, in the county of Philadelphia and State of Pennsylvania, have invented a certain new and useful Improvement in Composition of Matter Produced by Agglomerating or Cementing Granular Materials, whereof the following is a specification.

My invention relates to the agglomerating or cementing of granular materials such as coal, coke breeze, ores, concentrates and the like, by the use of a cementing material such as coal, or other solid hydrocarbons which have the property when heated of coking or of agglomerating into a coherent cementing substance. Anthracite coal or coke, consisting of particles less than one-half inch in diameter cannot readily be burned in furnaces adapted to the combustion of coarser fuels, especially when the fuel consists largely of particles one-eighth of an inch or less in diameter, because sufficient air for combustion can be forced through such a bed of fuel only by using an air pressure that tends to blow the finer particles out of the combustion chamber. To overcome this recourse has been had to two methods in which the fuel is agglomerated by the use of a cementing material. Bituminous coal has been thus used by mixing it with the fuel before firing, the heat of the furnace coking the bituminous coal, thus cementing the particles of coke or anthracite coal together before the temperature of the latter reaches that at which its combustion occurs. To effect this by methods heretofore used requires the addition of a relatively large percentage of bituminous coal. In other methods of utilization the coal is briquetted by the use of a binder prior to its introduction into the combustion furnace.

In describing my invention I will describe its application to the making of briquettes from anthracite "culm" or "dust" which consists of fine coal (smaller than about $\frac{1}{8}$ or $\frac{3}{8}$ inch) rejected as waste in the preparation of anthracite coal for market, it being understood that other materials such as those above mentioned may be cemented in like manner. By employing a combination of the improvements which I

have invented I have succeeded in making 55
briquettes of anthracite fines, typical of high-grade culm, and bituminous coal, of extraordinary hardness and strength, and capable of being subjected to rough handling without appreciable breakage. Tests 60
to determine the strength of this material under compression, using cylinders with a height about $1\frac{1}{2}$ times their diameter, show breaking strengths varying from 1200 up to 8400 pounds per square inch. The 65
strength varies with the character and composition of the materials used, their relative coarseness or fineness, the proportions in which they are mixed, the character of temporary binder used, the compression 70
under which they are molded, the temperature at which they are coked or baked, the character of the atmosphere in which they are coked and the time consumed in coking or baking. Briquettes made of the 75
same materials under the same conditions show remarkably uniform strength, for example, one set of briquettes made with 75 per cent of anthracite fines and 25 per cent of Westmoreland, Penna., bituminous coal 80
(Pittsburgh bed) with about 3 to 4 per cent of Texas oil distillate as temporary binder, showed breaking strengths under compression of 7330, 7750 and 7820 pounds per square inch. Another similar set coked at 85
slightly lower temperature showed strengths of 5920, 6080 and 6160 pounds per square inch, and another set duplicating these conditions showed strengths of 5960 and 6070 pounds per square inch, while similar 90
briquettes made in which 8 per cent of water was used to replace the oil as a temporary binder, showed strengths of 5500 to 6200 pounds per square inch. The material thus produced is very much stronger than anthracite coal or than the best grades 95
of coke, which latter show compression strengths of 1000 to 3000 lbs. per square inch. Its specific gravity varies with the variables above noted, usually ranging from 100
about 1.00 to 1.20. It is porous and therefore quick burning.

The several sets of briquettes above described were all made with about 74 to 75 per cent of anthracite fines and 24 to 25 per 105
cent of Pennsylvania bituminous coal, were all formed under practically the same pressure of about 7500 pounds per square

