Thermal Magnetic Behavior of Nanocrystalline Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ Ribbons

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Abstract. Thermal magnetic behavior and microstructure of Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ ribbon annealed at different temperatures were investigated. It was indicated that the onset crystallization temperature of the ribbon was around at 530 °C and its Curie temperature can be enhanced from 320 °C to 385 °C by annealing. X-ray diffraction structural analysis showed that a single α -FeSi crystalline phase with grain size about 12 nm was precipitated from amorphous matrix for FeCuNbSiB ribbons annealed between 530 °C and 580 °C. The α -FeSi phases were octahedral shape confirmed by the morphology simulation and the AFM method. The thermal magnetic behaviors were complicated, as the μ_i can be divided into two modes during increasing temperature, but for the Bs and Ms shown a normal decrease with increasing temperature. The coercive force Hc and remanence ratio α have shown a continuous monotonic decline in single-phase ribbon with increasing temperature, and increased to a max value or a peak and then decreased to zero in biphasic ribbons. The thermal behaviors of magnetic properties were controlled by the exchange coupling interaction between the α -FeSi grains and the intergranular amorphous phase. It is important to maintain an efficient exchange coupling interaction between α -FeSi grains and the residual amorphous matrix, and it is necessary to sustain the higher Curie temperature for the application of the FeCuNbSiB ribbons in high temperature by controlling the size and crystallization degree of α -FeSi grains.

Introduction

With the unique duplex ultrafine structure of nanocrystalline and amorphous phases, the nanocrystalline soft magnetic alloys successfully achieve excellent performances [1-4], such as high saturation flux density, high permeability, and low loss. Nevertheless, the magnetism of nanocrystalline soft magnetic alloys are sensitive to exoteric temperature, and can be lost completely as the temperature exceeds Curie temperature (T_c)[5]. Studying on the thermal behaviors can be obtain necessary information of the nanocrystalline magnetic alloys for the potential applications. Due to the two-phase structure with quite different Curie temperatures, the effective magnetic anisotropies and saturation magnetizations, the thermal behavior of nanocrystalline soft magnetic materials are complicated and affected by many factors [6-8]. Kulik and Hernando [9] investigated the relationship between the crystallization annealing temperature (T_a) and the coercive force (H_c) of Fe_{73.5}Cu₁Ta₃Si_{13.5}B₉ nanocrystalline alloy in the temperature range from 480 oC to 580 oC and found that a maximum value (T_k) existed at H_c-T curve and changed with the annealing temperature. Hernando proposed [10] a two-phase structure of phenomenological theory to explain this phenomenon. Yanzhong Zhang [11] studied the thermal behavior of permeability on the nanocrystalline alloy, and made a qualitative interpretation for the experimental results.

In this paper the influence of annealing temperature on the structure and thermal magnetic properties of Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ nanocrystalline alloy have been investigated. The thermal magnetic properties after a thermomagnetic treatment are characterized by the quasistatic hysteresis loops concerning its certain shapecoercivity, , saturation magnetization, remanence and so on. The

structures of nanocrystallines have been investigated using X-ray diffraction in transmission geometry. The correlation of structure and magnetic properties of the nanocrystalline alloy has been analyzed and discussed.

Experimental

Cast ingot of the soft magnetic alloy with nominal composition of Fe_{73.5}Cu_{1.0}Nb_{3.0}Si_{13.5}B₉ (in at.%) was obtained by arc melting of pure metals Fe, Cu, Nb, Si and Fe-B alloy (the purities of all raw metals are higher than 99.8 wt%, and Fe-B alloy is 99.9% wt%) under pure Ar atmosphere. The amorphous ribbons with about 10 mm in width and 30 μ m in thickness were prepared by a single-roller melt spinning method using above alloy ingot. The phases of as-quenched and annealed samples were examined by X-ray diffraction (XRD) using D/max-2500/PC with CuK α radiation. The microstructure of the nanocrystalline soft magnetic alloy was evaluated using atomic force microscope. Afterwards these ribbons were wound into toroidal core samples of 12 mm inner diameter and 20 mm outer diameter. To obtain the characteristic nanocrystalline structure, these toroidal samples were submitted to isothermal annealing at temperatures between 100 and 580 oC for 0.5 h under the protection of hydrogen atmosphere. Then the initial magnetic permeability of the samples before and after the thermal treatment was measured by the impact method with H of 0.08A/m. The DC magnetic properties were measured by the CL6-1, and the saturation magnetization (Ms), saturation magnetic flux density (Bs) and coercivity (Hc) were attained from the DC hysteresis loop, and the rectangle ratio Br/Bs also been calculate by the datum.

