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METHOD OF MAKING MAGNESIUM

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This invention relates to a new and advantageous method for producing magnesium by reduction of magnesium oxide, and relates more particularly to an improvement on the known carbon reduction of magnesium oxide-containing bodies.

Many attempts have been made to develop a commercial process for the production of magnesium from compounds and mixtures containing its oxide, based upon the reaction:

$$MgO+C \rightarrow Mg+CO \qquad (A)$$

It has long been recognized that the prime difficulty inherent in the above-stated simple reaction is the ease and rapidity with which the reverse reaction occurs:

$$MgO+C \leftarrow Mg+CO \qquad (B)$$

It has been proposed to dilute and to cool the gas-vapor mixture of CO and Mg with large volumes of a gas inert to the magnesium vapors, such as hydrogen, to minimize the possibility of obtaining magnesium oxide as a reversion product.

Another proposal of the prior art includes the step of bringing the Mg+CO vapor mixture into intimate contact with an extremely large and relatively cold condensing surface with the object of chilling the vapors rapidly through and below the temperature range in which the reverse reaction (B) occurs, so that little reversion of Mg to MgO takes place. Yet another prior proposal includes the application of a quick quenching of the hot vapor mixture with a cool hydrocarbon oil, or other inert liquid, either as a mist or as a liquid film, to condense the magnesium and thus remove it from contact with carbon monoxide.

We have found that each of the prior proposed methods, while reducing slightly the extreme tendency of the mixture of magnesium and carbon monoxide, or other oxidizing gas, to revert to magnesium oxide, still provides a condensate wherein at least about 20 per cent of the magnesium originally liberated in the reduction zone is reconverted to magnesium oxide before it can be recovered as metal. Each of the prior methods outlined above yields a condensate in a finely-divided state, except when the oil quench is used, which makes it explosively dangerous to handle in converting it to compact metallic magnesium. The prior method which involves the quenching action of a hydrocarbon oil reduces the hazard of explosion while handling the finely-divided product, but adds to the difficulty of recovering the magnesium either by vaporization or by melting under a protective flux of fused inorganic salts.

Among the objects of the present invention are (1) to condense magnesium from the vapor mixture containing magnesium, carbon monoxide, and possibly other reactive materials resulting from the thermal reduction of magnesia-containing raw materials with carbon, with substantially less than the usual amounts of the solid reversion products magnesium oxide and carbon, and (2) to obtain magnesium from the said carbon reduction reaction in such form that the product is at most but slightly pyrophoric and may easily be recovered as compact magnesium metal. Other related objects will become apparent from the following detailed description of the invention.

According to the invention, a volatile hydrocarbon, in the gaseous or dry vapor state, is added to the hot vapor mixture comprising magnesium and carbon monoxide liberated by the carbon reduction of magnesium oxide, resulting in a reaction with the magnesium vapor to form a carbide of magnesium (either MgC_2 or Mg_2C_3, or both) and minimizing formation of the usual reversion products magnesium oxide and carbon. The condensate, containing a magnesium carbide, does not consist entirely of such carbide, but is a mixture of metallic magnesium and magnesium carbide, and at most but small traces of magnesium oxide, and contains from as little as 20 per cent or less up to as much as 80 per cent of magnesium, the balance being substantially entirely the carbide. Further, the condensate comprising magnesium and one or more of its carbides may be heated to melt or to vaporize the magnesium in known manner, utilizing the ordinary precautionary measures to prevent oxidation of the metal. The magnesium carbides decompose at or below the temperature at which magnesium is vaporized, and yield a mixture substantially composed of magnesium and carbon, from which the magnesium can be vaporized and then condensed in known manner as compact liquid or solid metallic magnesium. It is not necessary to distill the magnesium to recover it from its carbide, as a simple remelting operation, in the presence of a customary inorganic salt flux, results in gradual liberation of magnesium from its carbide and causes coalescence of all the magnesium under the same conditions of temperature as those under which magnesium alone is ordinarily melted in a flux bath.

In a preferred method of carrying out the invention, a magnesium oxide-containing material, preferably in finely-divided form, is mixed with carbon and the mixture heated to the well-known reduction temperature, whereby magnesium vapor and carbon monoxide are liberated. There is then introduced into the vapor space to mingle with the magnesium-containing vapor stream a gaseous cooling medium which may be all gaseous or dry vaporized hydrocarbon, or it may be large-

ly hydrogen or other inert gas with just enough hydrocarbon to ensure the prevention of the reverse reaction between magnesium and carbon monoxide as well as to ensure the formation of some magnesium carbide. The amount of gaseous diluent introduced in the vapor stream may vary widely, but is commonly 40 to 50 volumes for each volume of Mg+CO. Of this, as much as 80 per cent may be hydrogen, and the rest gaseous hydrocarbon, though the aim is to produce as much magnesium carbide as possible, and, consequently, the entire diluent employed may be hydrocarbon if this is economically feasible. The magnesium carbide-containing product is not nearly as pyrophoric as magnesium alone, and can be handled safely and economically for the production of magnesium by distillation or by fusion in a flux bath as previously disclosed.

