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Application of high magnetic fields in advanced materials processing

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Abstract Recently, steady magnetic fields available from cryogen-free superconducting magnets open up new ways to process materials. In this paper, the main results obtained by using a high magnetic field to process several advanced materials are reviewed. These processed objects primarily include superconducting, magnetic, metallic and nanometer-scaled materials. It has been found that a high magnetic field can effectively align grains when fabricating the magnetic and non-magnetic materials and make inclusions migrate in a molten metal. The mechanism is discussed from the theoretical viewpoint of magnetization energy.

Keywords: stead magnetic field, advanced materials, alignment, magnetization force.

1 Introduction

The magnetic field, like temperature or pressure, is one of the important thermodynamic parameters of the inner energies of the materials. The magnetic field can enhance a Lorentz force even in a weak electric current and a magnetization force, which is tangible not only in magnetic materials but also in nonmagnetic ones such as paramagnetic and diamagnetic materials. With the development of cryogenic and superconducting technologies, nowadays it is quite easy to generate high magnetic fields above 10 T by using cryogen-free superconducting magnets, which can be used for a long term without filling with liquid helium[1,2]. The cryogen-free superconducting magnet has opened up a new field in advanced materials (such as magnetic materials, oxide superconductors and organics) processing, and also in the research of nanometer-scaled materials, spintronics, polymer, bio-chemistry and so on.

Now, high magnetic field is a promising means in materials processing[3], and it is expected that new phenomena will be discovered by high magnetic fields under extreme conditions of high pressure and ultra-low temperature. In this paper, recent progress in magnetic field processing of new advanced materials is introduced, and the prospect is discussed.

2 Principle of magnetic field functions during materials processing

Processing in a magnetic field is a novel means to produce oriented materials. Due to the persistence of magnetocrystalline anisotropy at high temperature, the crystal orientation in materials (including paramagnetic and even diamagnetic materials) can be controlled by applying a high magnetic field[4,5]. Besides, it is found that diamagnetic materials can be levitated in gradient magnetic fields, when the magnetic force produced by a gradient magnetic field balances the gravity[6]. The availability of high magnetic field technologies gives nowadays development perspectives to magnetic and contactless processing for fundamental research and for some applications.

When a material with anisotropy susceptibility is put in high magnetic fields, the field will exert anisotropy energy on the grains in the material. The interaction energy of a material with a magnetic field is

$$E = -\frac{1}{2\mu_0}\chi V B^2, \qquad (1)$$

where V is the volume of a grain, B is the magnetic field applied on the material and χ is usually anisotropic on the scale of the grain. This means that the energy varies depending on the direction of the grain with respect to the applied magnetic field. Defining $\Delta\chi$ as the difference between the maximum and minimum susceptibilities, the energy difference is

$$\Delta E = -\frac{1}{2\mu_0}\Delta\chi V B^2. \qquad (2)$$

And then the grain tends to orient itself in the direction of the lowest energy. In other words, when a material is placed in homogeneous magnetic fields, the induced magnetic moment has a torque, which contributes to the orientation of the material. According to eq. (2), if the volume of the grain is large enough and the magnetic field is strong, the anisotropy energy will be greater than the thermal disordering energy and the grain will be oriented in the magnetic field. In this case, the magnetic alignment effect takes place. In terms of paramagnetic materials ($\chi > 0$), the magnetic energy is minimized when the axis of maximum susceptibility is parallel to the magnetic field, while the orientation, along which the absolute value of χ is smaller, will be preferred in the case of the diamagnetic substance ($\chi < 0$). Therefore, the magnetic alignment effect is associated with the anisotropic susceptibility of the material as well as the field strength. The processes related to the crystal orientation by use of a high magnetic field are mainly as electro-deposition, vapor-deposition, solidification, ceramic sintering process and slip-casting. Now it has been recognized that the application of a high magnetic field is a quite promising method in electromagnetic processing of new materials.

