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論　文

Electrostatic Assembly of Particle Chains from Fine Particles Suspended in Gas Phase

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Fine Al_2O_3 particles and LSM (Sr doped $LaMnO_3$) particles were agglomerated in the form of particle chains, controlled by electrostatic forces. The result shows that particle chains can be formed from both dielectric particles and conductive particles suspended in gas phase. The deposition of the particle chains onto substrate formed tangled networks, a structure peculiar to electrostatic dry deposition. After sintering under proper conditions, the particles were connected together without using any binder and the particle chains became fiber-like. The method presented in this paper will provide a novel and promising option for fabricating functional fiber or porous ceramics.

1. Introduction

Electrostatic coagulation of aerosols is a growing process of aerosol particles in contact with each other under the action of electric field. Frequently, the aggregates formed from particle coagulation have very unpredictable shapes, some more chainlike, some more clustered. In some cases it is desirable to control fine particles forming chainlike aggregates. H. Yamamoto and S. Masuda reported a new method for forming ceramic membrane.[1,2] Ultrafine particles synthesized by thermally activated chemical vapor deposition (CVD) were controlled to form particle chains by electrostatic forces, and a very specific ceramic membrane with a structure of three-dimensional network was obtained. B. Su and K.L.Choy[3] reported a novel method, termed Electrostatic Assisted Aerosol-gel Deposition (EAAD), to produce crack-free porous silica film with a unique reticular structure. One more example, K.D. Hermanson *et al.*[4] reported that a new class of microwires that can be assembled by dielectrophoresis from suspensions of metallic nanoparticles.

In our former work,[5] we reported Al_2O_3 fine particles could be controlled to form particle chain in electrostatic field. In the present work, we experimentally study the electrostatic formation of particle chains from fine particles suspended in gas phase. The aim is to obtain a three-dimensional layer on a substrate and conductive fiber. Fine particles are prepared form dry powder dispersion, which is different from the above methods. In this work, we intend to develop a new method for fabricating functional fiber and porous ceramics. The basic processes of this study are as follows. The first step is to supply a well-dispersed powder to the gap between electrodes with an appropriate particle concentration. The dispersion achieved by accelerating particles within a compressed N_2 stream, together with particle and wall collision. The second step is to deposit fine particles onto a substrate, directly after dispersion, under the action of an electric field without corona generation. In the field, particles are polarized, not charged. In this step, we expect that particle chains can be formed on the substrate. The third step is sintering the formed particle chain. The field-induced chain aggregates are expected to become fiber, and fixed.

2. Experimental Set-up

Two types of fine powders, Al_2O_3 and Sr doped $LaMnO_3$ (LSM), is used in this experiment. The Al_2O_3 powder has average particle size 0.5 μm according to catalog data. LSM powder is conductive, having average particle size 0.5 μm and conductivity larger than 100 Scm^{-1}.

Figure 1 shows the schematic diagram of the experimental setup, which includes two parts: powder jet feeder and coating chamber. The powder jet feeder consists of a powder container (1) and a nozzle (4). Fine powders, normally 4 g, were dried sufficiently by an electric oven before placed in the speed controlled vibratory container, which is 10 mm in diameter and 10 cm in length. The particles pass through the nozzle (0.5 mm in diameter) accompanying with a jet of carrier gas (N_2), and then impact onto the backside of the substrate holder (6). This approach prevents powder build-up, clogging and the inconsistent feed rate normally experienced in systems that use tubing to transport powder. Dispersed particles pass through the wall of a stainless steel sieve cylinder (5), 2 cm in diameter, 5 cm in length, 38 μm sieve opening (400 mesh), which also supports the substrate holder. Then, the particles enter into the coating chamber in which DC high voltage is imposed. The electrode configuration is point-plane. The point electrode (8) is a 1 mm wire with hemispherical tip and the plane electrode (6), which also acts as the substrate holder, is 5 cm in diameter. The gap between the electrodes is 4.5 cm. Under this configuration, the onset potential of