Results

Microstructure Characteristics

X-ray Diffraction

In order to analyses the microstructure of annealed Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ ribbons, XRD experiments are conducted and shown in Fig. 1. It is well know that the as-quenched ribbon, a typical amorphous phase, exhibited abroad scattering peak in the diffraction curve. While the patterns of the annealed samples show the characteristic (110), (200), (211), (220) and (311) diffraction peaks of a-FeSi crystalline phase and (200) diffraction peaks of Fe₃Si crystalline phase, indicating that all the annealed samples have crystallized partially and their microstructure consists of the residual amorphous phase, the a-FeSi crystalline phase and a little Fe₃Si crystalline phase. The content of crystalline phase increase accordingly with the increase of annealing temperature, and the intensity of diffraction peaks become stronger, as shown in the Fig. 1.

Figure 1. XRD patterns of annealed FeCuNbSiB alloy

From the X-ray diffraction structural analysis, crystal lattice constant of a-FeSi (a_0), the content of Si in a-FeSi, average grain size of a-FeSi (d_a), the degree of short-range order δ , average grain size of amorphous phase (d_A), and t can be estimated by means of the Scherrer formula, as shown in Table 1.

It can be seen that the d_a of the annealed FeCuNbSiB ribbons are about 12 nm. Though there exist a little difference in grain size between the annealed samples, the grains size are small enough to randomly average out the local magneto crystalline anisotropies, and achieve an excellent magnetic softness according to Herzer's random anisotropy model [12].

Table 1. Micro-structural parameters of the FeCuNbSiB ribbons obtained by X-ray diffraction structural analysis

No.	Ta(°C)	α Fe(Si)		Am			
		a_0 (nm)	Si(at%)	d_a (nm)	δ (nm)	d_A (nm)	t (nm)
1	500	0.283715	20.49399	9.5	2.672675	0.25346	1.5166
2	520	0.283415	21.82911	13.5	2.716663	0.25161	2.2996
3	530	0.28303	23.5425	12.4	2.409599	0.25267	1.3258
4	540	0.283654	20.76547	10.6	2.782814	0.24921	1.0537
5	580	0.283557	21.19715	14.3	2.749161	0.25149	1.3575

X-ray Topography Analysis of Nanocrystals

To achieve further information the internal structure of the FeCuNbSiB ribbons is investigated by the morphology simulation method according to popa theory [13] and the AFM technique. The morphology simulation result and the AFM micrographs for the sample annealed at 580 °C for 40 min are shown in Fig. 2. In Fig. 2a the simulation image of the nanocrystalline shows octahedral shape. Fig. 2b is the AFM morphology of the surface of the ribbon which agrees with the positions predicted from the simulation image. The bright yellow field images are due to the strong force between the sample surface and the probe, especially for the up part of nanocrystalline (the up part of the octahedron as shown Fig. 2a). The AFM results confirm the high percentage of crystallization and the crystallite size measured by the X-ray diffraction structural analysis. It should be observed that the average crystallite size determined by the XRD analysis (table. 1) is usually lower than the grain size observed in AFM (the d_a the annealed FeCuNbSiB ribbons are about 14nm from the Fig. 2b). The grains in the annealed samples contain sub-grains or dislocation cells which are separated by the low-angle grain boundaries [14]. The grain size obtained from the X-ray diffraction structural analysis is equal to the average size of the domains which scatter X-rays strongly. As a result, the XRD differentiates between sub-grains and dislocation cells, even if the miss-orientations are very difficult and small.

