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Low frequency damping behavior associated with sintering process in Al powder compact

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Abstract: The internal friction behavior of Al green power compact duxing the sintering process was studied as a function of temperature. The internal friction measurements were performed from room temperature to 600 °C. Two typical internal friction peaks were detected corresponding to heating and cooling processes, respectively. The heating peak corresponds to a recrystallization process of deformed Al particles, which is influenced by many extrinsic parameters, such as measuring frequency, strain amplitude, heating rate, power particle size and compacting pressure. However, the intrinsic nature of the peak is originated from the micro-sliding of the weak-bonding interfaces between Al particles and increased dislocation density induced in compressing. The cooling peak with the activation energy of (1.64±0.06) eV is associated with the grain boundary relaxation, which can be interpreted as the viscous sliding of grain boundaries. The similar phenomena are also found in the Mg green powder compact.
Key words: Al powder compact; internal friction; sintering; grain boundary

1 Introduction

Sintering in powder metallurgy (PM) is generally defined as a process or phenomenon occurring with heating green powder compact under an appropriate temperature and atmosphere. The sintering plays a decisive role in the performance of the final products [1]. Therefore, to understand and describe the microstructure change of the green powder compact during the sintering process is of great importance. But the theoretical foundations for describing the sintering process in crystal metal powder compacts used in technical applications have up to now remained controversial [2]. During the sintering process of the metal powder compact, the powder contact boundary develops from poor mechanical bonding to strong metallurgical bonding. The driving force is generally considered as the reduction of free energy at the expense of minimizing the internal and external surfaces. However, the physical mechanisms on the driving force are still under discussion because of the complicated defect configurations induced in the metal powder compact [3,4].

Microstructure transition associated with the crystal defects evolution during the sintering process of the metal powder compacts has been widely detected by various techniques, such as position lifetime spectroscopy [1,5], transmission electron microscopy (TEM) direct observation [6], measuring shear viscosity or viscous Poisson's ratio [7]. In addition, the indirect measurements of electrical conductivity, thermal expansion, tensile strength, etc, are traditionally applied to understanding the sintering process [1]. However, the methods mentioned above are limited to a large extent due to the fact that the measurement of the samples in situ cannot be achieved. As a result, obtaining a dynamic changing rule of the microstructure during the sintering process is still a great challenge for metal powder compacts.

Internal friction (IF), Q^{-1}, is defined as energy dissipation from mechanical vibration energy irreversibly changing to thermal energy due to the intrinsic nature of materials, which is so sensitive to solid defects in materials that can offer detailed information even at atomic scale [8]. The Al particles during compressing are subjected to a severe deformation, as a result the powder compact is in a deformed state. Though the IF mechanical spectroscopy associated with the

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microstructure transition of the deformed metal materials has been widely investigated, such as Al−Mg alloys after cold-working with the deformation degree of 80% [9,10], pure Mg [11], Mg−Cu−Mn alloy [12] and ultra-fine-grained Cu [7,13,14] which were deformed through an equal channel angular pressing (ECAP); however, to detect and understand the sintering process of the green powder compacts using the measurement of IF has not been found as yet. In our previous study, an idea was proposed to understand the microstructure transition and crystal defects evolution of the green powder compact during the sintering process using the IF technique [15]. The results disclosed that the evolution law of grain boundary of the Al powder compact during the sintering process can be effectively detected according to the appearance of corresponding internal friction peaks. With the aim to obtain comprehensive and systematical information of the metal powder compact during the sintering process, the internal friction behavior of the Al powder compact was further deeply investigated. We are also expected that the results achieved are meaningful and helpful for better optimization of the sintering procedure.

2 Experimental

The elemental materials of Al and Mg powder with the purity of 99.99% were supplied by Sinopharm Chemical Reagent Co., Ltd., China. Different particle sizes with 70, 90, 130, 200 and 315 μm were achieved by a series of sieves. The green powder compacts of Al and Mg with the dimensions of 65 mm × 5 mm × 1.2 mm were prepared through an uniaxial compression under 400 MPa in a rectangular steel die for the measurement of IF. Any treatments for the metal powder compact were not carried out before IF measurement.

