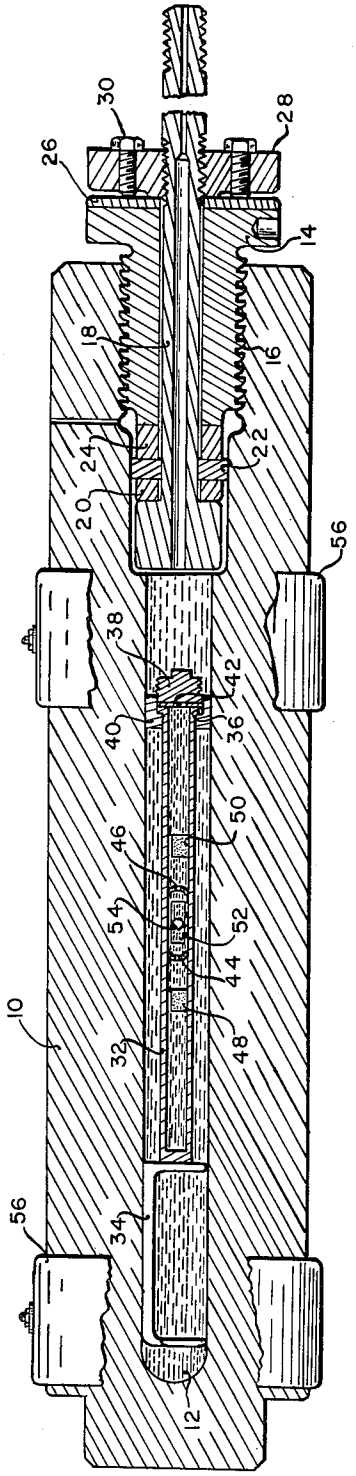
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CRYSTAL GROWING APPARATUS

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CRYSTAL GROWING APPARATUS

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This invention relates to apparatus for growing crystals synthetically. More particularly it is concerned with a novel structure for the hydrothermal growth of ferrite crystals.

Ferrites in general have the formula $MeFe_2O_4$, where the Me designates such metals in the devalent state as Fe, Ni, Co, Zn, Mn, Cu and Mg. Another mode of writing the formula is $MeO \cdot Fe_2O_3$, indicating that the ferrites are compounds of composite oxides, of which the divalent metal oxide is one and ferric oxide the other. The presence of a plurality of divalent metals gives a mixed ferrite (Me_1, $Me_2O \cdot Fe_2O_3$. Minerals and synthetic products are mostly nonstoichiometric, containing an excess of Fe_2O_3.

Ferrospinels crystallize in the cubic system. The most frequent structure among the ferrites is the octahedron. Eight elements of $MeO \cdot Fe_2O_3$ form a unit cell whose lattice constant for all of the spinels is in the immediate neighborhood of 8.40 A.

The term hydrothermal process implies that the system operates at high temperature and high pressure, and that one of the ingredients of the charge is an aqueous medium. For practical growth rates the required temperature and pressure ranges of a system are 450°–550° C. and above 20,000 p.s.i. respectively. In the hydrothermal processes, thick walled, metal vessels are used. They must have reasonable mechanical strength and resistance against excessive creep and corrosion under the severe operating conditions. The closures must be so designed as to be leakproof.

Although the latter problem has been satisfactorily solved, the corrosion of the autoclave used in growing the crystals still presents difficulties. Various steel alloys have been tested in these cases, but the corrosion of each has introduced new variables into the grown crystal. Accordingly, any precise analysis of the growth was difficult to obtain. Attempts have been made to plate the inner walls of the autoclave with a noble metal, e.g. silver. This was unsuccessful since "bleeding" through the plating occurred under the extreme temperatures and pressures requisite to the process.

Therefore, it is a primary object of the invention to provide an improved crystal growing apparatus which effectively eliminates the autoclave corrosion problem.

More specifically, it is an object of this invention to provide an apparatus wherein crystal growth takes place within a silver tube positioned inside of the autoclave.

The above and still further objects, advantages and features of my invention will become apparent upon consideration of the following detailed description thereof, especially when taken in conjunction with the accompanying drawing, in which the single figure shows a sectional view of an autoclave and silver tube constructed in accordance with my invention.

In the drawing, an autoclave is shown to have a body portion **10**. The autoclave is constructed of Inconel, a commercial nickel alloy. The particular autoclave employed was designed for a maximum pressure of 25,000 p.s.i. at 500° C. One end of the body **10** is closed. The opposite end is open, and a chamber **12** extends from said open end through almost the entire length of the body **10**. A cover **14** closes the open end of the chamber **12**. Threaded sections on said cover and said body engage

at **16**. The cover **14** is annular in shape, and a pressure fitting **18** passes through the central aperture and extends from both ends thereof. The outer end of said fitting is threaded for connection to a pressure gauge. At its inner end, the fitting is enlarged to fit the chamber in body **10**. A series of three packing members **20**, **22**, and **24** are compressed between said enlarged end and the inner end of cover **14**. The members **20** and **24** are conventional, high pressure, sealing material, while the middle member **22** is formed of silver. An annular thrust washer **26** fits over the outer end of the fitting **18** and abuts the cover **14**. A lock nut **28** is threaded on said fitting and holds the washer **26** in place. A plurality of set screws **30** are carried in said lock nut and serve to increase the pressure on the thrust washer **26**.