On the other hand, the force F acting on a material in a field is given by the gradient of E and its z component parallel to the field direction (vertical in this case and positive for upward) is expressed as

$$F = -\text{grad}E = \frac{\chi}{\mu_0} B \frac{\mathrm{d}B}{\mathrm{d}Z}. \tag{3}$$

One can note that, if a material is placed in an inhomogeneous or gradient field, the direction of the force depends on the sign of χ but if the sign of χ is negative, which is realized in diamagnetic materials, the force is repulsive from the center of the magnet and it is possible to make the force act upwards in a vertical field. When this force is strong enough to exceed the force due to the gravity, the specimen levitates. The advantage of material synthesis in levitation condition is, in principle, the contactless processing.

3 New materials processed by means of a magnetic field

3.1 Superconducting materials

The critical current density (J_c) of high-T_c superconductors (HTS) is enhanced by aligning the grains to produce a c-axis (00l) texture. One effective method of aligning HTS grains is to apply a large magnetic field during HTS phase formation or sintering. Magnetic field induced texture in HTS results from the anisotropy in the paramagnetic susceptibility associated with the Cu-O conducting planes. In 1991, Rango *et al.*[5] demonstrated that it is possible to prepare textured YBCO ceramic bulks by solidification in a 5 T magnetic field. Since then, Noudem *et al.*[7] using an 8 T magnetic field during processing of Bi-2223 pellets, also achieved a high degree of texture. Ma *et al.*[8] obtained a high degree of texture in Bi-2223/Ag tapes melt-processed in a magnetic field. Liu *et al.*[9] reported an increase in the degree of texture and transport critical current of Bi-2212 thick tapes melt-processed under the influence of an elevated magnetic field, which was further confirmed by a group in Sendai[10]. To illustrate the effect of the magnetic field, Fig. 1 shows two Bi-2212 tapes that were melt-processed in the absence (Fig. 1(a)) and the presence (Fig. 1(b)) of a 10 T magnetic field. It is clear that the increase in grain alignment is produced by the thermomagnetic process.

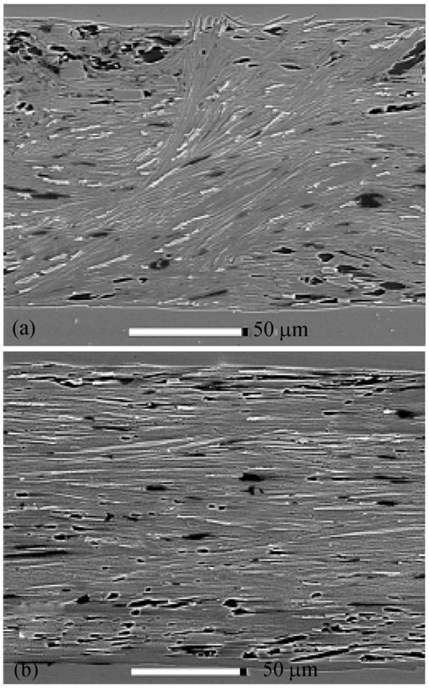

Fig. 1. SEM cross-sectional images from Bi-2212 tapes processed in the absence (a) and in the presence of a 10 T magnetic field (b).

On the other hand, the high irreversibility field of second-generation YBCO superconductor at liquid nitrogen temperature makes it a strongest candidate for future applications. Awaji *et al.*[11] synthesized YBCO bulk samples by the melt textured seeding process in a field of 10 T, and found that the critical current density increases with the applied magnetic field. Ma *et al.*[12−14] carried out systematical research work on the crystal growth of YBCO films by MOCVD method under high magnetic fields, and emphatically studied the influence of magnetic field on the microstructure and superconducting properties of YBCO films. It is found that the magnetic field influences the grain size during deposition. For instance, the square-shape grains with 8−10 μm in size are observed in the 0 T sample. With increasing magnetic field at the deposition, however, the grain size decreases and the microstructure changes to the irregular-shape grains below 1 μm in size. Most importantly, the grain connectivity was improved with increasing magnetic field[13]. Hence, with increasing magnetic field applied during deposition, the critical current density increased as shown in Fig. 2.