inch, three sets being made with oil as a temporary binder and another series of three sets of the same composition but with water as a temporary binder, the first set, that showing the greatest strength, having been coked for about 20 minutes in a furnace at a bright red heat, and the second and third sets at an ordinary red heat, and those of the water binder series were coked at temperatures about the same as those of the oil binder series, the strength of the resulting briquettes being thus shown to be affected in greater or less degree by the character of temporary binder used and the temperature at which coking was effected. In making these briquettes the anthracite fines were so graded that they consisted of particles that varied in size from 60 mesh down to 120 mesh, being passed through a 60 mesh sieve and having had removed material fine enough to pass through a 120 mesh sieve, and the bituminous coal consisted of finely ground material all of which passed through a 120 mesh sieve and containing also much finer material down to and including more or less impalpable dust. Other illustrations of composition of briquettes we have made are as follows: Briquettes of fair strength made by molding at a pressure of 800 pounds per square inch and coking for 45 minutes at a dull red, the briquettes being of an eggette shape $1\frac{1}{2}$ to $1\frac{3}{4}$ inches in diameter, and composed of 33.3 per cent of anthracite coal from $\frac{1}{4}$ inch to 20 mesh, 50 per cent of anthracite from 20 to 120 mesh and 16.7 per cent of bituminous coal smaller than 120 mesh, about 10 per cent of water being used as a temporary binder; Eggette briquettes $1\frac{1}{4}$ by $2\frac{1}{8}$ inches in diameter made with 80.75 per cent of a mixture of anthracite comprising sizes of 20 mesh to 120 mesh, and 60 mesh to 120 mesh, with 19.25 per cent of bituminous coal smaller than 120 mesh, with 8 to 10 per cent of water added before molding, molding at pressure of about 800 pounds per square inch and coking 50 minutes at a dull red, these briquettes being dropped twice from a height of 10 feet upon a hard yellow pine floor without injury, but breaking into several pieces on the third drop; Cylindrical briquettes, hard, bright and capable of taking a high polish, made of a mixture of 80 per cent anthracite sized between 60 mesh and 120 mesh sieves and 20 per cent of bituminous coal ground to pass through a 120 mesh sieve, mixed with 8 per cent of water and molded at low pressure in a tube $1\frac{3}{8}$ inch in diameter and coked at a dull red temperature in tube for 25 minutes; Massive agglomerated anthracite, made from a mixture of 75 per cent of anthracite sized between 60 and 120 mesh, and 25 per cent of bituminous coal smaller than 120 mesh, with 8 per cent of water, placed in a pasteboard box $1\frac{1}{2}$ x 1 x $3\frac{1}{2}$ or

4 inches in a cold oven and brought up to an orange heat in about 40 minutes, which was strong enough to bear ordinary handling without serious breakage.

Many attempts have been made to utilize anthracite culm by coking with bituminous coal, and experiments have been made also in briquetting a mixture of anthracite culm and bituminous coal and heating the briquettes to coke the bituminous coal. The procedure adopted in making anthracite coke has usually been to mix the culm with bituminous slack coal, or lump coal crushed to the size of slack (about $1\frac{1}{4}$ inch mesh) or perhaps smaller, and to charge the material so prepared into a beehive or by-product coke oven. The character of coke thus obtained depends upon the proportion of anthracite culm to bituminous coal, it being found that when much more than one-half of the charge consists of anthracite culm the coke produced is too weak and friable to bear handling or transportation or to be suitable for use as a metallurgical coke. Hence this method of utilizing anthracite culm has not come into general use. In the experimental production of briquettes the procedure has generally been similar, except that the bituminous coal and anthracite culm have in some cases been ground to small size before briquetting, in some cases both being ground to pass through a 20 mesh sieve, and the material has then been mixed with a binder such as tar, water, molasses, etc., and briquetting under pressure prior to being subjected to coking in a baking or coking oven. These experiments were unsuccessful when using water or tar as temporary binders, but by using 11 per cent of molasses with a mixture consisting of 75 per cent anthracite culm and 25 per cent bituminous coking coal, Parr and Olin, obtained moderately strong briquettes, described in University of Illinois Bulletin No. 60, June 3, 1912, as having a crushing strength of 650 pounds per square inch. Briquettes of moderate strength have been made by coking a mixture of coke breeze using pitch, and also by using coke breeze or crushed coke mixed with tar and without the admixture of bituminous coal, and briquettes have also been successfully made from coke made by low temperature coking (partially coked coal) by the admixture of tar and subsequent coking at high temperature. Coke has also been made successfully from anthracite culm by mixing it with tar, asphalt or similar coking material without the use of bituminous coal and coking the mixture, the residual carbon from the destructive distillation of the tar forming a coke-like cementing material which binds the particles of culm together, thus forming a mass of reasonable strength. None of these products approaches in strength or in value the ma-

terial which can be produced by the use of the improvements and inventions herein described.

