Effects of glass addition on sintering and magnetic properties of $3Ba_{0.5}Sr_{0.5}O{\cdot}2CoO{\cdot}12Fe_2O_3$ for high frequency applications

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The effect of glass addition on the magnetic properties of $3Ba_{0.5}Sr_{0.5}O{\cdot}2CoO{\cdot}12Fe_2O_3$ (Co_2Z) ferrites was investigated. The densification of Co_2Z ferrites was enhanced by addition of glass due to the liquid phase sintering. Although the initial permeability decreased slightly, the quality factor was improved over a wide frequency range with the addition of glass. The resonant frequency shifted to a higher frequency range with increasing the addition of glass. Therefore, the addition of glass in Co_2Z ferrites is an effective method to improve the magnetic properties for application in the radio frequency range.

1. Introduction

In order to reduce the size of devices in communication systems, magnetic components must also be miniaturized. In recent years, the multilayer chip inductor has been developed to increase the volume efficiency. The present operating frequency is 200–300 MHz for much electronic equipment, such as pagers, car telephone and cordless phones. The most commonly used materials used in multilayer chip inductors for high frequency applications are NiCuZn ferrites or nonmagnetic materials, such as low temperature cofiring ceramics (LTCC). The use of nonmagnetic materials is clearly essential for high frequency operation since NiZn ferrites typically exhibit severe property changes above 200 MHz due to the Snoek limit [1]. The maximum quality factor of multilayer chip inductors made with nonmagnetic materials is above 500 MHz. However, the quality factors at frequencies around 200–300 MHz are much lower than the values at higher frequencies. Therefore, it is desirable to develop a material that has a higher quality factor than nonmagnetic materials at 200–300 MHz and can be used in making multilayer chip inductors.

Improved densification of ceramics at lower temperatures can be achieved by optimizing the powder morphology, adding glass flux and optimizing the sintering profile [2–4]. Of the above methods, lowering the sintering temperature by with addition of glass is the most effective and least expensive technique.

Magnetoplumbite ferrites with hexagonal structure have revealed a higher dispersion frequency than that of nickel ferrites, because of the magnetic anisotropy of the magnetoplumbite [5]. Among those ferrites, Kimura [6] observed that $3Ba_{0.5}Sr_{0.5}O{\cdot}2CoO{\cdot}12Fe_2O_3$ (Co_2Z) ferrite has the best magnetic properties (such as permeability and quality factor) above 200 MHz. However, the densification temperature of Co_2Z always exceeds above 1200°C and the magnetic properties of Co_2Z with glass addition have not been reported.

In this paper, the sintering behavior and magnetic properties of Co_2Z with addition of glass flux were investigated. Relationships between the sintering temperature, microstructure evolution and magnetic properties of flux-sintered chip inductors are presented.

2. Experimental procedure

$3Ba_{0.5}Sr_{0.5}O{\cdot}2CoO{\cdot}12Fe_2O_3$ ferrites used in this study were prepared following the methods reported by Kimura *et al.* [6]. The Co_2Z ferrites were prepared from reagent-grade $BaCO_3$, $SrCO_3$, Co_3O_4 and Fe_2O_3, which were mixed and then calcined at 1200°C for 1 h. The calcined powders were added with various amounts of Bi-Zn-B glasses (EG 2735, Ferro glass) and then milled for 60 hr using YTZ balls. The powders were dried in an oven and then added with PVA for granulation. The powders were dry-pressed at 150 MPa into toroidal bodies. These specimens were then debindered at 500°C and sintered at various temperatures from 850°C to 1250°C for 2 hr. The density of the sintered samples were determined by the Archimedean method. The microstructure was observed by scanning electron microscopy. Magnetic properties (initial permeability (μ'), imaginary permeability (μ'') and quality factor (Q)) were measured by the LCR meter (YHP 4291A, YHP Co. Ltd.) from 1 MHz to 1.8 GHz.

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3. Results and discussion

3.1. Densification of Co_2Z ferrites by addition of glass

Fig. 1a shows the variation of linear shrinkage of Co_2Z ferrites containing various amounts of glass versus sintering temperature, indicating that the densification was enhanced by liquid phase sintering. It can be seen that the linear shrinkage of the ferrites fired below 1050°C was substantially increased by doping glass to the Co_2Z ferrites and that the density increased steadily with temperature. Above 1050°C, the sintering density of Co_2Z ferrites decreased slightly with addition of higher glass content. The temperature at which the maximum density was achieved depended on the glass content. The larger the amount of glass addition, the lower the temperature.

