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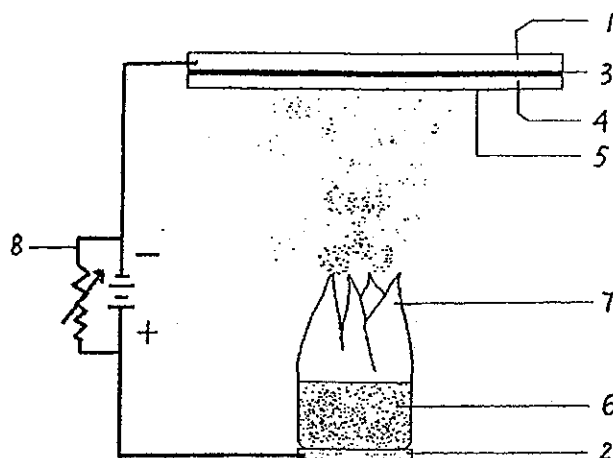
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[CN/CN]; 中国湖北省武汉市东西湖区吴家山革新大道南、五支沟西, Hubei 430040 (CN)。(74) 代理人: 永新专利商标代理有限公司北京办事处(NTD
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(54) Title: METAL COMPOUND MICROPARTICLES AND ULTRA-MICROPARTICLE POWDERS

(54) 发明名称: 火焰晶析法制备金属化合物微粒子或超微粒子粉体的方法



(57) Abstract: The present invention provides a process for manufacturing metal compound microparticles or ultra-microparticle powders by the flame crystallization process. The process includes sufficiently mixing the ions or precursors of metal salts or metal compounds with organic solvents, and flaming the mixed solution, to obtain the metal compound microparticles and ultra-microparticles through co-inflaming. The organo-metal salts may be a mixture of two or more kinds of organo-metallic salts. The growth rate and particle size of the microparticle can be controlled by adjusting the concentration of the mixed solution of the precursors and organic solvents. The obtained metal compound microparticles and ultra-microparticle powders can be used in a non-vacuum plating film process or in a powder collecting equipment to collect the powders produced by flaming.

[见续页]



(57) 摘要

本发明提供一种火焰晶析法制备金属化合物微粒子或超微粒子粉体的方法。该方法将金属盐或金属化合物的离子或母体与有机溶剂的充分混合，将混合溶液点火燃烧，经共烧获得金属化合物的微粒子或超微粒子粉体。有机金属盐可以是两种或两种以上有机金属盐的混合物。调整母体与有机溶剂混合溶液的浓度，可以控制微粒子生成的速度与粒子的大小。所制得的金属化合物微粒子或超微粒子粉体可用于非真空镀膜，或使用集粉设备，收集燃烧生成的粉体。

METAL COMPOUND MICROPARTICLES AND ULTRA-MICROPARTICLE POWDERS

BACKGROUND OF THE INVENTION

5 Field of Invention

[0001] The present invention relates to a method of metal compound microparticles and ultra-microparticle. More particularly, the present invention uses solution-gelatin process and flaming process to obtain high quality microparticles powders.

10 Description of Related Art

[0002] Generally speaking, using solution-gelatin transition process to make microparticles powders must to mix prepared powders matrix and organic solvent, and then keep still in oven for heating. The heat temperature is higher than flash temperature of solvent and lower than boiling point of solvent. So the heating temperature is the lower
15 the better. After more than ten hours or even several months, the solvent evaporated completely so as to obtain the powder-matrixes but not the final powders. During the evaporating process, mass of powder-matrixes accompanied solvent to evaporate.

[0003] As the manufacture process has heating treatment, the energy of heating accompanied raising the cost. Besides, the manufacturing process with evaporating makes
20 the solvent to bring out some powder-matrixes that cause a lot of loss in manufacturing process, powder pollution and further more serious air pollution. Its also pollutes the oven. Furthermore, the time of manufacture is longer, the powders will agglomerate and cohere

each other that increase the diameter of the powders and the quality of final products is decreasing.

[0004] Moreover, manufacturing powders can use the solvent-evaporating method, freeze-drying method, spray-drying method, spray-thermal decomposition, depositing
5 method, or flame with hydrogen and oxygen and drying method, but all of these methods need to increase the energy cost and make a lot of pollution. It is also very difficult to make the powders to become the micro-particles.

SUMMARY OF THE INVENTION

10 [0005] The present invention provides a process for manufacturing micro-particles or ultra-microparticles.

[0006] The present invention uses a flame crystallization process to manufacture microparticles or ultra microparticles.

15 A first aspect of the present invention provides a method of manufacturing microparticles or ultra microparticles, the method comprising the steps of:

(a) mixing metal ions and matrix of metal salts or metal compounds with organic solvents to obtain a mixing solution; and

(b) igniting the mixing solution directly to create a self-sustaining flame to produce
20 the microparticles and ultra microparticles.