I have discovered that to agglomerate or cement together granular materials of the above described types in such manner as to secure maximum strength of the finished product, both the material to be cemented and the material used as the cementing agent should be properly prepared. If the material to be cemented is inherently stronger than the cementing agent, then maximum strength will be attained when the particles of the stronger material are in more or less intimate contact and the cementing agent is less, or does not greatly exceed, in volume the volume of the pores or interstices between the particles of material to be cemented. If, however, the material to be cemented is inherently weaker than the cementing agent, then the strength of the finished product will be increased by increasing the percentage of the cementing agent. Failure to recognize these principles and failure to prepare the ingredients used in conformity therewith are the causes of failure to produce coked fuel and coked briquettes of satisfactory strength.

In the agglomerating by a cementing substance of materials of the above described types, the cementing substance may be considered to act either as a cement to hold the particles together or as a cellular matrix with the individual particles embedded in its cells. In the latter case the cementing material should have a volume equal to or greater than the sum of the interstices, or the porosity, of the granular material; but if the cementing material merely performs the functions of a cement, then its volume should not exceed and advantageously may be less than the porosity of the granular material. I find that when the granular material consists of particles of relatively strong material, the greatest strength can be obtained in the finished product by close juxtaposition of the particles of granular matter and I attain this end by introducing the cementing material in a very fine state of subdivision and in sufficient quantity to insure a film of such substance between all adjacent particles of the granular material which is to be cemented or agglomerated. In other words, the cementing material must be distributed throughout the granular aggregate to form a series of films adhering to the contiguous particles, and not as layers separating these particles by an appreciable space or interval. If the cementing material is a solid such as bituminous coal, or like substance, it must therefore be ground, crushed or pulverized so that its individual particles will be relatively smaller than the individual particles of which the granular material consists. Unless the cementing ma-

terial is thus finely comminuted, its coarser particles will separate the particles of the granular material by an appreciable space and thus increase the porosity of the granular material. As an illustration of this principle, I have made briquettes of anthracite coal ground or crushed to pass through a 20 mesh sieve and mixed it with 25 per cent by weight of bituminous coal similarly ground to pass through a 20 mesh sieve and have made other briquettes of the same materials except that the 25 per cent of bituminous coal was ground to pass through a 120 mesh sieve, the materials in both cases being thoroughly mixed, moistened with about 8 or 10 per cent of water and molded in a briquette press under the same pressure and coked or baked at the coking temperature of said bituminous coal, a red heat, in identical containers (to exclude oxygen) for the same length of time at the same temperature; the first set of briquettes were relatively weak and friable and when tested broke under compression at 400 to 600 pounds per square inch, while the second set made with the finely ground bituminous coal were hard and strong and broke at 1300 to 1400 pounds per square inch. I have made briquettes also from the same anthracite and bituminous coal, both of which were ground to pass through a 120 mesh sieve, using the same proportions as to weights, the same pressure and the same coking temperatures as in the other experiments described in this paragraph and found they were uniformly weak and friable and broke under compression at about 300 to 400 pounds per square inch, still further illustrating the requirement that to produce strong briquettes the cementing substance must be so comminuted that its particles will be materially smaller than those of the granular material which is to be cemented or agglomerated.

My invention is therefore based primarily upon the improvement which consists in the use of a coking material composed of particles materially smaller in size than those of the granular material which is to be cemented or agglomerated, said coking material being in quantity sufficient substantially to fill the voids between the particles of the granular material. By using this method I have successfully briquetted magnetic iron ore concentrates ranging from 40 to 100 mesh by the use of bituminous coking coal ground to pass through 120 mesh.