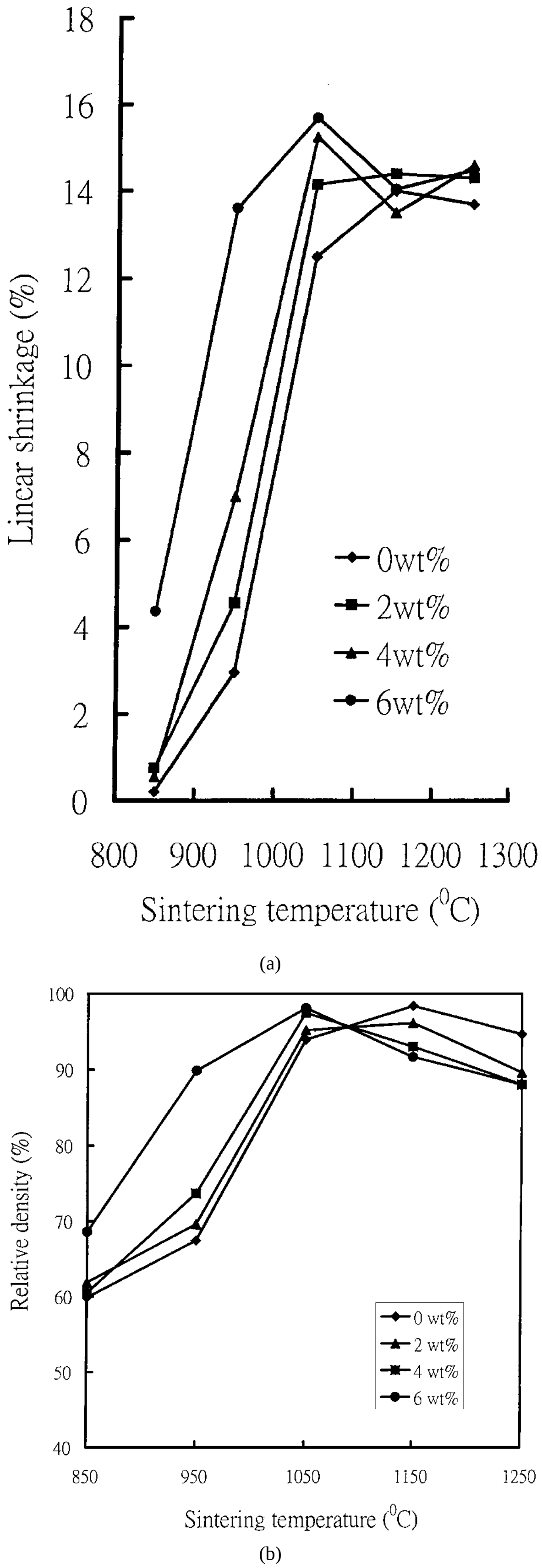


Figure 1 Linear shrinkage (a) and relativity density (b) of Co_2Z ferrites containing various amounts of glass versus sintering temperature.