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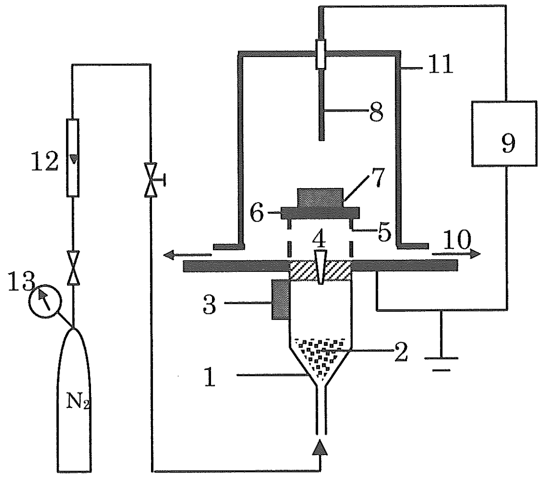


Fig. 1 Experimental Set-up.

1.Powder container 2.Powder 3.Vibrator 4.Nozzle (0.5 mm inner diameter) 5.Sieve cylinder (Stainless steel, 400 mesh in pore size) 6.Substrate holder (Copper) 7.Substrate 8.High-voltage electrode 9.DC high voltage power supply 10.Gas outlet 11. Plastic chamber 12.Flow meter 13.Gas regulator

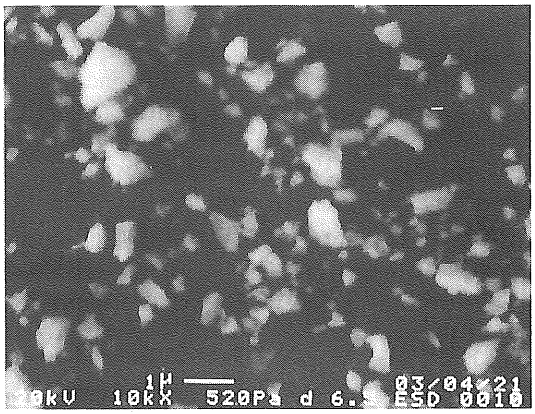


Fig. 2 Al_2O_3 particles after dispersion.

corona discharge is about 9 kV. The electrode assembly is sealed in a transparent plastic chamber (11), which is 10×8×14 cm in volume. Dispersion of the fine powders into ambient gas is an important operation in this experiment. Homogeneously dispersed system of primary particles, not of aggregates, suspended in the gas is wanted. Figure 2 shows the Al_2O_3 particles (0.5 μm) after dispersion, which was obtained by the natural sedimentation of particles on a copper substrate.

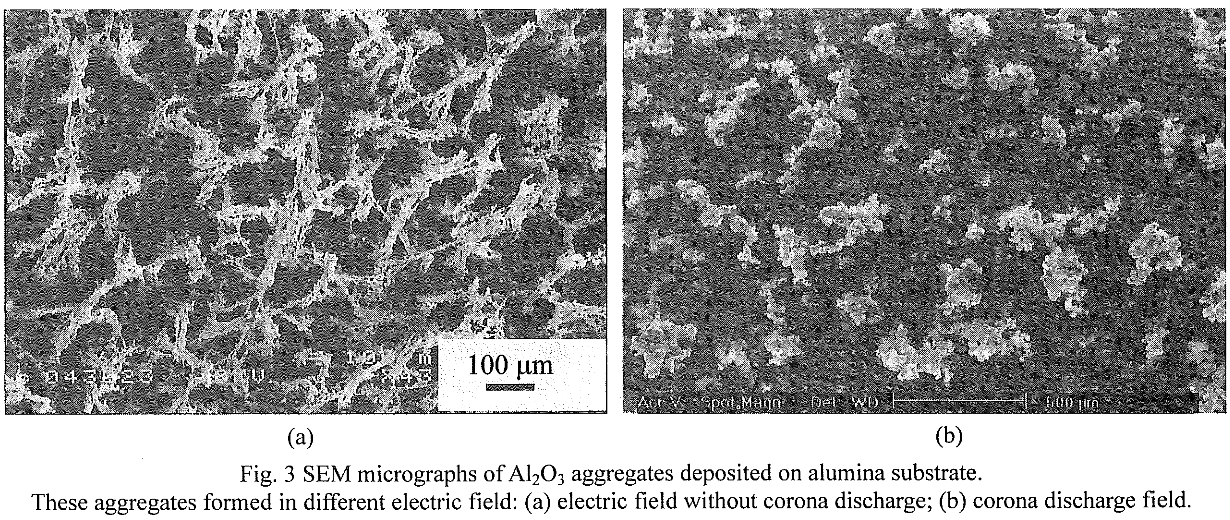
The Al_2O_3 particles are deposited onto an alumina substrate (7) (10×10×3 mm in size), and the LSM particles are deposited onto YSZ (Yttrium Stabilized Zirconia) substrate (Φ10 mm, 1 mm in thickness).