A multifunction internal friction apparatus (MFIFA), manufactured by Institute of Solid State Physics, Chinese Academy of Science, China, was used to measure IF. The MFIFA mainly consisted of an inverted torsion pendulum and an automatic computer controlled system. By setting different measuring parameters, the dependences of temperature, strain amplitude, frequency and time on IF can be correspondingly obtained. Reference [16] provides detailed information on the MFIFA. The measurement of IF was conducted under vacuum atmospheres of 1×10^{-3}−1×10^{-2} Pa. If there are no special instructions, the measuring frequency, strain amplitude and heating rate were set as 1.0 Hz, 10×10^{-6} and 2 °C/min, respectively. The characterization of microstructure of the green powder compact was performed using a Leica DMI−3000M optical microscope.

3 Results and discussion

Figure 1 shows the microstructure of Al green powder compact with a green density of approximate 0.80 ρ_0 relative to bulk Al where ρ_0 represents the density of bulk Al. The average particle size is about 70 μm. The grain particles have a lamellar morphology, which can easily cause an effective powder-particle size after compression. The size is slightly different from the particle size measured by screening analysis. This is because the Al particles were severely deformed during the compressing, leading to lots of very small Al particles and sometimes widening the area of Al compact.

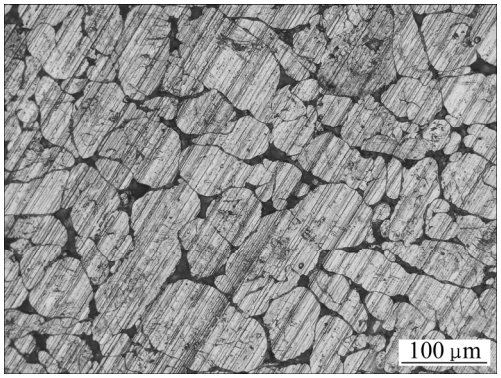

Fig. 1 Microstructure of Al green powder compact formed at 400 MPa

Figure 2 shows the typical IF and relative dynamic modulus (RDM) of the Al green powder compact during heating and cooling process from room temperature to 600 °C. The particle size of Al powder is 130 μm. It can be noted that the most outstanding feature is the appearance of two IF peaks corresponding to heating and cooling process, respectively. The positions of the peak temperature are separately at around 310 °C (named P_1) and 242 °C (named P_2) at the frequency of 1.0 Hz. The position of peak P_1 is independent on the measuring frequency. But the height of peak gradually decreases with increasing the measuring frequency. As a comparison, peak P_2 is dependent on measuring frequency which shifts towards higher temperature as the measuring frequency increases. In accordance with the appearance of the IF peaks, especially peak P_1, the RDM exhibits a much rapid drop. Peak P_1 is no longer observed during the cooling process and subsequent repeating heating process, which reflects a thermodynamically irreversible transition from green powder compact to a more equilibrium state. Peak P_2 is highly stable which always exists during cooling and repeated heating process after the first heating process. In other words, the appearance of peak P_2 should be related

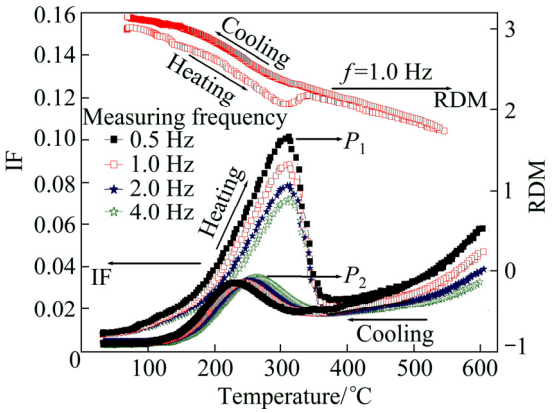

Fig. 2 IF and RDM dependence of temperature for Al powder compact during first run