[0007] The flame crystallization process as above described can manufacture the microparticles and ultra microparticles in a non-vacuum condition.

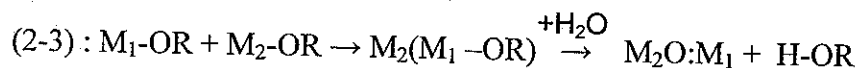
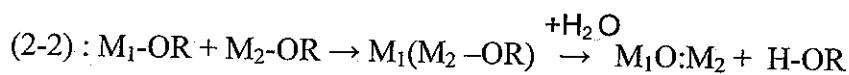
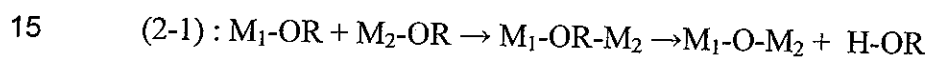
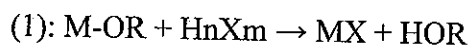
[0008] In one embodiment, the metal salts may include inorganic metal salts and organic metal salts, the metal compounds can include metal oxide, metal sulfide, metal
25 nitride, metal boron compound, metal selenium compound, metal phosphide, metal arsenide, metal

halide, complex metal oxide, complex metal sulfide, complex metal nitride, complex metal boron compound, and complex metal halide.

[0009] The inorganic metal salts may include metal phosphate, metal carbonate, metal sulfation, metal nitrate, metal borate and variety inorganic metal slats.

5 [0010] In one embodiment, organic metal salts may be chosen to be the metal salts. The first step is mixing the organic salts and hydride H_nX_m to make matrix, next step is sufficiently mixing the matrix and organic solvent to flame together and than the microparticles and powder of ultra microparticles is obtained. The X of formula H_nX_m can be oxygen, sulfide, nitrogen, selenium, phosphate, arsenide or boron. After mixing one
10 or two kinds of organic metal salts and fully stirring is obtained a mixture compound, we can input one or more hydrides with fully stirring to completely react and add organic solvent than flaming so as to obtain a micro-particle powder of complex metal compound.

The chemical formula is following;



, wherein M is metal, R is alkyl.

[0011] We can fully blend one or more then two metal-organic salts to mix or react.

20 Further adding one or more then two hydrides and also blend to get a first matrix compound within organic solvent. We can also fully blend one or more then two metal-organic salt to mix or react, focus on the exchanging the species of metal. Further adding

one or more than two hydrides and also blend to get a second matrix compound within organic solvent.

[0012] Further, we mix and fully blend the above two kind of matrix with solvent. After burning the mix matrixes, we can micro powder of complex metal composite
5 compound.

[0013] The hydrides of present invention can be hydroxide, sulfhydrate, boron hydride, nitrogen hydride, selenium hydride, phosphorus hydride, arsenic hydride, tellurium hydride, mixing gas with nitrogen hydride, or more than one kinds of hydride compound.

10 [0014] In another embodiment, inorganic metal salts can be chosen to be the metal salts. First step is to dissolving the inorganic metal salts or other metal compound into acid solution or alkali solution to make matrix, next step is sufficiently mixing the matrix solution and organic solvent to flame together and than the microparticles and powder of ultra microparticles is obtained.

15 [0015] If we use metal halide in the present invention, the first step is to dissolving the metal halide into organic solvent to be mixing solution; after burning, we can obtain the microparticles and powder of ultra microparticles.

[0016] The type of organic solvent as above described does not have limitation, but the best choice of the organic solvent which has flammable character, especially is able to
20 fully flaming without carbon particles while burning the organic solvent, like alcohol type organic solvent.

[0017] In one embodiment adjusting the concentrate of mixing solution that is metal salts or metal compounds mix with organic solvent can control the particle growing

rate and sizes. We can use scanning electron microscope or transparence electron microscope to observe or test the microparticle or ultra microparticle. According to the test result to adjust the concentrate of mixing solution.

[0018] In another embodiment, the mixing solution can be homogenized solution,

5 even the matrix solution may be added more over organic solvents before flaming the mixing solution, so as to adjust the ions of metal compounds in matrix solution, and the ions density and relative concentration of organic solvent. The process can easily control and adjust collision probability of each ion monomer in metal compound matrix that also decide the particle size and growth rate of microparticles or ultra microparticles, and
10 increase flame temperature.

[0019] While the concentrate of metal compound matrix is higher in organic solvent, the growth rate of microparticles (or ultra microparticles) is higher so the size of microparticles (or ultra microparticles) is bigger and the particle size distribution is broad.