Scanning Electron Microscope (SEM) is used to observe the surface structure of the particle layer deposited by electrostatic force. The chain aggregates formed on the substrates were sintered in an electric furnace.

3. Results and discussions

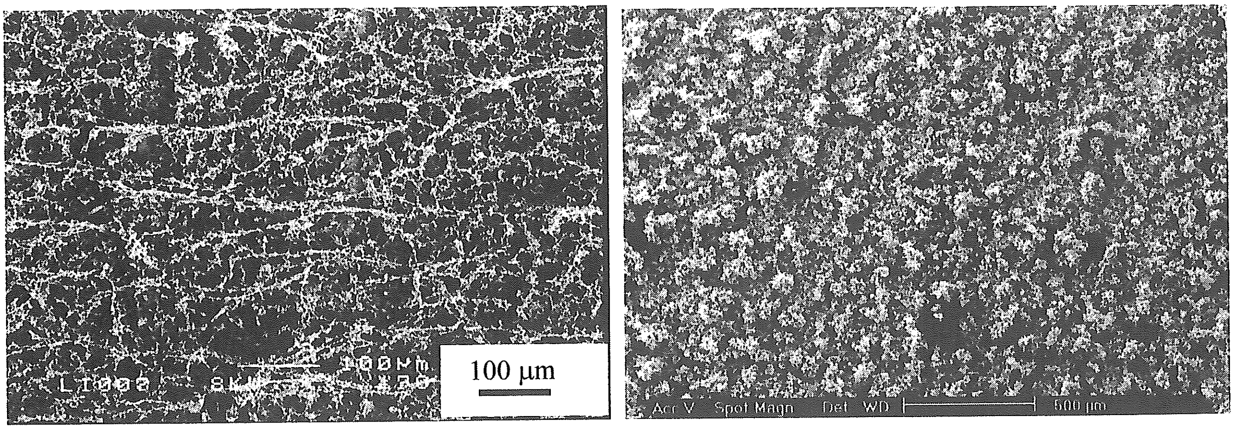
3.1 Influence of electric field intensity to chain formation

Figure 3 shows the Al_2O_3 particles deposited on the alumina substrate under different electric field intensity after sintering. The particle concentration in the coating chamber was around 20 g/m^3 while the N_2 gas pressure was kept at 1 atm. As can be seen, the shape of particle aggregate is greatly influenced by the electric field intensity. In the electric field without corona discharge (8 kV, positive, DC potential imposed on the point electrode), the Al_2O_3 particles agglomerated to chain-like aggregates, and the deposition of the chain aggregates onto the substrate formed a layer with the structure of tangled network, as shown in Fig.3 (a). However, in corona discharge field (15 kV DC potential imposed on the point electrode, discharge current was around 10 μA), the Al_2O_3 particles coagulated and formed the clustered aggregates, as shown in Fig. 3 (b). The same phenomena appeared in testing LSM fine particles, which is conductive, as shown in Figs. 4 (a) and (b).