to the appearance of peak P_1. In order to further understand the dependence of two peaks, the IF measurements were carried out in different temperature ranges from room temperature to 290, 310, 330 and 350 °C, respectively. Figure 3 shows the dependence as has been reported in Ref. [15]. It can be found that the appearance of peak P_2 is predominantly controlled by peak P_1, i.e., the appearance of peak P_1 in heating process and subsequently the appearance of peak P_2 in cooling process, no peak P_1, no peak P_2. Although the temperature of peak P_2 is around 242 °C, it appears during cooling process only when the temperature exceeds 310 °C during heating process. The results clearly demonstrate that the appearance of peak P_1 is the origin of the appearance of peak P_2. Figure 4 shows the heating rate dependence of peak P_1. With increasing the heating rate, the position of peak P_1 shifts towards higher temperature and the height increases. The dependence of Al particle size on peak P_1 is exhibited in Fig. 5. It can be noted that with the increase of particle size, the peak height increases and the peak temperature shifts towards higher temperature.

Peak P_2 is dependent on the measuring frequency, indicating a thermally activated relaxation process. Therefore, the activation energy E_a of the peak can be used as an index to identify its IF mechanism. In order to obtain exact peak temperatures at different frequencies, the net peak must be achieved by subtracting background IF (Q_b^{-1}) using the following equation:

$$Q_b^{-1} = A + B\exp[-C/(k_B T)] \quad (1)$$

where A is a material constant, k_B is the Boltzmann constant, T is the thermodynamic temperature, B and C are empirical parameters determined by the intercept and