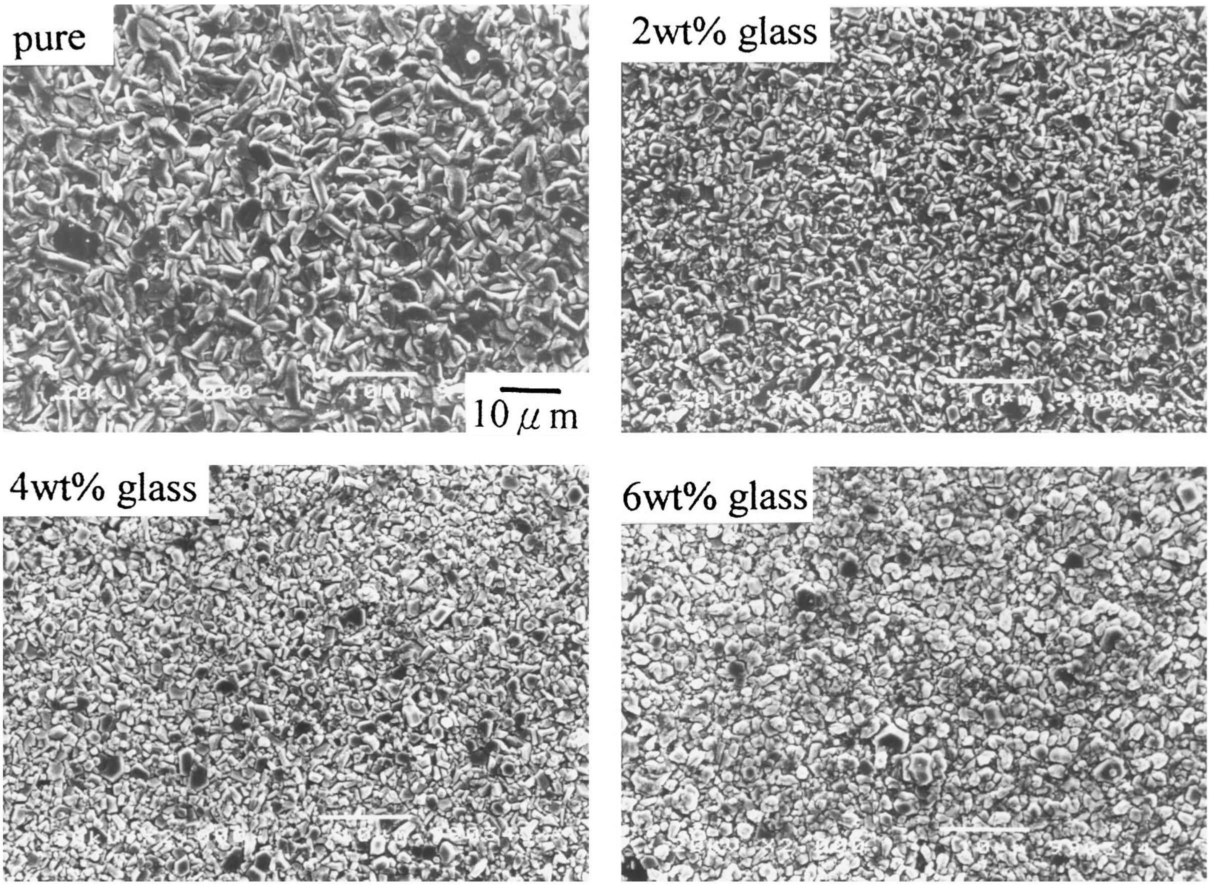
3.2. Microstructure of Co_2Z ferrites with addition of glass

Typical microstructures of sintered ceramics are shown in Fig. 2. At 1050°C, the amount of pores appears to decrease with increasing amount of glass added. The microstructure of the samples all show well-formed hexagonal plate-like grains of quite uniform size (Fig. 2a). Moreover, the grain size of the ferrites with added of glass is smaller than that of pure ferrites. This is an interesting observation and suggests that the thick films of glass, rich in grain boundaries of ferrite, interfere with the crystallization and inhibit further grain growth at 1050°C. At 1150°C, the particle size and density of pure Co_2Z ferrites increased. An important observation at this temperature was the loss of the well-formed hexagonal plate-like structure in the glass added ferrites and the start of discontinuous grain growth in the high glass added samples (Fig. 2b). Therefore, the density of Co_2Z ferrites, with the addition of glass above 4 wt%, sintered at 1150°C decreased slightly compared with samples sintered at 1050°C (Fig. 1b) due to porosity associated with discontinuous grain growth. When the sintering temperature increased to 1250°C, there was a sudden increase in grain size, as shown in Fig. 2c.

