

UNITED STATES PATENT OFFICE.

WILLIAM C. ARSEM, OF SCHENECTADY, NEW YORK, ASSIGNOR TO GENERAL ELECTRIC COMPANY, A CORPORATION OF NEW YORK.

CHEMICAL PROCESS.

979,363.

Specification of Letters Patent.

Patented Dec. 20, 1910.

No Drawing. Original application filed July 2, 1906, Serial No. 324,408. Divided and this application filed September 16, 1907. Serial No. 393,227.

To all whom it may concern:

Be it known that I, WILLIAM C. ARSEM, a citizen of the United States, residing at Schenectady, county of Schenectady, State of New York, have invented certain new and useful Improvements in Chemical Processes, of which the following is a specification.

My present invention relates to the treatment of metallic compounds, and my present application is a division of application Serial No. 324,408, filed by me July 2, 1906.

My invention includes the reduction of metallic compounds in a vacuum or inert atmosphere, with the production of pure metal.

In performing the reduction of the metallic compound I prefer to make use of the vacuum furnace shown in U. S. Patent No. 785,535, issued to me March 21, 1905. This patent discloses a furnace of the resistance type in which the heating conductor is a helix of carbon provided with a central aperture through which a crucible may be introduced containing the charge to be treated. These parts are all inclosed in a vacuum tight metal case connected permanently with a vacuum pump and provided with external cooling means and other auxiliaries. The furnace is provided with a peep hole through which the reactions within may be observed, and the pump is provided with a manometer for indicating the state of the vacuum within the furnace.

In carrying out the first step of my process I mix a reducible compound of the refractory metal, such as the oxid, with a finely divided reducing agent such as aluminum or carbon and I introduce this mixture into the crucible of the vacuum furnace, and after exhausting the air I heat up the charge until the desired reduction is effected. If a pure metal is desired I so proportion the charge that the carbon will be entirely used up in the reduction of the oxid without leaving a residue of unreduced oxid. In order to insure a good contact between the particles of carbon and of oxid I find it sometimes desirable to compress the mixture into briquets or lumps. This intimate contact between the particles facilitates the reaction.

The process of reduction may be closely watched by means of the manometer, for when reduction begins, carbon monoxid is evolved and the gage reading increases, and

after the completion of the reduction the carbon monoxid ceases to be formed and the vacuum rapidly improves. The vacuum pump is maintained in operation throughout the run, and the pressure within the inclosure never exceeds a few millimeters of mercury. By the above method of procedure I may reduce the oxids, sulfids, or other compounds, of such refractory metals as tungsten, tantalum, vanadium, or molybdenum, and obtain the metal in a pure state.

I am aware that it has been heretofore proposed to reduce the oxids of some of these metals with carbon in an arc furnace in the open air, but the method I have above outlined possesses many advantages and yields a superior product. In an arc furnace the temperature is variable and not easily controlled and the metal becomes fused and, unless a large excess of oxid is used, takes up carbon. In the vacuum furnace, the ease of temperature control makes possible the reduction of the oxid at a temperature lower than the melting point of the metal. Contamination with graphite is thus avoided and the unfused powder is much more pure than would be the case if fusion had occurred. The metal is ordinarily obtained in a coherent block. In a vacuum, oxidation by air is not possible, and the work may be carried on with accurately weighed quantities. By proper adjustment of the amounts of carbon and metallic oxid, an exceptionally pure metal may be produced.

In addition to the advantages above pointed out, it should be noted that reduction *in vacuo* may take place at a temperature lower than the same reduction would take place at atmospheric pressure. This is probably due to the fact that when the surrounding medium is of low density, as in the vacuum, the reducible substance vaporizes and comes into chemical contact with carbon at a lower temperature.

From this description of my process it will be understood that the material to be reduced is heated by radiant heat given out by a resistance conductor within the furnace. The reducible material is never traversed by current, and is, therefore never subjected to the sintering or fusing action which sometimes takes place when current passes through a powder; a fusing action probably due to minute arcs between adjacent and

poorly contacting particles. By the use of radiant heat, the charge is evenly heated, and furthermore, is protected from any tendency toward electrolysis or electrolytic action by the heating current. I have pointed out somewhat in detail the advantages of using radiant heat, as I consider that this method of heating contributes materially to the success of my process, especially when dealing with such compounds as the oxids of tungsten, tantalum, or molybdenum, and the manufacture of filaments therefrom. These metals are very easily contaminated with carbon, and are very difficult to purify, and I consider that anything which contributes materially to the purity of the product is of importance.

In the general process above outlined I have referred to the use of carbon as a reducing agent and I now desire to point out certain advantages which arise from using this carbon in an exceedingly fine state of sub-division. In reducing metals from their oxids by carbon the purity of the product as regards freedom from carbon depends both on the vapor pressure of the oxid at the reduction temperature and also on the size of the particles of carbon. If the oxid is volatile and the metal does not fuse at the reduction temperature, each grain of metal is a pseudomorph of a grain of carbon, since the metal is produced when the vapor of the oxid diffuses into the carbon particles. Hence, if the grains of carbon are too coarse, they leave centers or cores of carbon which have not been reached by the diffusing oxid. If the oxid is non-volatile, the reduction takes place only at the surface of contact between the grains of carbon and oxid, and hence complete reaction between the oxid and carbon to form a pure metal can only take place when the particles are very small.

To secure carbon in a very finely divided condition I first obtain a deposit of "lamp black" from volatile hydrocarbons and I then heat the lamp black to a very high temperature in a vacuum to remove all gases and volatile hydrocarbons, or I may heat it to a temperature approximating perhaps 3000 or 4000 degrees centigrade to graphitize the carbon. The resultant product is an absolutely pure carbon in a fine state of division. A much more perfect mixture of oxid and carbon can be obtained

with this fired lamp black than with any other kinds of carbon with which I have worked, and furthermore, the reduction of oxids takes place more rapidly than with other forms of carbon.

As previously stated, the metal obtained by the reduction of the oxid is in the form of grains having the same size and shape as the original carbon particles; consequently, by using lamp black, I can obtain the refractory metal in a powdered form much finer than any that has heretofore been produced.

What I claim as new and desire to secure by Letters Patent of the United States, is,—

1. The process which consists in reducing a metal compound in a vacuum by heat treatment while in contact with graphitized lamp black.

2. The process which consists in mechanically mixing a finely divided refractory metallic oxid with carbon deposited from a volatile compound and subsequently fired, and heating said mixture to cause a reduction of said metal compound.

3. The process which consists in producing a carbonaceous deposit from a volatile carbon compound, heating said deposit to produce pure graphite in a finely divided condition, mixing said graphite with a reducible compound, and heating said mixture in a vacuum to effect reduction.

4. The process which consists in mixing an oxid of tungsten with a dry powdered reducing agent and then heating said mixture in a vacuum by radiant heat to cause reduction of said oxid.

5. The process which consists in forming an intimate mixture of a powdered reducing agent and a refractory metal oxid, and heating said mixture to a reducing temperature in a vacuum by means of a resistance conductor insulated from said mixture.

6. A product consisting of finely divided refractory metal, the grains of which are pseudomorphs of graphitized lamp black.

7. Powdered tungsten the grains of which are pseudomorphs of lamp black particles.

In witness whereof, I have hereunto set my hand this 13th day of September, 1907.

WILLIAM C. ARSEM.

Witnesses:

BENJAMIN B. HULL,
MARGARET E. WOOLLEY.