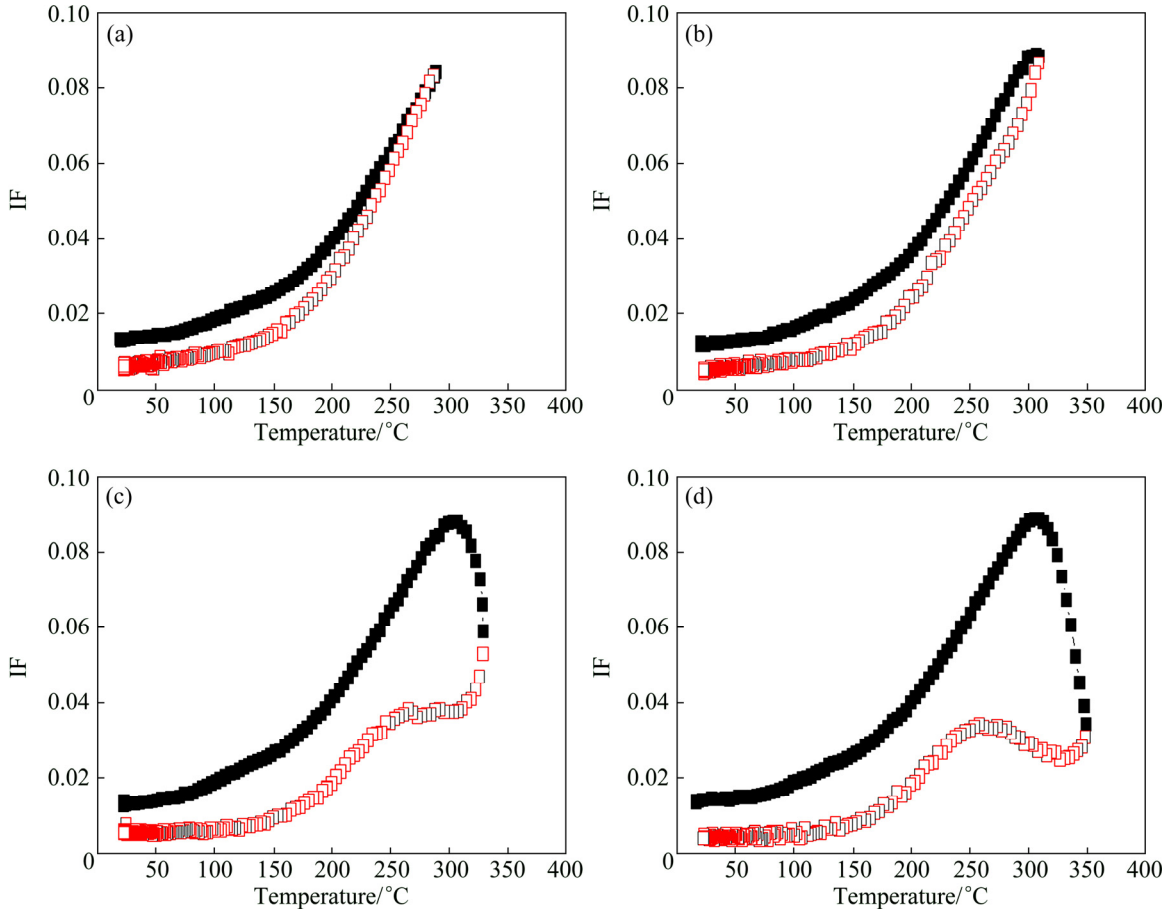

Fig. 3 Dependence of peaks P_1 and P_2 on heating temperature range from room temperature to 290 °C (a), 310 °C (b), 330 °C (c) and 350 °C (d) during first run

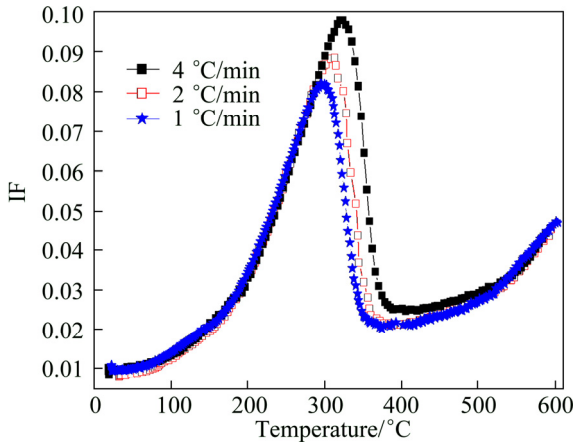

Fig. 4 Dependence of peak P_1 on heating rate at frequency of 1.0 Hz

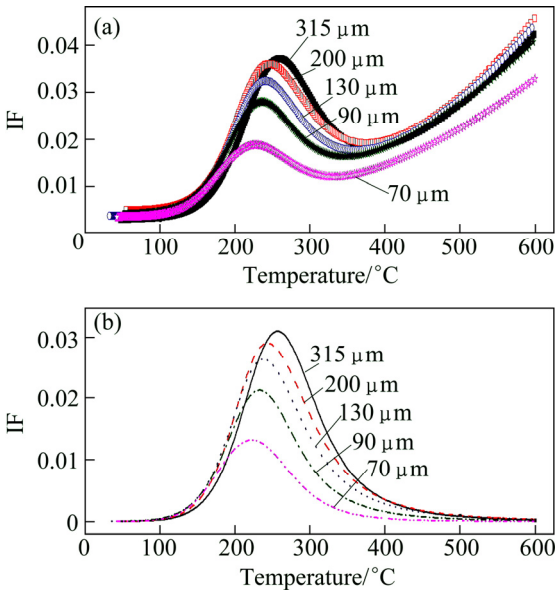

Fig. 5 Dependence of peak P_2 on Al particle sizes during heating process: (a) Original measuring results; (b) Treated results after subtracting background IF

slope of a semilogarithmic plot of ln Q_b^{-1} vs $1/T$ [8]. In practice, subtracting the background IF was conducted through a fitting procedure [17]. As a relaxation-type IF peak, its activation time τ should meet the Arrhenius relation:

$$\tau = \tau_0 \exp[E_a/(k_B T)] \quad (2)$$

where τ_0 is the pre-exponential factor. At the peak position, $\omega\tau_p=1$ is satisfied, where $\omega=2\pi f$ is the angular frequency and τ_p is the relaxation time at the peak temperature [18]. According to the τ_p values corresponding to different frequencies, an Arrhenius plot of (ln ω) versus $1000/T_p$ is given in Fig. 6. From the slope of the Arrhenius plots, the activation energy E_a of the peak was calculated as (1.64±0.06) eV, nearly independence of original Al particle sizes. The activation energy is almost the same to 1.692 eV in pure aluminum for the grain-boundary relaxation activation energy in Ref. [17]. According to the position and activation parameters of peak P_2, it is reasonable to suggest that the peak P_2 is related to the grain-boundary sliding. Traditionally, the grain boundary was described as an inhomogeneous structure composed of both the "good" and "bad" regions [19,20]. The sliding of the grain boundary is attributed to the sliding and climbing of the dislocations in the bad regions. Due to the viscous character, the grain boundaries are unable to support the applied shearing stress and have to slide in the direction of the stress, leading to relaxation under the applied stress. The temperature position of peak P_1 characterizes the onset of the crystallization process of Al.