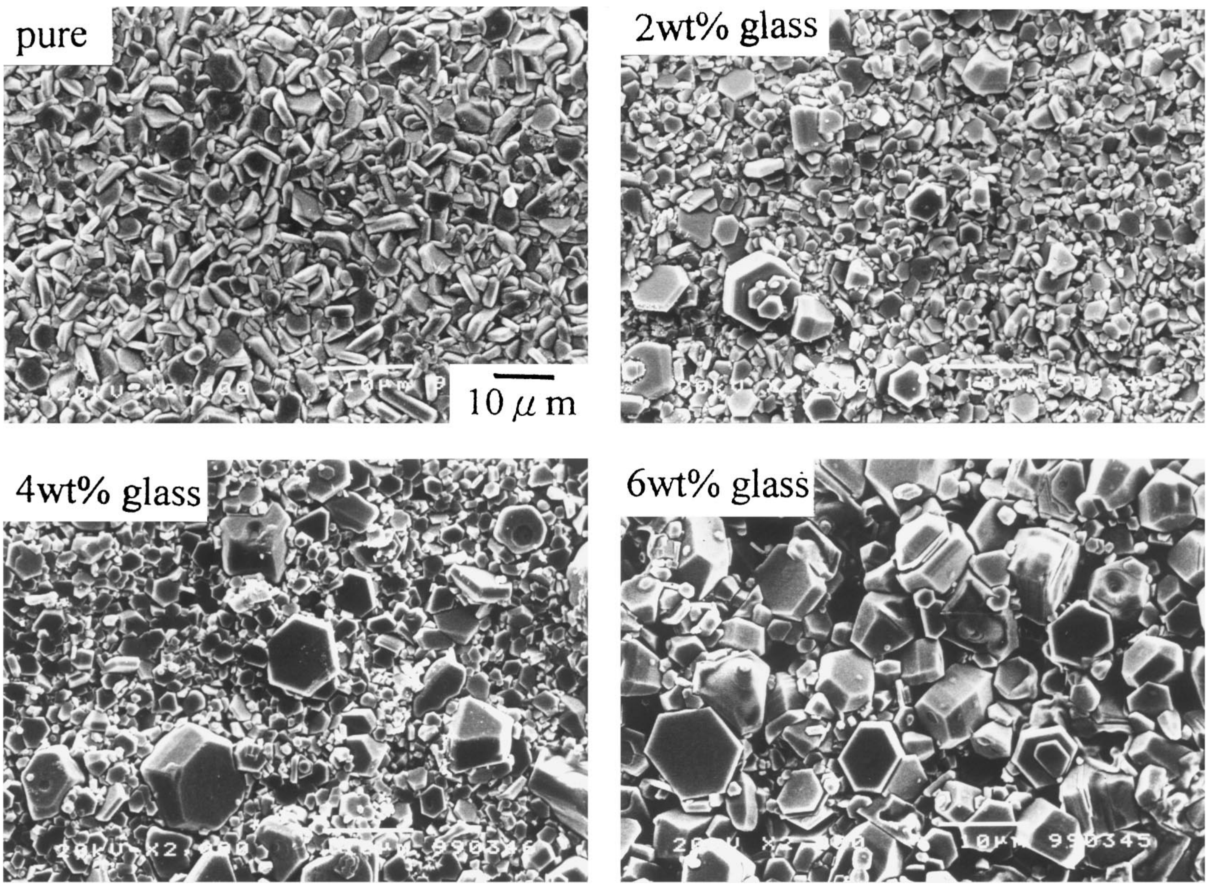
3.3. Magnetic properties of glass-sintered Co_2Z ferrites

The initial permeability and Q-factor of the ferrites containing various amounts of glass sintered at 1050 and 1150°C are shown in Figs 3–5. The changes in magnetic properties were strongly influenced by glass content. The initial permeability of the ferrites sintered at both 1050 and 1150°C decreased with greater levels of added glass (Fig. 3a and b). This may result from the nonmagnetic ions by the glass entering the crystal lattice of the ferrites, thus reducing the saturation magnetization, and hence the initial permeability.

The initial permeability of pure ferrites and ferrites containing 2 wt% glass increased as the sintering temperature increased from 1050 to 1150°C. This may result from the increase of the grain size and density, as shown in Figs 1 and 2. The pure Co_2Z ferrites show a lower resonant frequency than the ferrites with added glass (Fig. 4a and b). In general, the imperfections such as second phase on grain boundaries and pores will induce local demagnetizing fields, which will add to the anisotropic field and raise the resonance frequency [7]. As the sintering temperature increased from 1050 to 1150°C, the amount of pores in the pure ferrites decreased and the grain size increased. Therefore, the