(a) (b)

Fig. 3 SEM micrographs of Al_2O_3 aggregates deposited on alumina substrate.
These aggregates formed in different electric field: (a) electric field without corona discharge; (b) corona discharge field.



(a) (b)

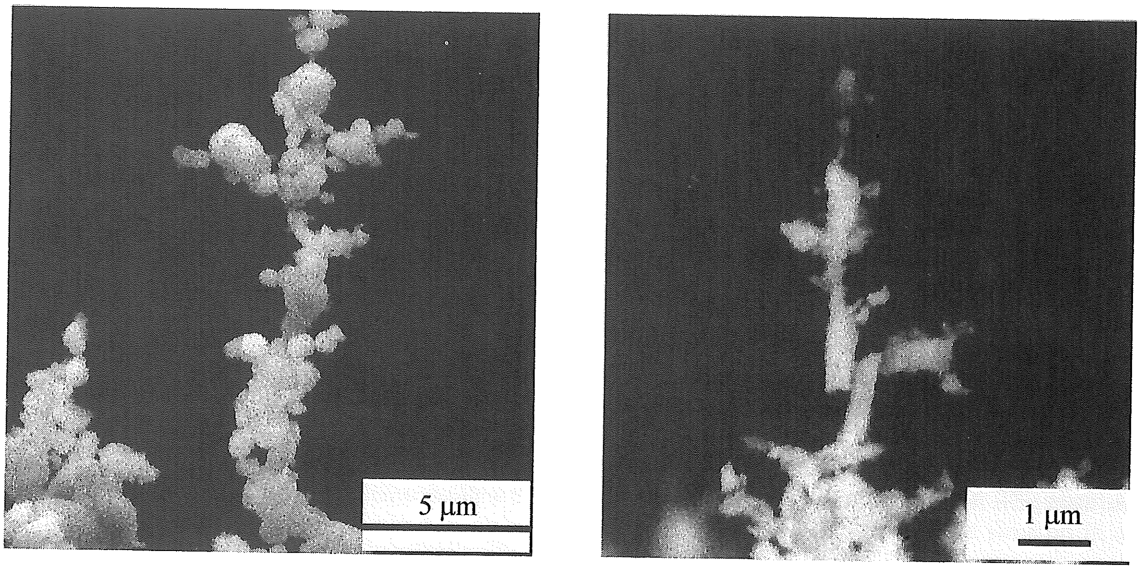
Fig. 4 SEM micrographs of LSM aggregates deposited on YSZ substrate.
These aggregates formed in different electric field: (a) electric field without corona discharge; (b) corona discharge field.

The above results show that the shape of aggregates can be controlled with the discharge current. In corona discharge field, the charged particles and ambient ions serve as nucleation sites for particle agglomeration. Charged particles with opposite sign attract each other, and repel each other owing to the same charge sign. Consequently, the shape of particle aggregates is very difficult to be controlled. Although dielectric particles can also agglomerate in the form of chain aggregates due to dipole interactions, the ionic flow will continually charge the aggregates, and the chain structure is difficult to remain. As shown in Fig. 3(b), the aggregate shape is dominantly clustered. For the conductive particles, the particles on the substrate are charged by induction to become negative polarity. The positive charged particles are attracted to the negative charged particles, and form clustered aggregates on the substrate, as shown in Fig. 4 (b). In an electric field without corona generation, polarization effects may induce formation of particle chains. The electric field polarizes a particle, and at close spacing, the polarized particles interact each other. Depending on the relative alignment of the particles, the mutual forces of interaction can be attractive or repulsive. In general, similar particles attract each other when aligned parallel to an applied field and repel each other when in perpendicular alignment.[6)] Therefore, dipole-dipole interactions induce particle chain formation.