Figure 2. Microstructure of the FeCuNbSiB ribbons: a) morphology simulation by the Popa theory, b) the AFM micrographs of the sample annealed at 580 °C for 40 min

Magnetic Properties

Thermal - Saturation Magnetization Curve

Fig. 3 presents the dependence of the M_s of the annealed samples at different temperature. The M_s display a slight variation with different treat condition, except for the sample treated at 520 °C. The curies temperature (T_c) of the sample treated at 520 °C is lower than that of others samples obviously. This result is induced by fellow reasons. Firstly, the different can be responsible for the different crystallization degree in the samples with various treatments. Secondly, the increase of treat temperature accelerates the crystallization of ribbons, and result in an increase in grain volume fraction, which in turn leads to an increase in the curies temperature. Due to an increase in grain volume fraction, this can reduce the effective magnetic moment by change the magnetic exchange interactions of the magnetic neighbor grains. It is worth mentioning that the M_s of the samples decrease with the increase of the temperature. But with the increase of treat temperature, which induces the increase of grain volume fraction, the decrease of the M_s became slower until to the curies temperature.

Figure 3. M_s of the annealed samples at different temperature

The Thermal Behavior of the Initial Permeability

In order to characterize the high-temperature magnetic response as well as the Curie temperature of the FeCuNbSiB ribbons, the temperature dependence of initial permeability (μ_i - T curves) was measured. Fig. 4 showed the μ_i - T curves of as-quenched samples and the annealed ribbons (from 100 °C to 580 °C). On all μ_i - T curves a characteristic sharp Hopkinson peak is observed at the Curie point of the amorphous phase. Fig. 4a showed the μ_i - T curves of as-quenched ribbons, which is due to the faster decrease of magnetic anisotropy than that of annealed ribbons. It can be easily seen that an obvious T_c for the samples, which annealed less than 500 °C, is around 320 °C. When the annealing temperature exceeded to 500 °C, the T_c for the samples became more difficult to identify, indicating that the nanocrystals play an important role to change and enhance the T_c for annealed ribbons.

The above results implied that, when the annealing temperature is low, only one low temperature amorphous phase Hopkinson peak is appeared, indicating that under low annealing temperature the ribbons remains the single-amorphous phase structure, but it exist structural differences between the as-quenched samples and the low treat temperature annealed ribbons. With increasing annealing temperature, the Curie temperature of the amorphous phase is increased, and the width of the Curie peak is broadened (the temperature between the peak and the end point of peak temperature). When the annealing temperature reached 500 oC, besides the amorphous phase Hopkinson peak, the crystalline phase Hopkinson peak of α -FeSi phase also appeared in the μ_i - T curves with a double Hopkinson peak characteristic. The results are similar with the Fan's researches [15]. So the result descript that the α -FeSi phase has precipitated from the matrix, and compose biphas structure with the residual amorphous phase, which has been proven by X-ray diffraction results. With further

increasing annealing temperature, the Hopkinson peak width of right branch curve continues to increase, but the Curie temperature of the amorphous phase has shown a downward trend. When the annealing temperature was higher than 540 oC, the permeability decrease with increasing temperature, only appear α -FeSi phase Hopkinson peak at high temperature, and amorphous phase Hopkinson peaks is disappeared. But X-ray diffraction results indicate that the annealed ribbons still remain a little of amorphous phase at this stage.

Taking these findings, it can be found the thermal initial permeability behavior of nanocrystalline soft magnetic material has diversity, and was divided into single and two Hopkinson peaks or other two types. For the FeCuNbSiB ribbons, their transformation temperature was in range of 530 oC -540 oC.

Figure 4. Thermal behavior of the initial permeability for the FeCuNbSiB ribbons with different treatment: (a) as-quenched; (b) 100 °C; (c) 200 °C; (d) 300 °C; (e) 350 °C; (f) 400 °C; (g) 500 °C; (h) 520 °C; (i) 540 °C; (j) 560 °C; (k) 580 °C.