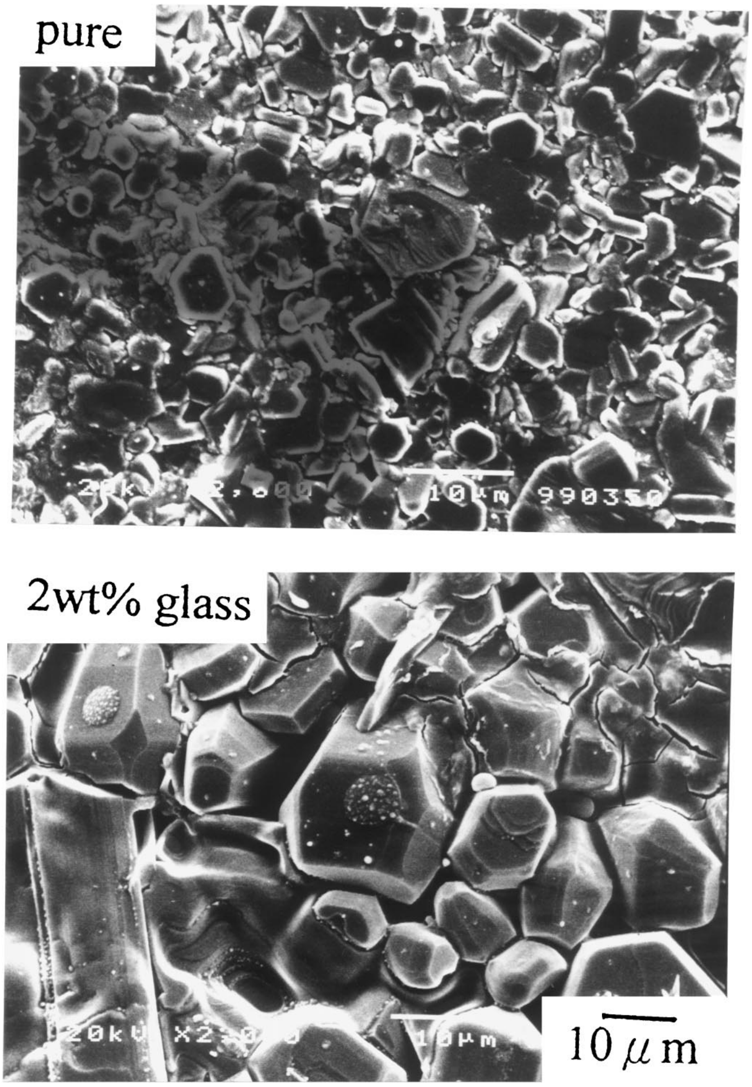


(a)



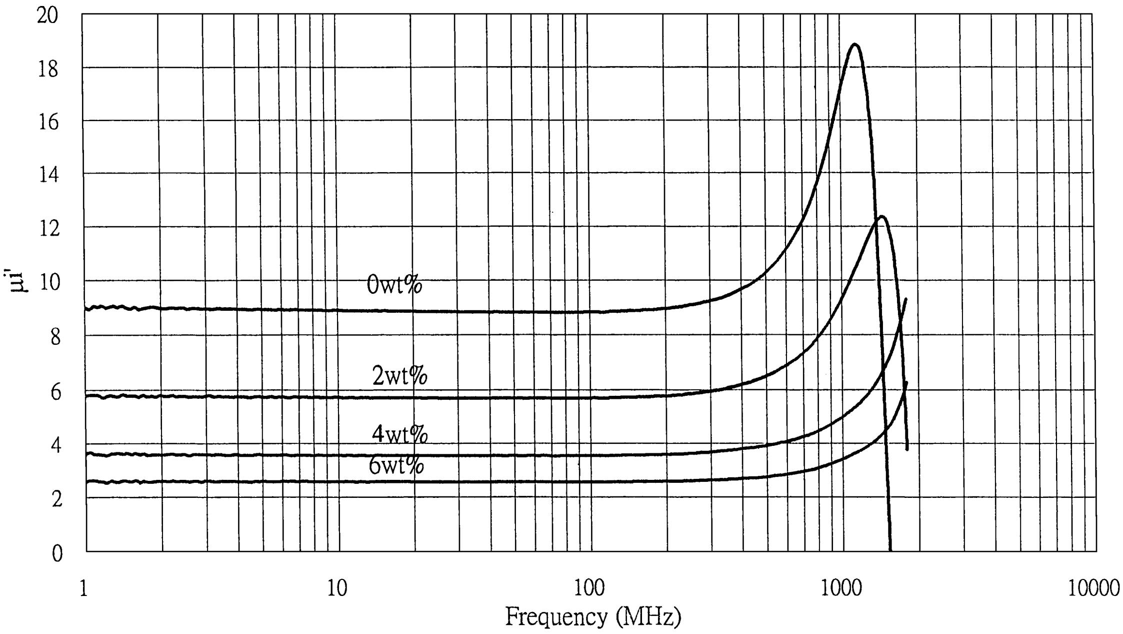
(b)