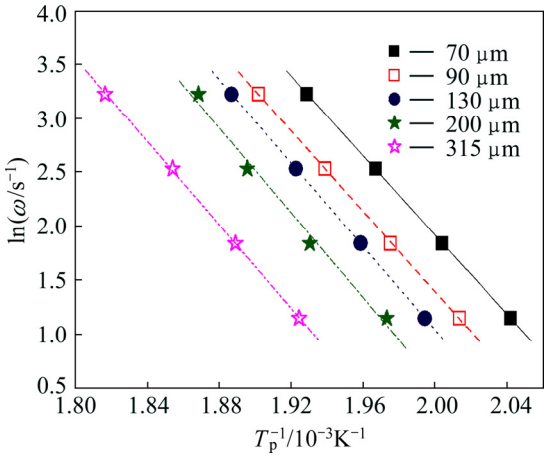

Fig. 6 Arrhenius plots of peak P_2 corresponding to different Al particle sizes

Now that peak P_2 appeared during cooling process is originated from the grain boundary relaxation, it can be concluded that a recrystallization process should take place during the heating process which is in good accordance with the appearance of peak P_1. The characteristics of peak P_1 are so similar to those of phase transformation IF peak, which is generally considered to result from the interface sliding between new phase and the parent phase [21,22]. Therefore, the IF mechanism of peak P_1 is suggested to associate with the weak-bonding interfaces between Al particles for the green powder compact. It is well acknowledged that during the preparation process of Al power compact, the applied compacting pressure will provoke the mechanical deformation and the displacement of Al particles, leading to the formation of interfaces among the Al particles in virtue of the mechanical interlocking bonding [2]. Relative to strong-bonding grain boundary, this kind of mechanical bonding interface can be reasonably considered as weak-bonding interface. During the IF measurement, when the applied external shear stress

exceeds the bonding strength of the weak-bonding interface, an interface microsliding process will be initiated by means of a bonding and debonding process, leading to the energy dissipation [23]. With increasing the temperature during the heating process, the viscous bonding force will become smaller and the interfacial microsliding becomes strong and easy. The energy dissipation gradually increases. But when the heating temperature approaches the temperature point of Al recrystallization, the vibration amplitude of Al atoms is enhanced and reaches the action range of the atomic force. The mechanical bonding between weak-bonding interfaces is gradually replaced by the metallurgical bonding. Accordingly, the weak-bonding interfaces among the Al particles are changed into grain boundaries, and the IF rapidly decreases [15]. From another point of IF background that is mainly controlled by the dislocations, there are large numbers of dislocations distributed in the deformed aluminum particles induced in compressing due to the severely deformation of Al particles. The intense increase in the IF background can begin in deformed state even at lower temperatures [10]. The dependence of peak P_1 on the temperature also reflects the evolution of the dislocation density. The rapid disappearance of the peak after recrystallization process is partly related to the decrease of dislocation density in the Al powder compact.

For better understanding the interface-sliding mechanism of peak P_1, the dependence of the peak on original Al particle size and compacting pressure applied during compressing was investigated. The results are shown in Figs. 7 and 8, respectively. It can be found that the height of peak increases with the decrease of Al particle size. Moreover, decreasing the compacting pressure, an increasing peak height can also be achieved. There is no doubt that the green powder compact prepared by the Al particle with smaller size possesses more amount of weak-bonding interface relative to that