The present method, which includes the use of a hydrocarbon gas or dry vapor to convert magnesium at least in part to a carbide and to minimize reversion of magnesium to its oxide, may be employed together with any of the prior known methods. The use of a mixture of hydrocarbon gas and hydrogen or other inert diluent has already been described. Similarly, the hydrocarbon gas may be used in those cases where the vapors of magnesium and carbon monoxide are to be quenched in a liquid hydrocarbon. In such cases, the gaseous hydrocarbon which reacts to form the magnesium carbide is preferably added to the vapor mixture just before the latter is quenched in oil. The magnesium is converted instantaneously to carbide, and will be collected as a solid, suspended in the inert quenching liquid.

When the procedure to be used for quenching the Mg+CO mixture involves sudden chilling thereof on a large, clean and relatively cold surface, the hydrocarbon gas may be mixed therewith just before or at the same time that the vapor impinges on the said cooling surface. The condensate, containing substantial amounts of magnesium carbide, is in a finely-divided form which can be scraped from the condensing surface and then heated to liberate magnesium from the carbide and either to sublime or to melt the magnesium therein for recovery thereof in compact form.

Regardless of the precise method employed, the hydrocarbon gas should be introduced into the magnesium vapor stream in a manner to secure intimate mixing and commingling of the two streams.

The temperature of the hydrocarbon gas at the time of its mixture with or introduction into the magnesium vapor stream may vary over a considerable range. This temperature may be at or near room temperature, or it may be as high as 300° C. or higher. The lower temperatures are preferred, as they help the gas to serve the dual function of cooling agent and chemical reagent. Shock quenching of the magnesium and carbon monoxide vapor with a gas including the hydrocarbon gas, makes desirable a low temperature in the introduced gases. In any event, the temperature of the hydrocarbon should be high enough to prevent its condensation as a liquid or mist in the system before it has had opportunity to react with magnesium to produce a carbide. Regardless of the temperature of the introduced dry hydrocarbon gas, it reacts with the hot vaporous magnesium to form carbides.

The hydrocarbons which can be used in the invention to form magnesium carbides include such normal aromatic liquids (which must be heated to form a dry vapor) as benzene, toluene and xylene, and such aliphatic hydrocarbons as methane, ethane, butane, octane, acetylene, the butylenes, and the like or mixtures thereof. The preferred hydrocarbons are such commonly available gases as methane and ethane, which occur in natural gas. Natural gas itself can be used.

Regardless of what theory may be advanced to explain the phenomena occurring here, the fact remains that the introduction of a hydrocarbon gas (or dry vapor) to the stream of vaporized magnesium and carbon monoxide, is an effective means of suppressing the reversion reaction which has heretofore hampered the carbon reduction method of making magnesium from its oxide. One function of the hydrocarbon is undoubtedly that of a diluent to suppress reaction between magnesium and carbon monoxide by lowering the concentration of both these potential reagents with respect to one another in the gas stream. The function of the hydrocarbon gas which is important here is the formation of magnesium carbide, which may be explained by any of several possible reactions, the most probable of which may be illustrated somewhat as follows:

$$3CH_4+2Mg \rightarrow Mg_2C_3+6H_2 \qquad (C)$$

At present we endorse no particular theory, resting our claim to invention primarily on the advantageous and unexpected results obtained when operating according to the herein described method, as particularly pointed out in the appended claims.

We claim:

1. In a method for the carbon reduction of magnesium oxide, wherein a mixture of magnesium vapor and carbon monoxide is produced, and the vapor is cooled to condense magnesium, the step which consists in introducing a dry hydrocarbon gas into the hot magnesium-carbon monoxide mixture to suppress the reverse reaction to magnesium oxide and to convert a substantial amount of magnesium vapor to a carbide of magnesium.

2. The method of recovering metallic magnesium from the magnesium carbide-containing product produced according to the method of claim 1, which comprises heating the said carbide to its decomposition temperature.

3. The method which comprises heating a mixture containing carbon and a magnesium oxide-containing body to a reaction temperature to generate magnesium vapor and carbon monoxide, introducing into the magnesium vapor stream a diluent gas containing a dry gaseous hydrocarbon in quantity sufficient to suppress the reverse reaction between magnesium and carbon monoxide, and to convert a substantial amount of the magnesium vapor to a carbide of magnesium, condensing the carbide and remaining magnesium vapor, and thereafter heating the magnesium carbide-containing condensate to the decomposition temperature of the carbide and separating free magnesium from remaining carbon and carbide.

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