Figure 2 SEM micrographs of fracture surfaces of Co_2Z ferrites sintered with different glass contents at (a) 1050°C, (b) 1150°C and (c) 1250°C.



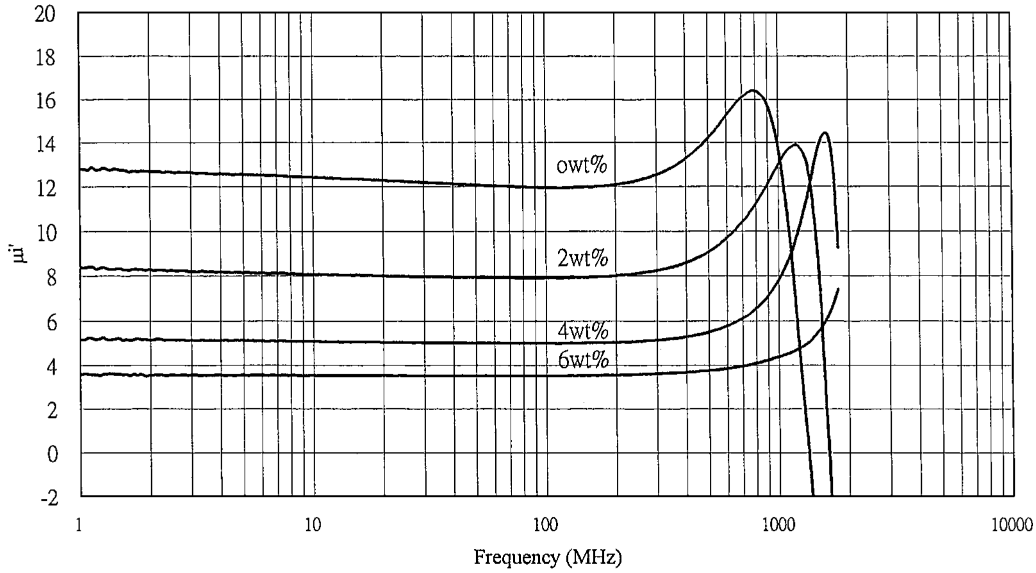
(c)

Figure 2 (*Continued*).



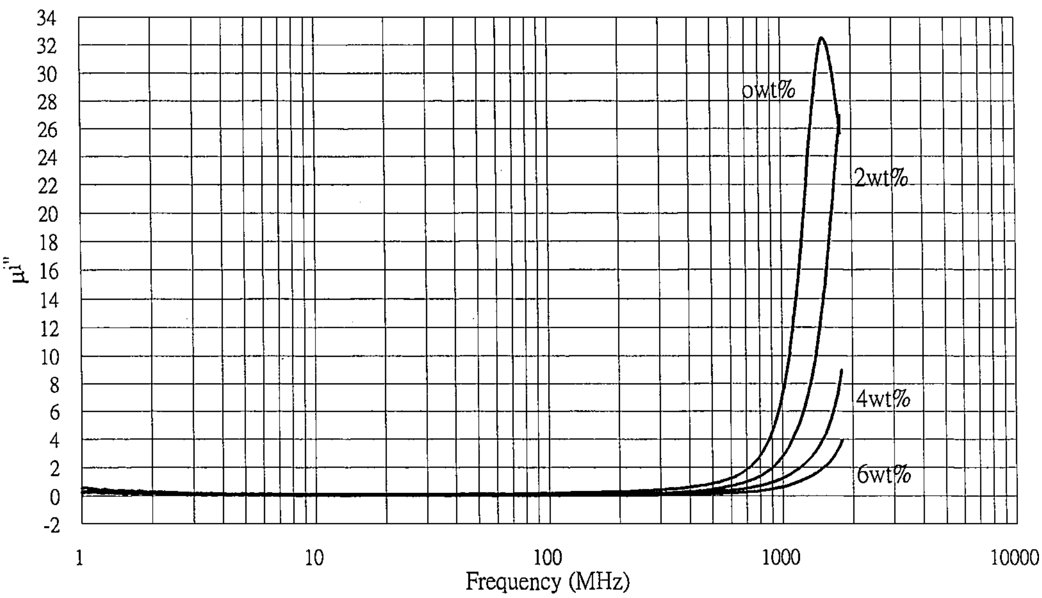
(a)