A further improvement which I may employ in the cementing or agglomeration of materials of the above enumerated types, consists in the preparation of the granular material to reduce its porosity and increase its strength, but I do not herein claim this improvement except in combination with the invention herein disclosed, as I have made it

the subject of another application for patent filed September 27, 1920, Serial No. 413,196. This latter improvement consists in grading the granular material as to the size of its particles, so that it shall be composed of classes of different size, and so proportioned that the weight or volume of each of such classes is sufficient approximately to fill the interstices of that of the next larger size, thus decreasing the porosity of the mass as a whole and bringing the individual particles into more intimate juxtaposition. In conformity with the principles already outlined this preparation will include the removal of particles as small as, or smaller than, those of the cementing substance to be used in cementing or agglomerating the granular material, but it will of course be understood that if these small particles of granular material be present in relatively small quantity their removal may not be necessary to obtain a satisfactory result.

The strength of many natural and some artificial products is increased by crushing to relatively small size, that is, the small particles may have greater relative strength to resist crushing than large particles. This is doubtless due to the presence of natural cleavage or fracture planes in the larger pieces; thus, while the strength of anthracite coal in cubes of one or two inches in size usually ranges from 2000 to 8000 pounds per square inch, I have found by crushing tests that cubes of $\frac{1}{4}$ inch or $\frac{1}{8}$ inch show crushing strengths of 10,000 to 18,000 pounds per square inch, in other words, the smaller pieces are relatively stronger than the larger pieces. Hence one of the improvements which I may employ contemplates crushing the coarser particles for the purpose of increasing the average strength of the aggregate. In preparing the granular material by grading as to the size of its particles, any crushing which may be necessary for that purpose will tend to increase the strength of the aggregate by increasing the relative strengths of the particles of which it consists. Followed to its logical conclusion, this premise would imply that the strongest aggregate would be obtained by crushing all of the granular material to very small size, but this is not practicable because the reduction of the particles to very small size greatly increases the sum of the superficial surfaces of these particles, and as these surfaces should be coated with a film of the cementing material, the quantity of the cementing material necessary to coat these surfaces becomes relatively too large to comply with the requirements essential to the production of a finished product of satisfactory strength, because the thickness of this film of cementing material between the individual particles tends to separate them by an interval that, compared

with the size of the particles, is relatively too great. If the size of the particles of granular material be very small, it becomes practically impossible to comminute the cementing substance to the still smaller size necessary, or to secure such complete and thorough admixture of the materials so prepared as to insure uniform distribution of the cementing substance throughout the mass. I am aware that in the manufacture of briquettes the materials have been crushed or ground to varying degrees of fineness for the purpose of securing a product of uniform texture, but I believe crushing to produce grading as above described and to increase the strength of the granular material is a new and useful improvement in the art. I do not, however, herein claim this improvement except in combination with the invention herein disclosed, as I have made this the subject of another application for patent as already stated.

When the particles of the material to be agglomerated are angular in form, it is evident that they cannot be firmly cemented together to produce a relatively strong product, if they are merely cemented together by a film of cementing material, because many of the points of contact will consist of a sharp edge or a sharp point of one particle in contact with a flat surface of another particle, and the contact will consist merely of a point or a line. If in such case the flat surface of the particle be coated with a mere film of a cement, angular particles in contact with such flat surface, where such contacts are merely lines or points, will not be cemented to the first particle in such a way as to develop material strength, and to insure a union between such particles that will be relatively strong, the cementing material must be built up around such point of contact, so that the sharp corners or edges of the particles are embedded in the cementing material, which thus forms a matrix which adheres to the sharp points or edges by a much greater area, than the area of actual contact of particle to particle. It will therefore be understood that to produce an agglomerated product of satisfactory strength, it becomes necessary to use more of the cementing material than that which would be required to merely coat the surface of such angular material with a thin film of the cementing substance, and that the strength of the product will be at a maximum when the cementing material is present in sufficient quantity to form a matrix that practically fills the voids or interstices between the individual particles which are to be cemented or agglomerated.