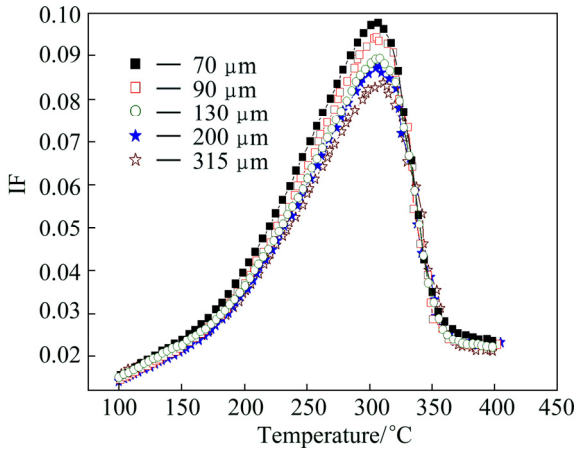

Fig. 7 Dependence of peak P_1 on Al particle sizes during heating process

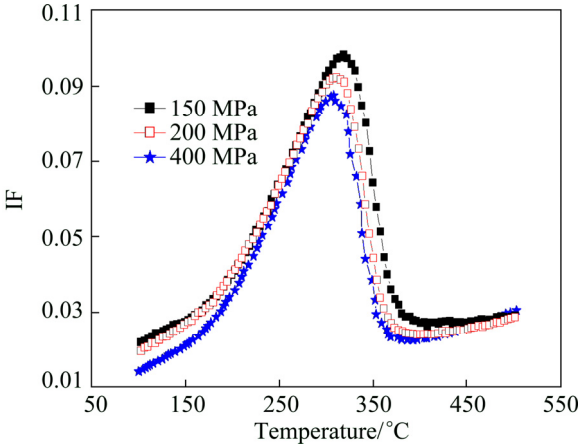

Fig. 8 Dependence of peak P_1 on compacting pressure during heating process

with larger Al particles size, i.e., the smaller the particle size, the more the interface amount. On the other hand, increasing the compressing pressure, the bonding strength between Al particles is greatly enhanced, and the micro-sliding of the interfaces becomes difficult owing to increased viscous force, leading to the decrease of peak height. The results mentioned above further confirmed the propose that the micro-sliding of interfaces between deformed Al particles should be the predominant reason for the appearance of peak P_1.

GOLOVIN et al [9,10,13,14,24] studied the IF behaviors of deformed metal alloys of Al−Mg, pure Cu and Grade-4 Ti. A pseudo-peak was found in the deformed state of the alloy. The appearance of the peak was interpreted as follows: the IF background in the deformed metals is higher owing to higher density of dislocations, and begins to grow intensely even at lower temperature than that in the as–annealed state. Upon heating in a temperature range of recrystallization, the IF values in this temperature range drop as the volume fraction of recrystallization in the sample increases and the dislocation density decreases therein. In addition, FAN et al [11] investigated the IF behavior of pure Mg deformed by ECAP technique. An irreversible peak was observed, which is related to the change of the structural state. It was suggested that the peak should be associated with the recrystallization involved with migration of grain boundary and reconfiguration of dislocations. Figure 9 shows the dependence of the height of peak P_1 on the strain amplitude for the Al green powder compact. The peak height increases with increasing the strain amplitude. The effect of strain amplitude is usually regarded as the evidence for the existence of dislocations and can be understood in terms of dislocation model developed by GRANATO and LÜCKE [25]. As a result, though peak P_1 is arisen from the micro-sliding of weak-bonding interface between Al particles, it is also

associated with the change of dislocation density during the sintering process.

As a comparison, the temperature dependence of IF of the Mg green powder compact was also investigated. Similar results, as shown in Fig. 10, were obtained as those in the Al powder compact. Two IF peaks are observed during heating and cooling process separately. The cooling peak with the activation energy of 1.25 eV originates from the Mg grain boundary relaxation. According to the experimental results, a propose is assumed that the IF results appeared in the Al green powder compact may be a general phenomenon for the meal powder compacts during the sintering process. Due to the first time to study the microstructure transition of green metal powder compacts using the IF technique, further investigations are urgently needed to deeply understand the IF mechanism, especially the formation of the two IF peaks.