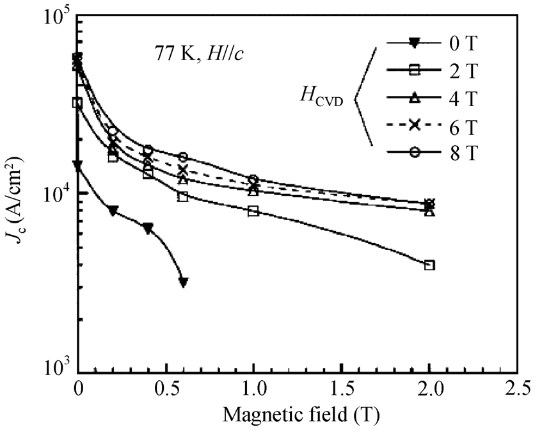

Fig. 2. Magnetic field dependence of J_c of YBCO films deposited in different magnetic field at 77 K.

As we know, new superconductor MgB_2 was discovered in 2001. Since it possesses a strong anisotropy structure, superconducting properties of MgB_2 may be improved when processed under a high magnetic field. Based on this idea, we have fabricated MgB_2 bulks and Fe-sheathed tapes under a high magnetic field for the first time[15]. It is found that application of a large magnetic field during processing resulted in quite uniform microstructure and better connectivity between the MgB_2 grains. As a result, the J_c of these samples has shown a much higher value than that of the MgB_2 samples in the absence of magnetic field.

3.2 *Magnetic materials*

It is very important to control the microstructure of magnetic materials because textured or anisotropic magnetic materials usually show better performance in practical applications. So far magnetic field processing is a well-proven technique to impose the desired texture and magnetic properties of magnetic materials. For permanent magnetic materials, the loose powders of single crystal grains can rotate so as to align their easy axis of magnetization along with the magnetic field direction. Thus the oriented permanent magnets with higher remanence and energy product can be obtained by solidification from the molten state in a high magnetic field. In the case of ferromagnetic alloys, texturing in high magnetic fields can be obtained by two means: when solidification directly occurs in the ferromagnetic state after a sufficient supercooling, and when annealing of quenched samples is performed below the Curie temperature.

It was reported that a high degree of orientation is obtained with samarium-cobalt compounds solidified in a static magnetic field[16]. Anisotropic bulk magnets with a coercive field up to 2250 kA/m and energy product above 160 kJ/m^3 are obtained. This process provides an alternative to the currently used industrial technology which is based on powder metallurgy. Recently, Cui *et al.*[17] found that there is noticeable improvement in the magnetic properties of the melt-spun $Nd_{2.4}Pr_{5.6}DyFe_{85}B_6$ alloy and $Nd_2Fe_{14}B$/Co nanocomposite after a magnetic annealing. The magnetic annealing can result in both the magnetic-field-induced crystallographic texture and an enhanced exchange coupling. High-field annealing was performed in sintered Nd-Fe-B magnets in order to increase the coercivity by modifying the interface between the $Nd_2Fe_{14}B$ and Nd-rich phases. When the sample, containing 1.3 at% Dy and 0.32 at% Al, was annealed at 823 K under a magnetic field of 14 T, the coercivity reached 1.92 T, which is 37% higher than that for the control sample annealed at zero field[18]. The reason is that a preferential alignment may take place in the solidification process of the Nd-rich phase which makes better lattice matching in the interface with a main $Nd_2Fe_{14}B$ phase, resulting in a higher coercivity. On the other hand, for the soft magnetic materials, it was found that annealing the cold-rolled Fe-Si sheet[19] in a high magnetic field of 10 T parallel to the rolling direction enhanced the selectivity of the <001> axis alignment. High magnetic field annealing was also applied in FePd system by Tanaka *et al.*[20]. It was shown that a so-called mono variant $L1_0$ structure was obtained by high magnetic field annealing at 780 K under 10 T.