Figure 3 Initial permeability of Co_2Z ferrites sintered with various amounts of glass at (a) 1050°C and (b) 1150°C.

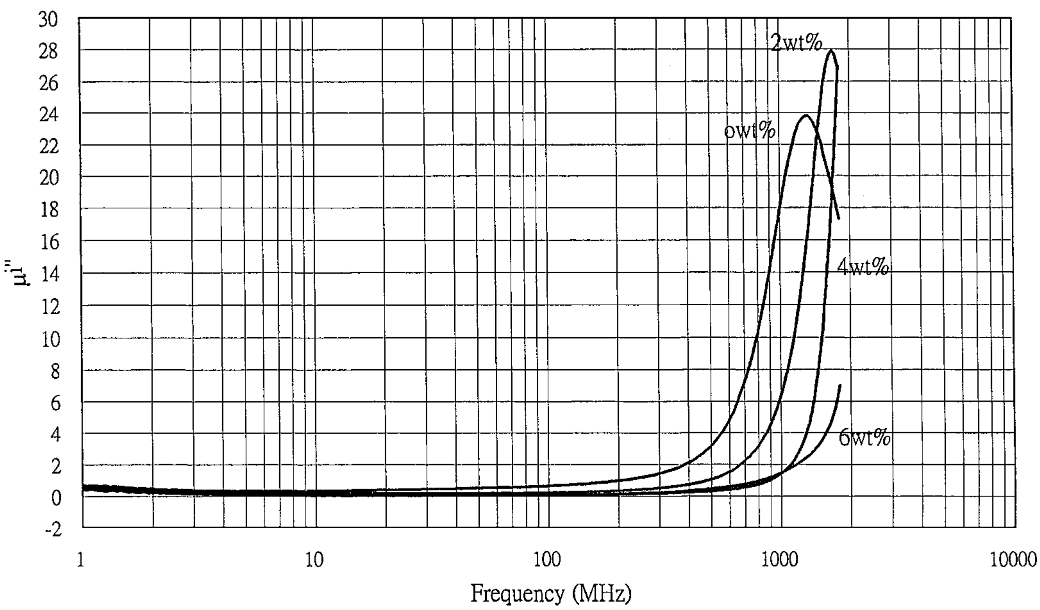


(b)

Figure 3 (*Continued*).



(a)



(b)

Figure 4 Imaginary part of the permeability of Co_2Z ferrites sintered with various amounts of glass at (a) 1050°C and (b) 1150°C.