To secure close juxtaposition of the particles of granular material with each other, and with the cementing substance, the mixture may be compressed. Moderate pres-

sure exerted upon a dry mixture of the described type does not however make much change in the relative position of the particles comprising the mixture. If, however, the mixture be moistened with water, oil, or other liquid, the particles seem to slide more readily upon each other, the liquid doubtless acting as a lubricant to reduce interstitial friction, and the mass is materially reduced in volume with corresponding increase in density. A further function of such liquid is its property of holding the material together by "surface tension", "molecular attraction," "capillarity," or whatever term may be used to designate the force that causes wetted particles to adhere to each other.

The quantity of cementing material to be used to attain the best results depends upon the porosity or percentage of voids in the material to be agglomerated or cemented, and upon its relative coarseness or fineness. I have successfully made briquettes of anthracite coal in which the particles of anthracite coal varied from $\frac{1}{4}$ inch mesh down to 120 mesh, grading the percentages of coarse and fine particles as described in suitable proportions, and using as the cementing material mixed therewith from 7.5 per cent up to 30 per cent of finely ground coking coal. The strength of the material usually increases with the percentage of coking coal up to about 25 to 30 per cent. If too large a quantity of coking coal be used the briquettes are likely to swell or suffer deformation, with the formation of blow holes or shrinkage cracks.

The presence of relatively coarse anthracite, that is, particles $\frac{1}{4}$ to $\frac{1}{2}$ mesh, tends to reduce the strength of the resulting product, and the presence of much anthracite "dust," that is, particles smaller than 100 or 120 mesh, also tends to reduce the strength. Hence, in the introduction of coarse material to reduce the porosity of the mass, regard should be had to the relative strength of the particles so added; otherwise the final product may be weakened by these coarser particles.

The coking coal used as a cementing material should, as already stated, consist of particles materially smaller than those composing the mass of material to be cemented. I have obtained excellent results by using bituminous coal ground to pass through a 100 or 120 mesh sieve, containing of course a large quantity of material of 200 and 300 mesh or even finer, as the cementing material to agglomerate anthracite coal that had passed through a 60 mesh and over a 100 or 120 mesh sieve, and to agglomerate anthracite coal most of which was 60 to 100 mesh but which in some cases contained some 20 mesh material, and in others contained anthracite coal that had passed through $\frac{1}{4}$ or

$\frac{1}{2}$ inch mesh sieves, and over 10 or 20 mesh sieves. In mixtures of these last described types the presence of the coarser coal particles assists in obtaining good results with relatively low percentages of the coking coal; hard and strong briquettes being produced when the bituminous coal forms but from 14 to 17 per cent by weight of the mixture, and fairly coherent briquettes being made from mixtures containing as little as 7.5 to 10 per cent of the bituminous coal.

The strength of briquettes depends to some extent upon the compression to which they are subjected. I have molded briquettes of these several types at pressures ranging from 200 pounds per square inch up to 10,000 pounds or more per square inch. While no exact rule or law has been worked out by which the effect of pressure used in molding can be predetermined, I have found that briquettes molded at 500 to 700 pounds pressure per square inch developed strengths upon coking of 2300 to 2500 pounds per square inch, while briquettes made of the same material but which were molded at a pressure of 7000 to 7500 pounds per square inch, developed strengths of 4000 to 6000 pounds per square inch, thus showing, in these cases at least, that a great increase in the compression pressure produced a relatively smaller increase in the strength of the resulting product.

In agglomerating or cementing materials by the method I have described, when the cementing material is one with a property of coking, the temperature used to effect coking materially affects the strength of the resulting product. In the manufacture of anthracite briquettes it may be desirable to use a relatively low temperature in order to retain a considerable proportion of volatile combustible constituents in the finished briquette. I have made many briquettes by coking at a dull red, or barely visible red, heat, and in some cases even below a visible red, and the briquettes produced have been sufficiently hard and strong to bear handling without disintegration. In making these briquettes the temperature has been raised sufficiently to drive off most, if not all, of the illuminating hydrocarbons (which are driven off from the coking coal forming the cementing material) at relatively low temperatures. I have thus made briquettes which retain 5 per cent or more of volatile combustible matter, chiefly hydrocarbons, but which may in part consist of hydrogen. These briquettes generally have a crushing strength from one-half to two-thirds the strength which is attained if the coking be conducted at a bright red or orange heat, or at a heat sufficient to drive off nearly all of the volatile combustible matter. These results indicate that where the maximum crushing strength is desired, as in the case