Within the chamber **12** is a silver tube **32**. Said tube is free floating within said chamber between the enlarged end of the fitting **18** and a stop framework **34**. The tube **32** is formed with one end thereof open. Said open end has a flange **36** formed externally thereon. A closure member **38** is threadedly engaged by an annular nut **40**. Said nut has a flange thereon which engages under the tube flange **36** whereby the closure may be tightened into place. A sealing gasket **42** is compressed between the closure **38** and the flange **36**.

The tube **32** is divided into three compartments by a pair of apertured partitions **44** and **46**. The compartments formed at opposite ends of the tube each receive one of the component oxides. These are seen at **48** and **50**. The center compartment has a mounting frame **52** therein, and a seed **54** is located on said frame. The apertures through the partitions permit free communication between the several compartments.

Tube **32** contains a suitable growing solution in an amount that will develop the desired pressure at the operating temperature. The chamber **12** within the autoclave also contains said solution, or any suitable fluid, in an amount that will develop the said desired pressure at said operating temperature. In this manner the internal and external fluid pressures exerted on the silver tube at operating conditions are substantially equalized so that stresses on said tube are minimized. A pair of cylindrical heaters **56** are positioned around the body **10** and serve to produce the elevated temperatures required for operation.

With the above described apparatus, trial runs were made using a nickel spinel seed. The component oxides were Fe_2O_3 and NiO. In each of the runs detailed below, a growing solution of ½ Normal NH_4Cl was employed. In the first run the tube **10** was filled to 70%, and subjected to a pressure of 16,500 p.s.i. at 473° C. The original weight of the seed was 0.0467 gram, and the weight after completion was 0.0510 gram. The amount of growth was accordingly 0.0043 gram. In another trial, the tube was filled to 75%. The pressure was raised to 21,000 p.s.i. at a temperature of 476° C. With a seed initially weighing 0.0516 gram, a final weight of 0.1656 was obtained. Thus, the growth in this instance was 0.1140 gram.

From these figures, it is clear that successful crystal growth is achieved in the apparatus disclosed above. It is to be understood that the single detailed embodiment is merely illustrative of the principles of my invention. Numerous other arrangements may be devised by those skilled in the art without departing from the spirit and scope of the invention.

What is claimed is:

1. Crystal growing apparatus comprising an autoclave having an open-end chamber therein, a silver tube in said chamber, means dividing said tube into three communicating compartments suitable for containing a com-

ponent oxide in each of two of said compartments and a seed in the remaining compartment, said tube and the portion of said chamber surrounding said tube each being filled with an amount of liquid that will develop substantially equal pressures at the operating temperature, at least the liquid in said tube being a crystal growing solution.

2. Crystal growing apparatus comprising an autoclave having an open-end chamber therein, a sealed silver tube positioned in said chamber remote from the walls thereof, said tube being divided into a plurality of communicating compartments suitable for containing a component oxide in each of two of said compartments and a seed crystal in another of said compartments, a liquid in each of said tubes and said chamber in an amount that will develop substantially equal pressures at the operating temperature, at least the liquid in said tube being a crystal growing solution.

3. Crystal growing apparatus comprising an autoclave, a chamber in said autoclave and having one open end, a silver tube within said chamber remote from the walls thereof, a pair of partitions within said tube and dividing the latter into three compartments, a seed crystal in one compartment, different component oxides in each of the other compartments, growing solution in said tube, said chamber containing an amount of a fluid that will develop the same pressure as said solution at the operating temperature, and means sealing the open end of said chamber.

4. Crystal growing apparatus comprising an autoclave having an open-end chamber therein, a sealed silver tube within said chamber, a pair of apertured partitions dividing said tube into aligned compartments, a seed crystal in the central compartment, a metal oxide in each of the other compartments, growing solution partially filling said tube and said chamber, heating means surrounding said autoclave, and means sealing said open end.

5. Apparatus for growing ferrite crystals comprising

an autoclave, an open-end chamber in said autoclave, a sealed silver tube in said chamber, means dividing said tube into a plurality of aligned compartments, a ferrite seed crystal in one of said compartments, a metal oxide in another of said compartments, ferric oxide in still another compartment, a growing solution partially filling the portion of said chamber and surrounding said tube, and means sealing said open end.

6. Apparatus for growing ferrite crystals comprising an autoclave having an open-end chamber therein, a silver tube positioned in said chamber spaced from the walls thereof, a plurality of apertured partitions spaced within said tube, a ferrite seed in a compartment formed between two of said partitions, a metal oxide in a compartment formed adjacent one end of said tube, ferric oxide in a compartment formed adjacent the other end of said tube, growing solution in said compartments and in said chamber, annular heating means surrounding said autoclave, and means sealing the chamber in said autoclave.

7. Apparatus for growing ferrite crystals comprising an autoclave having a chamber therein, one end of said chamber being open, a sealed silver tube in said chamber, a pair of partitions in said tube dividing the latter into three aligned compartments, said partitions being apertured to provide communication between adjacent compartments, a metal oxide in one end compartment, a ferrite seed crystal in the central compartment, ferric oxide in the third compartment, a growing solution partially filling said tube, said chamber containing an amount of a fluid around said tube that will develop substantially the same pressure as said solution at the operating temperature, and means sealing said chamber.

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