In above discussion, only the electrostatic forces interacting between particles were considered. In fact the adhesive forces between particles arising from Van der Waals force and surface tension can also force two particles into contact and the particles stick to form an aggregate. Van der Waals forces are due to instantaneous polarization of atoms and molecules.[7)] Surface tension effects arise from interactions of layers of adsorbed moisture on the surfaces. In electric field, electrostatic forces are the predominant forces because the results presented above have proved that the aggregate shape can be controlled with electrostatic forces. But particle coagulation depending on Van der Waals force or surface tension will be in a random way. In the situation without external field, however, the primary forces of particle adhesion are Van der Waals forces, especially for the sub-micron particles.[8, 9)] Figure 5 shows the chain aggregates of the two kinds of particles before sintering. As we observed, the structure of particle chains remains without the action of external field. In other words, the formed particle chains are self-supported.

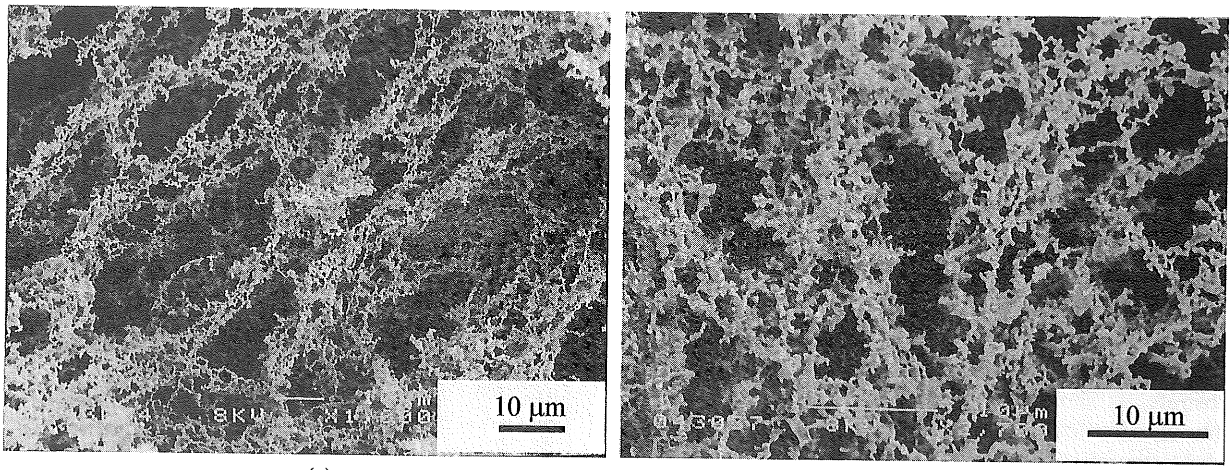
3.2 Preliminary results of fabricating porous ceramic and functional fibers

Porous oxide ceramics are important for a variety of applications such as thermal insulation, filtration, biomedical, and catalyst substrates. These ceramics may be classified into four basic structures: tangled fiber networks, closed cell structures, open cell structures, and membranes.[10)] As described in the former section, fine particles suspended in gas phase can be controlled to form chain aggregates by electrostatic forces, and the deposition of the chains aggregates onto substrate formed a tangled network layer. This will provide a new way for fabricating porous ceramics. As in the other methods, particle concentration is an important factor affecting the structure of the particle layer. Figure 6 shows the SEM micrographs of Al_2O_3 particle layers deposited on alumina substrate using different particle concentration. High particle concentration induces high coagulation rate and high deposition rate. Consequently, the structure of Fig. 6(b) is better than Fig. 6 (a). In practice, the particle concentration should be chosen according to the desired pore structure and thickness. But it should be larger than 50 mg/m^3, the lowest concentration for forming particle chain reported by R. Flossmann and A. Schütz.[11)] In future we will study the relationship between particle concentration and pore size, thickness of the porous layer in detail.



(a)　　(b)

Fig. 5　SEM micrographs of chain aggregates before sintering.
(a) Al_2O_3 particle chain; (b) LSM particle chain. The chain aggregates are self-supported due to the Van der Waals forces.



