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SYNTHESIS OF SPHERICAL MICROPARTICLES OF YTTRIUM ALUMINUM SILICATE BY MEANS OF PARAFFIN OIL GELATION COLUMN

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Abstract

Radioactive glass microspheres of yttrium aluminum silicate have been used in cancer brachytherapy treatment. ***.*** *Glass particles with irregular shapes were produced by different methods and transformed into spherical shape (microspheres) by using two different processes in solid phase: A) re-melting in a metal cylinder B) spheroidization by gravitational fall in vertical tubular furnace. But we could produce a spherical micro particle by using paraffin oil gelation column.*

The sol gel method was used to synthesise yttrium aluminum silicate sol and freshly prepared heated sol were loaded into a syringe and pumped through a 0.4 mm diameter nozzle into paraffin oil. The droplets fell into a double-walled gelation column (1 cm internal diameter, 90 cm length) filled with paraffin oil thermostated at 80ºC in the top of the column and coolanted at -15°C in the end of the column.

Key word*: yttrium aluminum, yttrium silicate, yttrium nanoparticle*

Introduction

Numerous processes have been reported for the synthesis of spherical particles from metal alkoxides. Hydrolysis of tetraethyl orthosilicate (TEOS) is a versatile technique to synthesize as Silica microspheres. An acidic medium has been found to synthesize these particles. Thus, Karmakar et al. prepared silica microspheres by hydrolysing TEOS with acetic acid and Izutsu et al. with tartaric acid (weak organic acids) whereas Kawaguchi and Ono demonstrated the same using nitric acid (strong inorganic acid).
The methods used for synthesizing garnets are sufficiently diverse; however, each of these methods has its advantages and essential disadvantages. For example, the solid- phase synthesis of yttrium aluminum garnets from yttrium and aluminum oxides requires high temperatures (>1600°C); therefore, it is difficult to control the purity and homogeneity of yttrium aluminum garnet powders synthesized under these conditions whereas sol gels methods help to solve these problems.

Glass microspheres of 17Y2O3–19Al2O3–64SiO2 (mol %) with sizes ranging between 20–40 μm have been used in cancer brachytherapy treatment. The function of Al_2O_3 is to disperse the rare earth ions uniformly in the glass matrices and also in preventing the cluster formation trends in the glass networks. The isotope 89Y is transmuted to 90Y by neutron activation resulting in a β_emitter with half-life of 64.1 h. Other elements that are part of the glass structure, such as 27Al, and 30Si, are also activated becoming β emitters; however, the half-life is only 2.25 min and 2.62 h, respectively. These microspheres are chemically resistant to body fluids and are non-cytotoxic.
Glass particles with irregular shapes were transformed into microspheres by using two different processes. In the first process, glass microspheres with particle size distribution in the range of 20–150 μm were obtained by re-melting irregular particles in a hot flame. A torch to burn a mixture of oxygen and petrol

liquefied gas was used for this purpose. The microspheres were collected inside a metal cylinder. Although this process has been previously reported and it is known as 'spheroidization by flame', for each type of glass the experimental parameters had to be adjusted to reach the best results. The second process consists of introducing glass particles with irregular shapes on the top of a vertical tubular furnace, and allowing them to fall down inside the furnace. This process is now defined as 'spheroidization by gravitational fall in a tubular furnace.

Bioinert glass microspheres, with an yttria aluminosilicate composition containing Y89, have been developed by Day and Day to deliver localized doses of radiation for the treatment of liver cancer. Prior to use, the microspheres are bombarded by neutrons to create Y90, a radioactive isotope which undergoes beta-decay, with a short half-life (64 h), and with a short range (2–3 mm) in the liver.

Experimental

Tetra-ethoxy-silane (TEOS) (purity_99%) was supplied by Aldrich; glacial acetic acid was supplied by (Merck). Y_2O_3 (purity 99.9%) was supplied by Aldrich. Al $(NO_3)_3.9H_2O$ (purity 99%) was supplied by guangdon guanghua chemical factory Co. Scanning of electron microscopy was performed on a LEO 1430VP.