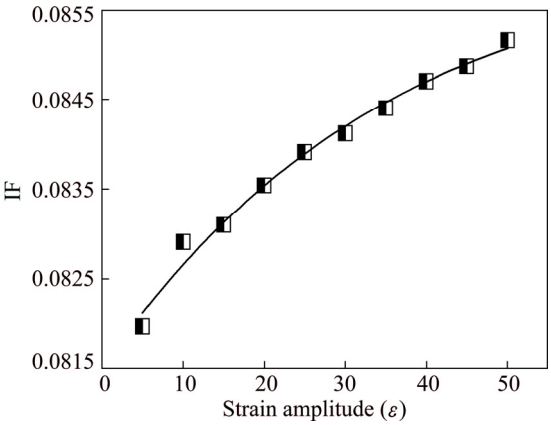

Fig. 9 Dependence of height of peak P_1 on measuring strain amplitude at measuring frequency of 1.0 Hz

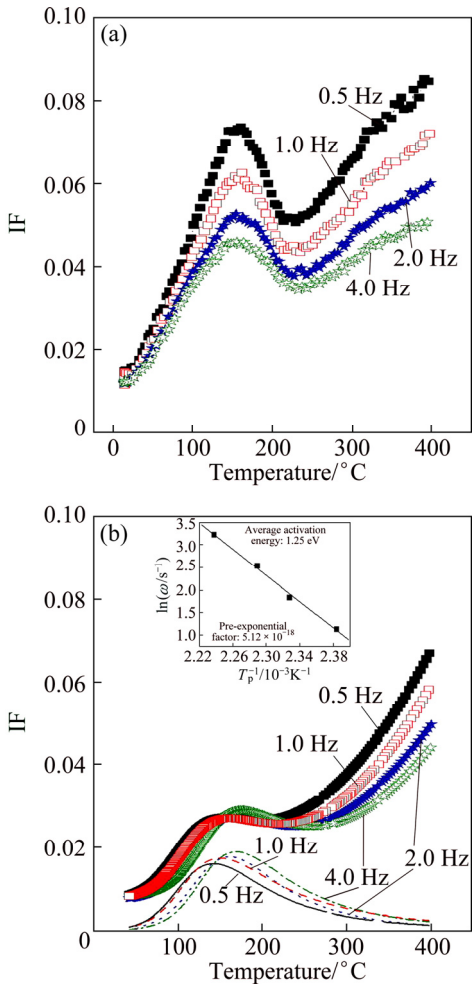

Fig. 10 IF dependence of temperature for green Mg powder compact during first run at different measuring frequencies: (a) Heating process; (b) Cooling process (Inset is Arrhenius plot of cooling IF peak)

4 Conclusions

1) The temperature dependence of IF of the Al powder compact during the sintering process was investigated using a multifunction internal friction apparatus ranging from room temperature to 600 °C. Two typical IF peaks (P_1 and P_2) were found corresponding to heating and cooling processes, respectively. Peak P_1 disappears after first heating process and peak P_2 appears in the subsequent cooling process.

2) Peak P_1 is irreversible and dependent on the measuring frequency, heating rate, compacting pressure and Al particle size, which is related to the recrystallization process. However, the intrinsic mechanism of the peak is resulted from the micro-sliding of weak-bonding interfaces between the deformed Al particles and increased dislocation density induced by compressing. The disappearance of the peak can be interpreted as the end of structure transition from weak-bonding interface to grain boundary due to the recrystallization process during first heating process.

3) Peak P_2 with the activation energy of (1.64±0.06) eV is associated with the viscous sliding of grain boundary formed in the recrystallization process.

4) Similar IF results as those in the Al powder compact are also found in the Mg green powder compact, indicating that there probably exists a general IF phenomenon during the sintering process for the metal powder compacts.

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烧结过程中铝粉末压坯的低频内耗行为

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摘　要：在室温至 600 °C 之间，研究铝粉末压坯在烧结过程中的内耗行为，其在升温和降温过程各出现一个典型的内耗峰。升温峰具有测量频率、应变振幅、升温速率依赖性，同时，随铝颗粒粒径、压坯成型压力的不同而变化。分析认为升温峰与形变铝颗粒的再结晶过程有关，但该峰产生的本征原因是形变铝颗粒之间弱结合界面的微观滑移，同时还与成型过程中位错密度的增加有关。降温峰与铝晶界的粘滞性滑移有关，属于晶界弛豫峰，其激活能为(1.64±0.06) eV。此外，镁粉末压坯具有与铝粉末压坯相似的内耗现象。

关键词：铝粉末压坯；内耗；烧结；晶界

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