Stronger effects or new effects are expected if magnetic interactions occur in the ferromagnetic state. Gaucherand *et al.*[21] observed the texturation of ferromagnetic Co-B alloys at high temperature in a high magnetic field. As a result of the solidification in the ferromagnetic state of the first cobalt particles, the microstructure is strongly anisotropic. As seen in Fig. 3, particles are stacked as long needles along the applied field. On the contrary, no order was observed in the zero field solidified samples where such cobalt particles were randomly dispersed.

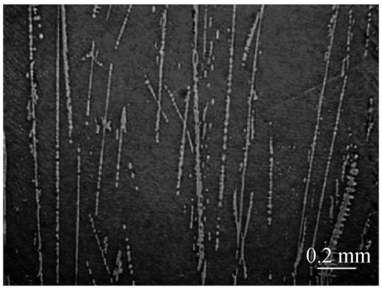

Fig. 3. Primary cobalt particles self organize as needle-like colonies in a 3T magnetic field when solidification of Co-B alloys starts in the ferromagnetic state. The magnetic field is vertical in the picture plane.

3.3 *Metallic materials*

Purifications of molten metal alloys or certain flow structures can be easily achieved through controlling the heat and mass transfer during metallic alloys solidification processes. This is due to different susceptibilities between the precipitates and the molten alloys and the influence of magnetic field on mass transfer. At present, the contactless handing of electrically conducting liquids by use of magnetic fields makes this technology attractive in many industrial practices.

The principle of this method is based on the magnetization force. Takagi *et al.*[22] reported that the magnetization force induced by a high magnetic field could move non-magnetic substances, for instance, when Al–18%Si alloy was melted and solidified in a magnetic field. The precipitated silicon particles, simulating inclusions, and the molten alloy are diamagnetic and paramagnetic, respectively. The macrostructure of the solidified alloys is shown in Fig. 4. Without a magnetic field, the silicon particles are located mainly at the bottom, as shown in Fig. 4(a). On the contrary, the applied magnetic field makes the magnetization force act on the upper direction of the specimen and the particles migrate to the top part, though some of them adhere to the wall surrounding the melt (Fig. 4(b)). Yasuda *et al.*[23] demonstrated that the static magnetic field reduced not only the rising velocity of the Cu-rich drops but also the coalescence rate of the liquid drops, resulting in a reduction in the macrosegregation during solidification of the Cu-Pb alloys. When directionally solidified Al-4.5% Cu alloy under a 10 T magnetic field, the aligned structure formed and the crystal <111> direction turned to the direction of the magnetic field, opposing to the dendrite growth in the crystal <100> direction[24]. As we know, a field usually brakes the flow of a liquid metal and this is observed in the liquid bulk during solidification. However, Lehmann *et al.*[25] found that AlCu and AgCu alloys were solidified directionally in the horizontal configuration under a transverse magnetic field; as a result, this force is opposite to the natural solutal buoyancy force. They argued that when a magnetic field is applied during a solidification process under a high temperature gradient, the effect of the thermoelectric Lorentz force cannot be neglected.

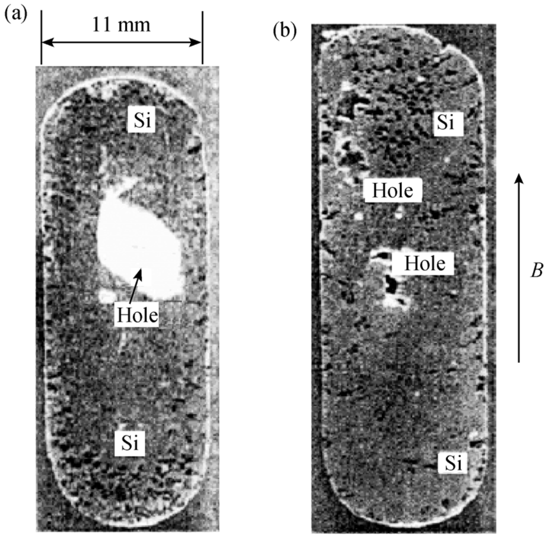

Fig. 4. Macrostructure of Al-18% Si alloy. (a) Without magnetic field; (b) with magnetization force.