Synthesis of the yttrium silicate sol

$YCl_3.6H_2O$ obtained by the reaction of yttrium oxide and hydrochloric acid, SiO_2 colloids were produced by adding TEOS to a mixture of glacial acetic acid and water under stirring at room temperature. The molar composition of TEOS: CH_3COOH: H_2O was 1: 2: 2, which is described as the optimum composition for obtaining Yttrium silicate sol. A typical reaction uses 1mL of acetic acid 1mL of H_2O, 135 mg of $YCl_3.6H_2O$ and 0.5mL of TEOS. The Y ions were incorporated in the SiO_2 through replacing the water by an aqueous solution of $YCl_3.6\ H_2O$, respectively. After stirring the mixture for 30 minutes the sol was heated to 100°C.

Preparation of spherical particle

Freshly prepared heated sol was loaded into a syringe and pumped through a 0.4 mm diameter nozzle into paraffin oil. The droplets fell into a double-walled gelation column (1 cm internal diameter, 90 cm length) filled with paraffin oil thermostated at 80ºC at the top of the column and coolanted at -15°C in the end of the column. The sol droplets were therefore converted into solid gel spheres in a few seconds. The oil temperature is an important parameter. If it was too high, the kernels did not have time to gel and get aggregated in the bottom of the column.

Microspheres washing, drying and thermal treatment

After natural heating of the paraffin oil to room temperature, microspheres were separated from the paraffin oil and rinsed with carbon tetrachloride in a beaker to remove traces of oil. The kernels were then let to age in 200mL of a 2 mol/L solution of ammonium hydroxide during 30 min, prior to washing in the same solution by agitation during 30 min. The solution was then withdrawn, acidified with nitric acid and then the kernels were washed again with 200mL of fresh ammonium hydroxide solution, using the same method. This process was repeated at least three times. The efficiency of these washing

steps was identified by measuring the conductivity of the washing solutions used.
The kernels were then rinsed with distilled water with the same procedure, until the pH of the washing solution was neutral. The kernels were finally allowed to dry at room temperature overnight and washed at least twice with pure ethanol.

Results and discussion

Glass particles with irregular shapes (see Figure 1) were transformed into microspheres by using two different processes in solid phase: A) re-melting in a metal cylinder B) spheroidization by gravitational fall in vertical tubular furnace. However, in doing so the spheroidization of particles is performed in liquid phase into paraffin column. Fig.2 shows the scanning of electron micrograph of microspheres after using paraffin oil galation column.