On the contrary, in organic solvent, as the concentrate of metal compound matrix is lower;
15 the growth rate of microparticles (or ultra microparticles) is lower so the size of microparticles (or ultra microparticles) is smaller, and the particle size distribution is narrow. The quality of microparticles (or ultra microparticles) is better, but the cost of manufacture increases. The proportion of adding organic solvent and metal compound matrix is between 2:1(the formed micro-particles is smaller than 100nm) and 350: 1(the
20 formed micro-particles is smaller than 20nm).

[0020] The method of a further embodiment can use powder collected means in flaming mixing solution to collect the formed microparticles or ultra microparticle powders.

[0021] The microparticles or ultra microparticle powder formed in one embodiment can be metal oxide, metal sulfide, metal nitride, metal boron compound, metal selenium compound, metal phosphide, metal arsenide, metal halide, complex metal oxide, complex metal sulfide, complex metal nitride, complex metal boron compound, and complex metal halide.

[0022] A second aspect of the present invention provides a method of manufacturing microparticles or ultra microparticles, comprising the steps defined in the first aspect and further includes putting an electro above the flame in the electric field, and adjusting the intensity of electric field to control the emitting microparticles or ultra microparticles which is separated, injects to the electro.

[0023] This invention is utilized chemical reaction. Under long time and fully blending, the reacted compounds are fully orientated. After high temperature burning, we can get a micro powder. Those powders have the listed characteristic:

- 1.The size of the powders is very small.
- 2.High surface energy and easily to melted.
- 3.Dry powder and free to select related & suitable solvent.
- 4.Easy to modify, such as capsule modification, surface modification, etc. al.
- 5.We can directly get the crystal,(mono crystalline with solid crystal outer shape).

The quality of this compound powder is better than those made by physical method.

We can get numerous and high temperature micro powers under non-vacuum electroing and fire burning. Compared with traditional vacuum electroing, those powders have almost perfect orientated crystal structure due to processing under long time reaction. Further more, putting the electroed subject under fire with certain distance by non-vacuum physical vapor electroing, we can get a high quality vapor ing product.

[0024] The advantage of this invention is: Combined with solving-concentration method and easy firing method to get high quality micro (or ultra fine) powders.

[0025] This invention is processing under non-vacuum condition and with no special requirement. Thus the suitable sorts of vaporized subject are highly increased. Such as metal, glass, ceramics, plastics, leathers, drapery and papers, all of them can processed by this invention. The vaporized subject can be easily rotated and moved due to no limitation under non-vacuum condition. Therefore, we can process vapor electroing with different sizes and shapes by the method in this invention.

[0026] The pollution of this invention is very low and only produces few & pure carbon dioxide gas and the organic solvent can be fuel in flame process for saving energy. The microparticles or ultra microparticles is made by the present invention that is high quality.

[0027] In conventional vacuum vapor, only one metal can be vaporized, so if you want to vapor complex metal compound or more than one metal compound, must to use sputter equipment that is very expensive.

[0028] An embodiment can also separate perfect crystal microparticle powders that conventional vacuum vapor cannot make, so the high quality product can be made by the present invention. Beside, the electroing process of present invention is cost lower and speeds the working rate easily.

[0029] Reference will now be made in detail to the present preferred embodiments of the invention, examples of which are illustrated in the accompanying drawings. Wherever possible, the same reference numbers are used in the drawings and the description to refer to the same or like parts.

BRIEF DESCRIPTION OF THE DRAWINGS

[0030] Fig. 1 is a schematic diagram shows flaming process and a non-vacuum physical ing device separate crystals;

[0031] Fig. 2 is a schematic diagram showing another embodiment that is more precise method to control the crystals size to be separate by flaming process and a non-vacuum physical-electroing apparatus.

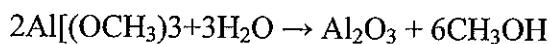
In the figure 1 and 2 includes electrode boards 1 and 2, heating board 3, electro 4, electro film powder 5, mixing solution 6 which is mixing matrix and organic solvent, flame 7, power 8, and cotton strip or capillary 9.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

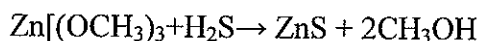
[0032] This invention is utilized organic or in-organic metallic compound to produce micro powder or ultra micro powder. The followers are some embodiments of this invention. Those embodiments are just the description of the invention but not the limitation of this invention. We can absolutely utilize other ways excepted described in this invention to produce the product.

Embodiment 1

[0033] First step is mixing organic salts $\text{Al}[(\text{OCH}_3)_3]$ and H_2O to make matrix, second step is fully mixing the matrix with organic solvent CH_3OH , than flaming to process non-vacuum vapor obtain Al_2O_3 microparticles or ultra microparticles.

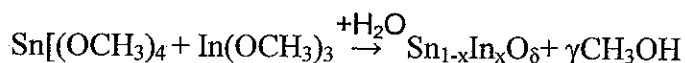
Embodiment 2

[0036] First step is mixing organic salts $\text{Zn}[(\text{OCH}_3)_3]$ and H_2S to make matrix, second step is fully mixing the matrix with organic solvent CH_3OH , than flaming to process non-vacuum vapor obtain ZnS microparticles or ultra microparticles.