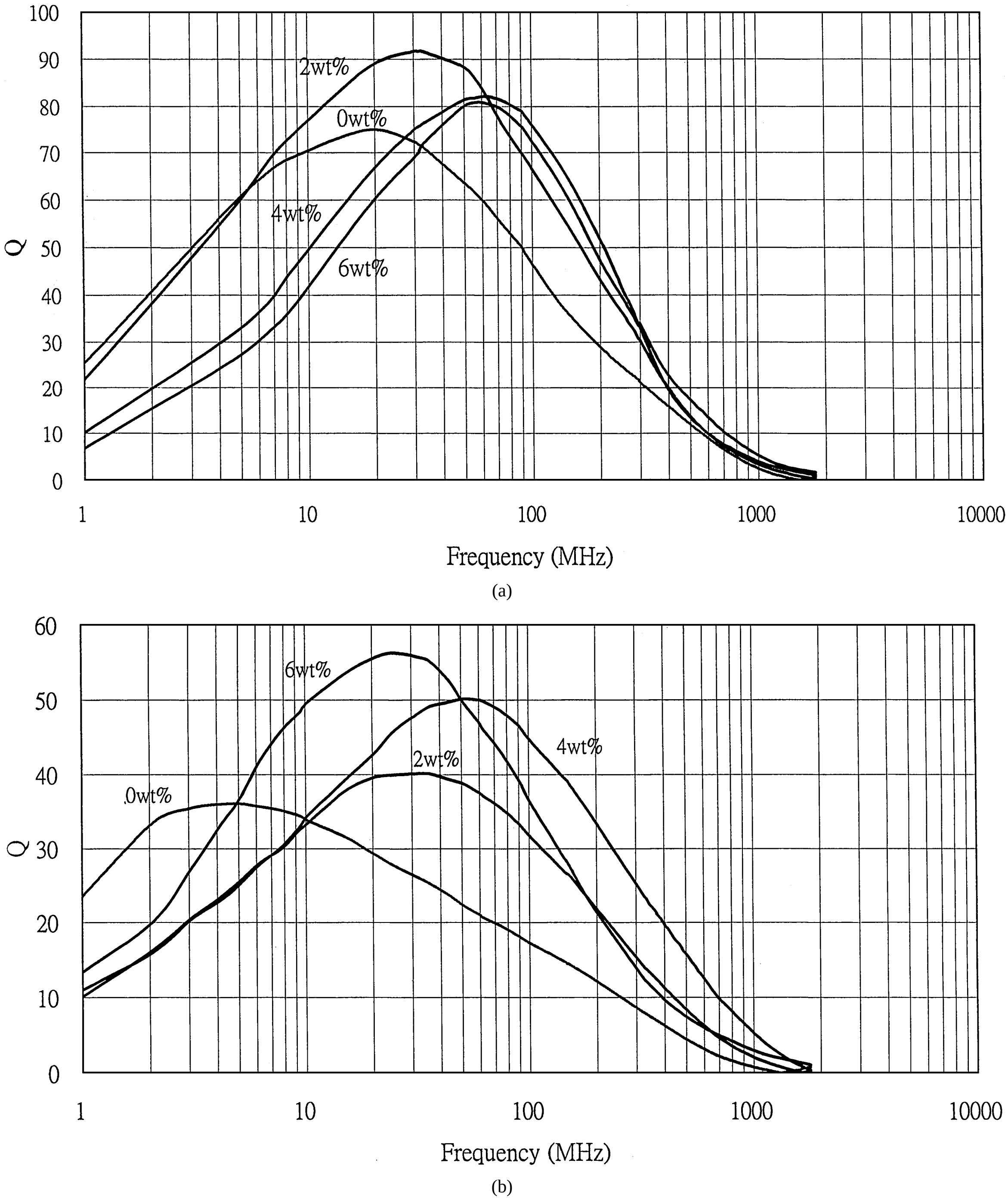


Figure 5 Q-factor of Co_2Z ferrites sintered with various amounts of glass at (a) 1050°C and (b) 1150°C.

resonance frequency of pure ferrites decreased as the sintering temperature increased from 1050 to 1150°C as a consequence of the decrease of demagnetizing field (Fig. 4a and b). It also has become clear that the main reason for magnetic loss in a broad frequency below the spin resonance frequency is the presence of domain wall [8]. As the grain size increases, the formation of domain walls becomes increasingly energetically favorable [9], so that their total effect on magnetic loss is greatly increased. As the sintering temperature increased from 1050°C to 1150°C, the grain size of the pure ferrites increased, which resulted in the value of pure ferrites sintered at 1050°C is larger than 1150°C as a result of the decrease of domain wall loss (Fig. 5a and b). However, the maximum Q value of the ferrites shifted to a higher frequency and increased as the amount of added glass increased, presumably because the glass phase prevented a loss contribution from domain wall movement. It is significantly to note that although the ferrites with added glass had a lower permeability, the Q factor was improved over a wide frequency range (above 200 MHz), as is indicated from a comparison of Fig. 3 with Fig. 5. Therefore, the addition of glass is an effective method in improving the magnetic properties of Co_2Z ferrites for application in the radio frequency range.

4. Conclusion

1. The densification of Co_2Z ferrites was enhanced by addition of glass due to the liquid phase sintering.

2. Glass addition decreased the initial permeability of Co_2Z ferrites slightly but improved the quality factor over a wide frequency range.

3. The resonant frequency of Co_2Z ferrites shifted to a higher frequency range as the added glass increased.

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