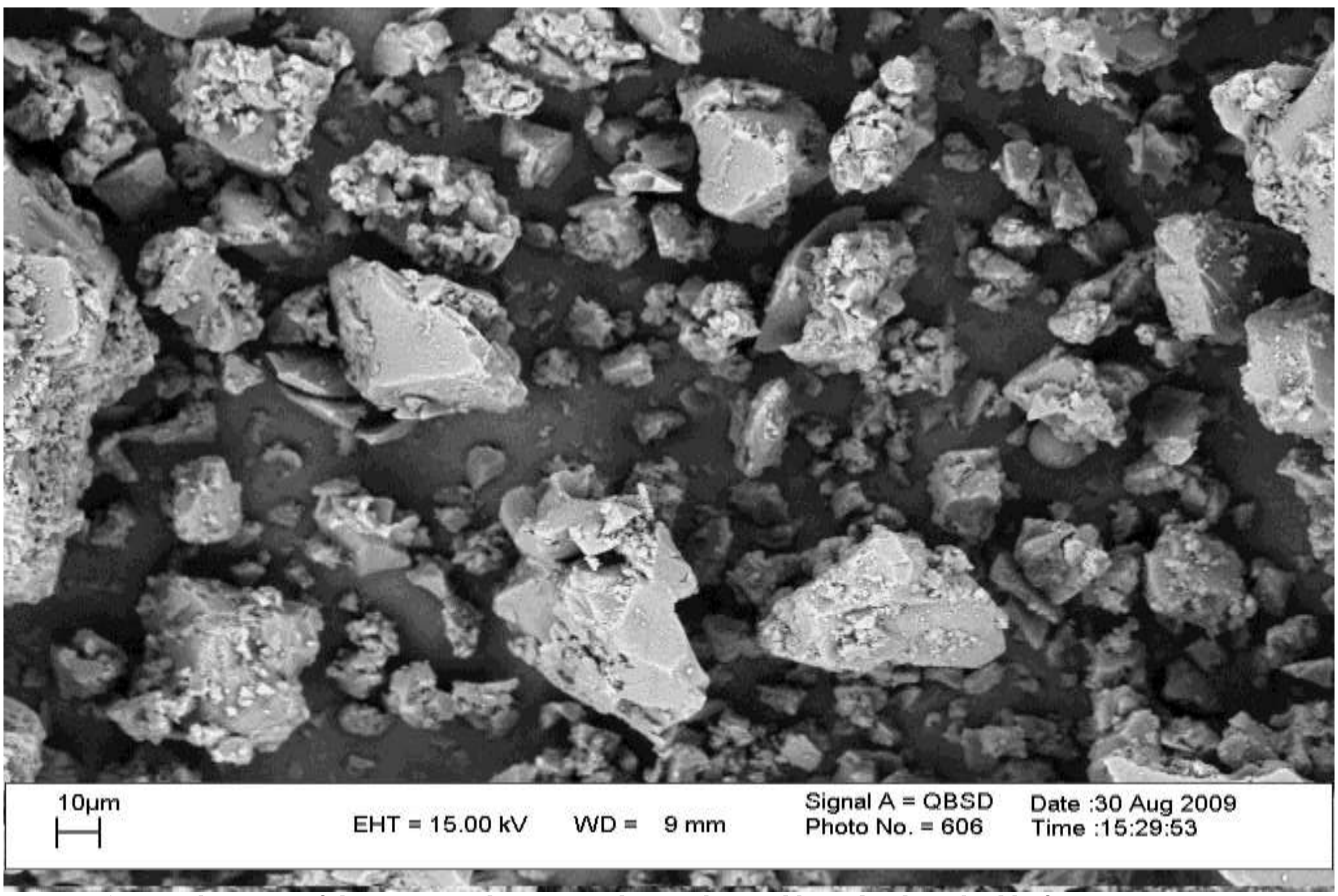

Fig. 1. The SEM of the Yttrium aluminum silicate obtained by sol gel method before spheroidization process

In this experiment, production of microspheres is dependent on the length of the column and partic size is dependent on the kind of the oil, the oil temperature and nozzle diameter.