of metallurgical fuel, the coking should be finished at relatively high temperature; in other words, at a temperature comparable to that used in the commercial manufacture of coke. Where, however, there does not exist any reason why maximum strength is necessary or desirable, coking can be done at a much lower temperature. The advantages of coking at lower temperature are a reduction in the cost of the coking process due to operation of the coking or baking ovens at a lower temperature, a reduction in the time necessary for the completion of the coking process and an increased fuel value of the product due to the retention of the combustible volatile matter. There is also some advantage in the domestic use of briquettes so made due to the free burning nature of briquettes containing an appreciable percentage of volatile combustible matter. It is, however, in many cases desirable that the coking should proceed far enough to expel the illuminating hydrocarbons which are the cause of the smoke and soot in burning bituminous coal, thus producing a briquette which is practically smokeless.

It will be understood that the time required for agglomerating materials when a coking coal is used as the cementing material, will depend upon the volume or mass of material introduced into the coking or baking oven, and upon whether the material is to be produced in massive form suitable for metallurgical use, or in the form of large or relatively small briquettes for steaming or for domestic use. In the manufacture of briquettes it is especially desirable to reduce the time required for coking to a minimum, because the process is not only cheapened thereby, but the coking can usually be completed with less loss of volatile combustible matter than where the coking period is prolonged. In the case of small briquettes weighing from $1\frac{1}{2}$ to 2 ounces, I have obtained satisfactory results by coking for periods of from 15 to 25 minutes in a muffle furnace heated to a dull red heat. In manufacturing on a large scale the time required for coking of such briquettes may perhaps be materially reduced below these figures, but I am not able to state with exactness the time required to produce the most satisfactory results, as this will vary with the character of the materials used, the size of the briquettes and the manipulation of the materials during manufacture.

It will be understood that if coking or baking of the cementing material be employed it should be carried out in a non-oxidizing atmosphere. After the temperature rises to a point where volatile combustible gases are given off, the gases so produced will of course furnish a non-oxidizing atmosphere, and this will continue during the baking or coking period. It is,

however, important that oxygen be excluded from the coking or baking ovens so far as possible. In the early stages, during which the briquettes are becoming heated, the gases or vapors given off by temporary binder used, whether this be oil or water, will of course tend to expel atmospheric oxygen, so that the briquettes will be surrounded, if water be used as the binder, by an atmosphere of water vapor, or if oil be used as a temporary binder, by an atmosphere of vapor of the oil so used. In the manufacture of most of the briquettes made, I have coked or baked them in a cylindrical sheet iron container with a lid or cover having a slip fit. The molded briquettes were placed within such container and the container introduced into a muffle furnace as before described. The looseness of fit of the cover of these containers permitted the escape of steam, volatile hydrocarbons and gases given off during the coking process, so that the briquettes as made were, after the coking process commenced, continuously in an atmosphere consisting of the volatile hydrocarbons given off by the coking coal forming the cementing material, and the presence of these gases in the container would permit only very small quantities of oxygen to gain access to its interior, thus simulating the conditions present in the interior of by-product or retort coke ovens.

In the making of small briquettes pressure is generally an essential part of the molding process, and the use of a temporary binder will usually be found advantageous. It is, however, possible in some cases to mold briquettes of sufficient strength to bear handling during manufacture, without the aid of a temporary binder, and thus to produce a product sufficiently strong to bear some handling without objectionable breakage. Agglomerated fuel suitable for steam making and similar uses, and perhaps for smelting, may be made by the use of my invention without the application of pressure prior to coking, the use of a temporary binder, or special grading of the granular material to be agglomerated, but generally speaking the strength of the product will be increased by pressure prior to coking, by use of a temporary binder and by preparation of the granular material as to size. I have, however, made a fair grade of agglomerated or cemented fuel from anthracite fines mixed with 15 to 25 per cent of more finely ground bituminous coal, without the addition of a temporary binder, the application of pressure or grading of the granular aggregate as to the size of its particles.