Embodiment 3

[0035] First step is fully mixing $\text{Sn}[(\text{OCH}_3)_4]$ and $\text{In}[(\text{OCH}_3)_3]$, second step is adding H_2O continue to fully stir until reacting completely and than adding organic solvent CH_3OH ,

flaming in the non-vacuum vapor to obtain complex metal oxide, $\text{Sn}_{1-x}\text{In}_x\text{O}_\delta$ microparticles or ultra microparticles.



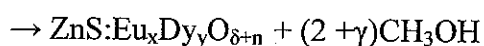
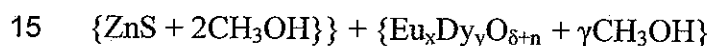
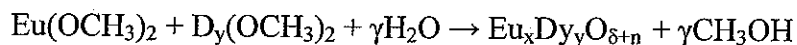
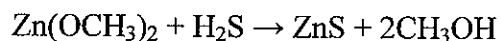
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Embodiment 4

[0036] The $\text{Zn}(\text{OCH}_3)_2$ is inputted H_2S to make a first matrix, ZnS (with CH_3OH).

Fully mixing $\text{Eu}(\text{OCH}_3)_2$ and $\text{Dy}(\text{OCH}_3)_2$ input H_2O and stir to react competently to make a second matrix (with CH_3OH). Next, mixing the first matrix and second matrix, fully

10 stirring and flaming in non-vacuum vapor obtain a complex metal compound, $\text{ZnS:Eu}_x\text{Dy}_y\text{O}_{\delta+n}$ microparticles or ultra microparticles.



Embodiment 5

[0036] Dissolving the copper oxide powders (crystal phase) into hydrochloric acid or hydrofluoric acid, the copper oxide powders will become non-crystal phase matrix.

20 Selecting optimal organic solvent (like methyl alcohol) to make mixing solution with metal compound matrix and organic solvent. Flaming the mixing solution processes non-vacuum vapor. Finally, the CuO microparticles powder is obtained.

embodiment 6

[0037] The first step is dissolving NaCl into methyl alcohol to make a mixing solution, the second step is flaming the mixing solution, or dissolving NaCl into water to make a concentrated salts water, adding a lot of methyl alcohol to make the mixing solution, and then flaming the mixing solution.

[0038] In the aforementioned embodiment 3 and 4 can implant varied metal ions.

[0039] It will be apparent to those skilled in the art that various modifications and variations can be made to the structure of the present invention without departing from the scope or spirit of the invention. In view of the foregoing, it is intended that the present invention cover modifications and variations of this invention provided they fall within the scope of the following claims and their equivalents.

WHAT IS CLAIMED IS:

1. A method of manufacturing microparticles or ultra microparticles, the method comprising the steps of:
 - (a) mixing metal ions and matrix of metal salts or metal compounds with organic solvents to obtain a mixing solution; and
 - (b) igniting the mixing solution directly to create a self-sustaining flame to produce the microparticles and ultra microparticles.
2. The method of claim 1, wherein the metal compounds selects from one of compounds as follows: metal oxide, metal sulfide, metal nitride, metal boron compound, metal selenium compound, metal phosphide, metal arsenide, metal halide, complex metal oxide, complex metal sulfide, complex metal nitride, complex metal boron compound, or complex metal halide.
3. The method of claim 2, wherein the metal salt is an inorganic metal salt that selects from one of compounds as follows: metal phosphate, metal carbonate, metal sulfation, metal nitrate, metal borate and variety inorganic metal salts.
4. The method of claim 2, wherein the method of manufacturing microparticles or ultra microparticles comprising the steps of:
 - (a) dissolving the metal ions and inorganic metal salts into acid or alkyl solution to make a matrix and then fully mixing with organic solvent to obtain the mixing solution; and
 - (b) igniting the mixing solution directly to create a self-sustaining flame to produce the microparticles and ultra microparticles.
5. The method of claim 2, wherein the mixing solution further has a metal halide is dissolved, and goes on.

6. The method of claim 1, wherein the steps of (b) further includes adding more over organic solvent into the mixing solution before flaming to adjust concentrate of the metal salts or the metal compounds in mixing solution so as to control growth rate and size of microparticles and increase flaming temperature.
7. The method of claim 1, 2 or 6 wherein the step of flaming the mixing solution further includes using a collecting mean to collect the microparticles or ultra microparticles that is separated by flaming.
8. The method of manufacturing microparticles or ultra microparticles, comprising the steps as the claim 1 described further includes putting an electro above the flame in the electric field, and adjusting the intensity of electric field to control the emitting microparticles or ultra microparticles which is separated, injects to the electro.