(a)　　(b)

Fig. 6　SEM micrographs of the particle layers with reticular structures.
The layers were deposited at different particle concentration (a) 50 g/m^3 (b) 100 g/m^3.

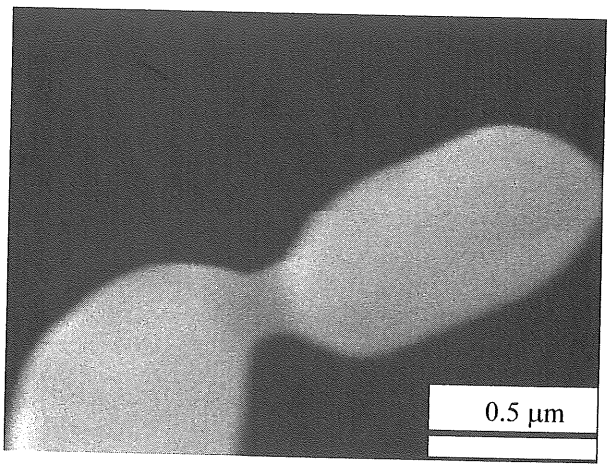


Fig. 7　Connection between two LSM particles after sintered at 1200℃, 2 h.

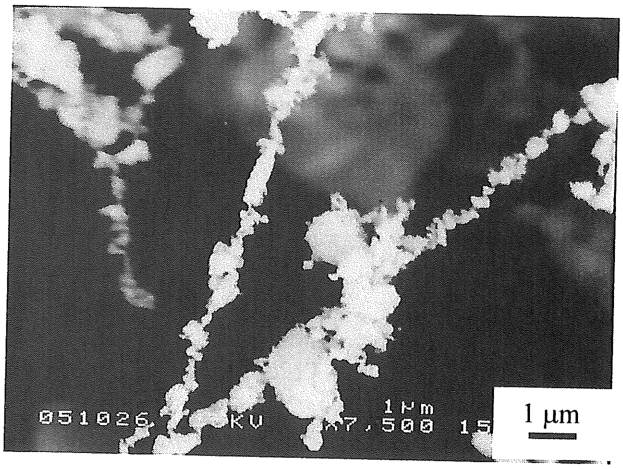


Fig. 8　LSM fiber formed from sintering LSM chain aggregate

In this experiment, particles in a particle chain were sintered together in atmosphere without using sintering aid. In the sintering process, careful control of the parameters such as temperature, time, *etc.* is essential. These parameters were determined empirically by observing the resultant structure. For Al_2O_3 particles, the sintering conditions were: heating rate 10℃/min, sintering temperature 1300℃, sintering time 2 h. For LSM particles, the sintering temperature was 1200℃, and the heating rate and sintering time were the same as those of Al_2O_3. Figure 7 shows the connection of two LSM particles after sintered. As we observed, the chosen sintering conditions just sintered the particles to the initial sintering stage of neck formation and growth. Particle chains became fiber-like.

Figure 8 shows LSM fibers formed from sintering LSM chain aggregates. As reported by K.D. Hermanson *et al.*,[4] conductive micro-wires have great meaning for miniaturization of photonic and electrical circuits. The simple assembly process presented here will provide a promising option for fabricating functional micro-wires.

4. Conclusions

This paper presented a method for assembling particle chains from fine Al_2O_3 particles and LSM (Sr doped $LaMnO_3$) particles suspended in gas phase. After sintering, the particle chains became fiber-like, and particle layers with the structure of tangled networks were obtained. Compared with other techniques, the key advantages of the method are:

(1) Fine particles are prepared from dry powder dispersion.
(2) The equipment is simple and low cost.
(3) Substrate may be simple or complex, ceramic or metal.
(4) It can be used to fabricate porous ceramics or functional micro-wire.

Additional work will be performed on the following aspects:

(1) The effectiveness of the method for fine particles of other materials.
(2) Fabricating porous ceramic filter for micro-filtration.
(3) Fabricating metallic micro-wire.

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