In the examples above given in which graded materials containing coal $\frac{1}{4}$ inch and 6 mesh, between 20 and 120 mesh and be-

tween 60 and 120 mesh were agglomerated with bituminous coal ground to pass through 120 mesh and which contained all the finer material produced by such grinding, including the impalpable powder or dust so made, the number of particles of the cementing material was in every case very much greater than the number of particles of the coal that was agglomerated, the number of particles in a given quantity of material theoretically increasing inversely as the cube of the diameter of the particles. For example, a given weight of 20 mesh size will have theoretically and approximately 64 times as many particles as an equal weight of 5 mesh particles; the same weight of 80 mesh size will similarly contain 64 times as many particles as the 20 mesh size and 4096 times as many particles as the 5 mesh size, and 160 mesh size will contain 8 times as many particles as the 80 mesh size, 512 times as many particles as the 20 mesh size and 32768 times as many particles as the 5 mesh size, so that if a graded mixture be made consisting of 25 per cent of anthracite coal averaging 20 mesh, 50 per cent of anthracite coal averaging 80 mesh and 25 per cent of bituminous coal averaging 160 mesh, every unit of such mixture containing 1 piece of 20 mesh coal, will theoretically contain 128 pieces of 80 mesh coal and 512 pieces of 160 mesh bituminous coal, the particles of bituminous coal therefore outnumbering the particles of anthracite coal about 4 to 1.

In carrying out the various applications of my invention, the details in each case should be varied to meet the requirements of the service, the character of fuel desired, and the shocks and attrition to which it may be subjected in handling, transportation and during combustion, sufficient strength to bear stresses and attrition being desirable if not essential. These requirements are at a minimum when the agglomerated fuel is burned without removal from the furnace in which agglomeration is effected and are at a maximum when long distance transportation and repeated handling and rehandling are unavoidable, as in fuel made for export to foreign countries. The quantity of cementing material to meet these requirements, grading as to size, compression before coking, the use of a temporary binder and the temperature at which agglomerating is effected can be varied as above described, the advantage of my invention being that in every case the necessary strength can be secured with a smaller quantity of the cementing material than with any method heretofore used, and in some cases, such as in direct firing, especial grading, compression and the use of a temporary binder may not always be necessary to secure satisfactory results.

While the manufacture of a fuel by agglomerating or cementing anthracite fines, coke breeze and the like is the most obvious application of my invention, the material so produced has characteristics that may fit it for other use. As already described this material may be thus made of great strength, as shown by briquettes I have made with a crushing strength of from 1200 up to 8400 pounds per square inch. It may be possible thus to produce material stronger than any I have yet made. This material is quite porous and may therefore be suitable for use as a filtering medium; it can be molded and produced in almost any desired shape or form; it is of light weight, usually from 60 to 70 pounds per cubic foot, its lightness and strength fitting it for use as a structural material; it is not attacked by ordinary acids or alkalis and is infusible except at extremely high temperature. Such properties may adapt it to many industrial uses.

In this specification and the claims hereof the terms "materially smaller" and "appreciably smaller" used in describing the relative size of the particles of coking material, are not intended to mean that all of the particles of coking material should or must be smaller than all of the particles of the granular or non-coking material, these terms being used to mean that the average size of the particles of coking material shall be materially or appreciably smaller than the average size of the granular or non-coking material which is to be agglomerated or cemented. I use the term "non-coking materials" in the claims hereof to mean materials such as anthracite or other non-coking coal, coke, ores and the like. The terms "coking material" and "cementing material" in this specification and the claims hereof are intended to include any material such as coal, or other solid hydrocarbons, and the like, which when heated have the property of coking or of agglomerating into a coherent cementing substance.