As for the magnetic solidification of Bi-Mn alloy, due to strong magnetic moments, MnBi crystals grow preferentially and congregate along the *c*-axis of MnBi. The good alignment of MnBi crystal can be accomplished under relatively moderate fields. The magnetization shows pronounced anisotropy in magnetization in directions normal and parallel to the fabrication field, resulting from this alignment[26]. The crystal orientation effect was also observed in the rapidly solidified Bi-20 at% Mn alloys when further subjected to the semi-solid processing under a magnetic filed of 4 T[27]. It is evident that the fabrication field improves the crystal quality of not only MnBi compounds, but also the Bi matrix by suppressing the convection and the movement of matter by Lorentz force and magnetization force. In addition, when 1Cr18Ni9Ti stainless steel tube blanks were produced by electromagnetic centrifugal casting, the as-cast microstructure of the blanks was refined due to electromagnetic stirring. As a result, the plastic deformation ability of the tubes was greatly improved by electromagnetic centrifugal casting[28].

3.4 Nano-materials

The research interest is now shifting to smaller scales, that is, to nanostructures, which are expected to provide a revolution in material research. Nanostructures are structures with dimensions in the range of 1—50 nm and the physical dimensions largely determine the properties of these structures. It is this inherently heterogeneous structure on a nanometre scale that dictates many of their attractive mechanical, electrical, thermal and chemical properties. Macroscopic materials comprising a large number of nanostructures in crystalline alignment with one another would constitute highly anisotropic materials with wide potential applications.

Walters *et al*.[29] synthesized a suspension of purified single-walled carbon nanotubes (CNTs), and then put the suspension to a strong magnetic field of 25 T to align the CNTs, and produced excellent aligned membranes of CNTs as shown in Fig. 5. The alignment of CNTs in magnetic fields arises from the anisotropic magnetic susceptibility of nanotubes. The mechanism is that (n, n) variety of CNTs is paramagnetic in the direction of their long axis, and tends to align parallelly to the ambient magnetic field. The other varieties of CNTs are diamagnetic, but their diamagnetic susceptibilities are most negative in the direction perpendicular to the tube axis, causing them to also align parallelly to the direction of an ambient field. The orientation energies for both the paramagnetic and diamagnetic varieties of CNTs are comparable, and the calculated CNTs susceptibilities predict that at room temperature, fields of an order of 10 T will produce the orientation of individual CNTs. Similarly, Kimura *et al.*[30] succeeded in using a high magnetic field to align CNTs in a polyestermatrix and obtained electrically conductive and mechanically anisotropic composites. They dispersed CNTs in the monomer solution of unsaturated polyester, and then applied a constant magnetic field of 10 T to align the nanotubes. Polymerizing this CNT-monomer dispersion under the magnetic field freezes the alignment of CNTs in the polyester matrix. Choi *et al.*[31] also demonstrated that the thermal and electrical properties of single wall carbon nanotube polymer composites were significantly enhanced by magnetic alignment during processing.

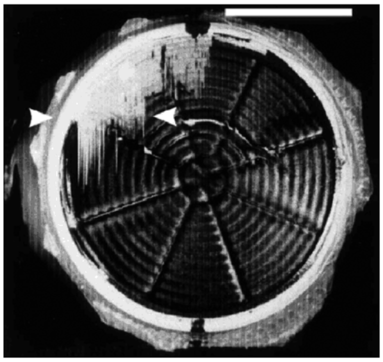

Fig. 5. In-plane-aligned membranes of CNTs, formed by filtration in a 25 T magnetic field (scale bar: 1 cm).