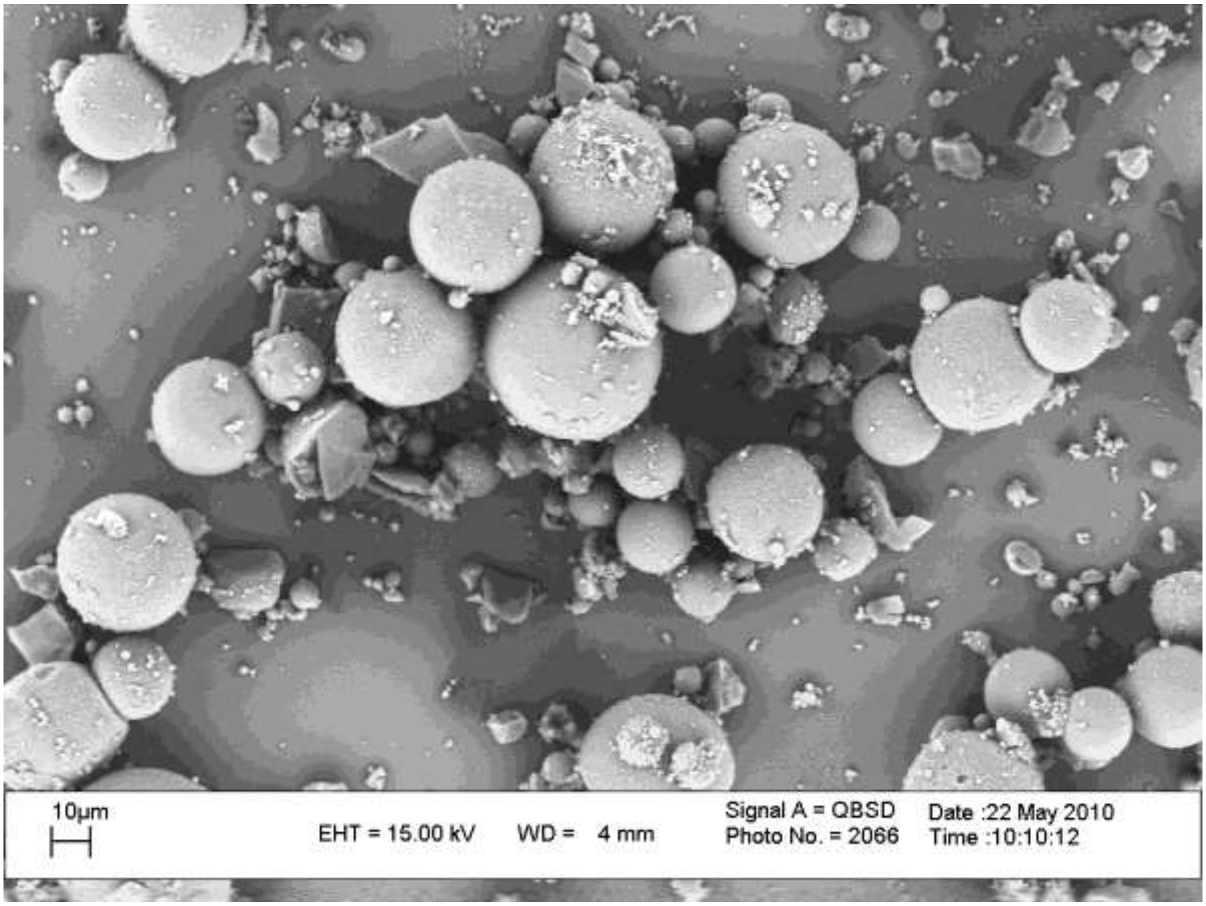

Fig. 2. The SEM of the Yttrium aluminum silicate microspheres by using of paraffin oil galation column

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QATILAŞDIRILMIŞ PARAFİN ƏSASLI NEFT SÜTUNU VASİTƏSİLƏ İTTRİUM ALÜMİNİUM SİLİKATIN KÜRƏŞƏKİLLİ MİKROZƏRRƏCİKLƏRİNİN SİNTEZİ

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Xərçəng xəstəliyinin braxiterapiya üsulu ilə müalicəsində ittrium alüminium silikatın radioaktiv şüşə mikrokürələrindən istifadə olunur. Mürəkkəb formaya malik şüşə zərrəciklər müxtəlif üsullarla əldə olunur və bərk fazada iki müxtəlif proses vasitəsilə kürəşəkilli formaya (mikrokürələr) salınır: A) metal silindrdə təkrar əritmək; B) şaquli boruşəkilli sobada cazibə qüvvəsi nəticəsində enmə üsulu ilə kürə şəklinə salmaq. Lakin biz parafin əsaslı neft sütunundan istifadə etməklə də kürəşəkilli mikrozərrəciklər əldə edə bilərik. İttrium alüminium silikat məhlulunu sintez etmək üçün zol-gel üsulundan istifadə olunmuşdur. Təzəcə hazırlanmış və isidilmiş məhlul diametri 0.4 mm olan şprislə parafin əsaslı neft sütununun içinə yeridilmişdir. Neft sütununun yuxarı hissəsində temperatur 80ºC-yə qızdırılır, aşağı hissəsi isə -15°C-yə qədər soyudulur.

СИНТЕЗ ШАРООБРАЗНЫХ МИКРОЧАСТИЦ ИТТРИЙАЛЮМИНИЙСИЛИКАТА С ПОМОЩЬЮ КОЛОННЫ, ЗАПОЛНЕННОЙ ПАРАФИНОВОЙ НЕФТЬЮ

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При лечении болезни рака методом брахитерапии используются радиоактивные стеклянные микрочастицы иттрийалюминийсиликата. Эти стеклянные микрочастицы имеют сложную форму и получаются разными способами, особенно в твердой фазе в шарообразной форме двумя методами: А) повторным плавлением в металлическом цилиндре и Б) путем падения под влиянием силы тяжести в вертикальной трубчатой печи. В данной работе получены сферические микрочастицы путем использования колонны, заполненной парафинистой нефтью. Для получения иттрийалюминийсиликатного раствора использован способ золь-геля. Свежеприготовленный нагретый раствор заполняется в шприц диаметром 0.4мм и вводится в колонну с двойной стенкой, заполненной парафинистой нефтью. В верхней части колонны температура нагревается до 80^{0}С, а в нижней части охлаждается до -15^{0}С.