I do not claim the use as agglomerating materials of tar, pitch, asphalt, oil residuum or the like which are liquids or become liquid upon increase in temperature, except in combination with my herein described invention.

It will be clear from the foregoing description of my invention that one of its chief objects is the production of a strongly agglomerated product from a mixture of anthracite coal or coke and coking coal, and that the strength of this product largely depends upon the strength of the coke or coke-like material which results from subjecting the coking coal to a temperature sufficient to coke it, or to convert it into a coke-like substance because this coke not only acts as a cement but it also forms a

matrix occupying more or less completely the voids or interstices between the particles of anthracite coal or coke, and the strength of the product must always depend largely upon the strength of this matrix. If this matrix be weak the product will be weak and vice-versa.

The coking of any coking material converts it into a more or less porous coke, or coke-like substance, in which the pores are cells, which may or may not communicate with each other, produced by the gases evolved from the volatile constituents of the coking material during the coking process. In coking, the individual particles of coking material, if not separated from each other by any foreign body, coalesce or run together to form the more or less porous material called coke. Some coking coals swell in coking while others shrink. The strongest coke is usually produced from materials that swell but slightly, or that shrink, during coking. It is well known that the volume and the porosity of coke depend largely upon whether the material during coking is free to expand, that is, is practically unrestrained, or whether it is under compression from its own weight, or under pressure or restraint from any other source. Coal that produces a good, strong, relatively dense and slightly porous coke, when its expansion is restrained during coking, as in a by-product coke oven, may produce a light, weak, spongy and very porous coke if coked under conditions permitting expansion in one or more directions.

As a particular object of my invention is to produce a strong agglomerated product with a minimum quantity of coking coal, this end can therefore best be realized by the use of a coking coal that under proper conditions will produce a strong coke, and by providing the conditions under which this result will be attained. The experiments described herein were all made with coking coal of this character. The agglomerated products I have produced with maximum strength, as already described, show a sufficient quantity of moderately porous coke, or coke-like substance, to substantially fill the voids between the particles of granular material, this being readily seen by examining a ground and polished section of the briquettes so made, the agglomerated product being seen to consist of granular anthracite coal held together by a matrix of more or less porous or cellular coke. In the agglomerated products of less strength, the matrix of coke is not so strong, the cells in the coke are larger and more numerous, the walls of the cells are not so

strong, and the product is correspondingly less strong, this condition being clearly shown by some of the briquettes already described.

Hence one of the most important elements of my invention is to provide conditions under which, with a minimum quantity of coking coal, the coked residue of this material forming the matrix will be of maximum strength. As already described this object is attained by reducing to a minimum the percentage of total voids and also the size of the individual voids or interstices between the particles of anthracite coal, and by so doing the swelling of the coking coal during coking is restrained by the relatively small size of the voids and not being free to expand, the cells formed are relatively small and the cell walls relatively thick, the coke matrix consisting of relatively dense, strong coke of relatively low porosity, this object having been attained by grading of the sizes of the particles of anthracite coal to reduce the percentage of total voids and also to reduce the size of the individual interstices of which the voids are composed.

The exact quantity of finely divided coking material necessary to produce any desired result cannot accurately be determined, except by actual trial, but I have sufficiently indicated the procedure to enable anyone conversant with the art to carry out my invention in practice.

I do not herein claim the method of agglomerating materials herein described as I have claimed this in an application for patent filed September 27, 1920, Serial No. 413,194, of which said application, this application is a division.

Having described my invention, I claim, 1. A composition of matter consisting of non-coking material cemented together by the coked residue of coking coal, the average size of the particles of said coking coal being materially smaller than the average size of the particles of non-coking material.

2. The composition of matter of claim 1, said non-coking material constituting at least 70 per cent of the article.

3. The composition of matter of claim 1, in which the average size of the particles of said coking coal is not larger than the average size of particles that will pass through a 120 mesh sieve.

In testimony whereof I have hereunto signed my name at Philadelphia, Pennsylvania, this first day of June, 1921.

THOMAS M. CHANCE.

Witnesses:

H. M. CHANCE,
CARL K. SCHULZE.