On the other hand, ferromagnetic nanowires (NWs) are scientifically important because they have unique magnetic properties originating from their one-dimensional structures and are also technologically important because they may be applied to a high-density perpendicular magnetic recording with their excellent magnetic properties. Lee *et al.*[32] prepared aligned ferromagnetic iron and cobalt nanocluster wires (NCWs) under a magnetic field with a diameter from 8 to 10 nm and a length up to a few millimeters. The NCWs grew through aggregation of metallic nanoclusters along lines of magnetic flux. Without a magnetic field, however, only metallic nanoclusters could be produced. Hangarter *et al.*[33] reported the fabrication of segmented nickel/gold/nickel and fabricated nickel/bismuth/nickel nanowires with controlled dimensions by template-directed electrodeposition. One hundred percent magnetic alignment of nanostructures to the imposed magnetic fields was achieved by applying a low external magnetic field. Niu *et al.*[34] prepared Co polycrystalline wires with an average length of 2 mm and a diameter of 13 μm by the self-assembly of Co nanocrystallites (15 nm on average) through inducing a 0.25 T external magnetic field. They found that although the nanocrystallites contained single domains, the orientation of each domain was spontaneously random when no external magnetic field was applied. However, when an external magnetic field was applied, the spherical particles tended to align along the magnetic line of force and favor the formation of linear chains. Therefore, the magnetic aligning process seems promising in fabricating large arrays of uniform wires of some materials and improving the magnetic properties of nanoscale magnetic materials.

3.5 Other materials

Finally, growing crystals of diamagnetic materials in a magnetic field is also interesting. As the susceptibility is very small in diamagnetic materials, the effects of a high-magnetic field on these materials are generally neglected. Recently, superconducting magnet technologies have been developed and used for such diamagnetic materials. For example, crystal growth of benzophenone in a field up to 8 T has been reported[35]. It was demonstrated that a magnetic field changes the morphology of protein crystals and increases the quality[36]. Another type of orientation effect occurs for biological materials. It has been found that there is good alignment in a field of 8 T for the polymerization of fibrin fibres in magnetic fields[37]. In addition, Sakka *et al*.[38] have demonstrated that the alignment of titania whiskers can be controlled by colloidal filtration of a well-dispersed suspension of the whiskers in a high magnetic field of 10 T when the direction of the magnetic field was perpendicular to the direction of the fluid. Whisker alignment seems to be an attractive way to improve the properties of whisker reinforced materials. More recently, Ma *et al*.[39] reported that the ferromagnetic transition temperature was significantly enhanced when $YNi_xMn_{1-x}O_3$ films were annealed in the presence of an 8 T magnetic field. Characterization study shows that the microstructure is affected, obtaining larger grains of uniform size. The improvement in the ordering temperature of all films is interpreted in terms of the grain growth caused by the magnetic-field driving force for boundary motion where the exchange coupling is high.

Thanks to the development of superconducting magnet technology, clearly, not only magnetic materials but also non-magnetic materials are orientatively ‘processed’ by the magnetic field. Surely, by improving electric, magnetic, thermal and mechanical properties of materials we can acquire new materials with the orientation effect of the magnetic field. It is expected that a breakthrough in magnetic field processing will surely bring up a rapid progress in the research of new functional materials.

4 Conclusions

The experimental studies on the materials processing using a high magnetic field, including superconducting, magnetic, metallic and nanostructural materials, have been reviewed and discussed from the theoretical viewpoint of magnetization energy. It has been found that a high magnetic field can make inclusions migrate in a melt and align grains in fabrication processes for the magnetic and non-magnetic materials.

Materials synthesis or processing in high magnetic fields is a new and useful method for creating new types of materials. Especially, with recent development of cryogen-free superconducting magnets, people are paying more and more attention to the ‘control of materials’ for materials science by means of a magnetic field, such as materials synthesis, chemical reactions or crystal growth in high magnetic fields.

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