Thermal Behavior of Saturation Magnetic Flux Density

In Figure 5, the dependences of saturation magnetic flux density (B_s) on temperature for the annealed FeCuNbSiB ribbons at 800 kA/m are shown. According to the experiment results, the B_s at the room temperature shows an increasing trend with increasing annealing temperature, from 0.8 T of the amorphous ribbon to the 1.2 T of the ribbon treated at 530 °C, 540 °C, and 580 °C. It is considered that the crystalline phase (α -FeSi phase) induced the increase of the B_s for the ribbons treated above the 530 °C. From the figure, the B_s thermal behaviors of the ribbons have shown the similar change trend with each other. The saturation magnetic flux density B_s decrease with the increase of temperature, and decrease to zero around the Curie temperature like the permeability thermal behaviors. The reason will be discussed in the next parts of the paper in detail.

Figure 5. Bs of the annealed samples at different temperature

The Coercivity and Remanence Ratio Thermal Behavior

The thermal behavior of coercivity and remanence ratio (α) of the ribbons were measured from the room temperature to the temperature above the Curie temperature, and shown in fig. 6. For single-phase soft magnetic amorphous ribbon (as shown in fig. 6a and b), the coercive force H_c and remanence ratio α have shown a continuous monotonic decline with increasing temperature. When the temperature rose to the peak of amorphous phase Hopkinson and the zero point's temperature of the B_s , namely the Curie temperature, the coercive force H_c and the remanence ratio α are reduced to zero, and the ribbons loss magnetic properties, which similar to the results Hernando^[10] studied. For biphasic nanocrystalline ribbons (as shown in fig. 6c-f), When the temperature rose to the peak of amorphous phase Hopkinson and the zero point's temperature of the B_s , namely the Curie temperature, the coercive force H_c and remanence ratio α also reduce to zero, which similar to that of the amorphous ribbon. But in the process of increasing temperature, there exist very different behaviors between the amorphous ribbon and the annealed ribbons. From the figure 6, it is clear that the coercive H_c and remanence ratio α firstly increased to a max value or a peak and then decreased to zero with the increase of temperature, but this phenomenon didn't occurred in the amorphous ribbon. The peaks of H_c -T curves and α -T curves are occurred in the temperature range, in the range the permeability μ change rapidly, indicating that the peaks of H_c and α are related to the ferromagnetic-paramagnetic transition in the ribbons during increasing temperature. Another phenomenon can be found from the figure, the peak of H_c did not coincide with the peak of peak α in the position of temperature, and the temperature at the peak of H_c is higher than that of the α , implying that there exist different mechanism in the thermal behavior of H_c and α .

Figure 6. The temperature curve of coercivity and remanence ratio

Discussions

Analysis of the Super Paramagnetic Properties of α Fe-Si Nanocrystals

From above results, it can be conclude that the microstructure of the FeCuNbSiB nanocrystalline ribbons have a stronger effect on the thermal behavior magnetic properties. From the X-ray diffraction structural analysis, the FeCuNbSiB nanocrystalline ribbons contain the nano- α FeSi grains and amorphous phase. According to the magnetic theory, when the ultrafine magnetic α FeSi grains are isolated by the amorphous phase, the ribbons will exhibit a superparamagnetic characteristic. In the superparamagnetic stage, there is no interaction between the nano- α FeSi grains, when the magnetic anisotropy energy of the grain equal to thermal motion energy of the grain, the thermal activation energy may be able to overcome the magnetic anisotropy barrier, this induce that the magnetization vector of the ultrafine grain will no longer along the easy magnetization direction of the grains, but ceaselessly back and forth reverse between the different easy magnetization directions. So the magnetic properties of ribbons have show very different between the amorphous ribbon and the annealed ribbons. But for the superparamagnetic theory, there exist a critical grain size (D_c) for the FeCuNbSiB ribbons, and the D_c can be calculated by the following formula 1:

$$D_c = \sqrt[3]{\frac{600kT}{\pi K_1}} \quad (1)$$

Where k is Boltzmann's constant; T is the absolute temperature; K_1 is the magnetic anisotropy constant. If the grain size of the ribbons is smaller than the superparamagnetic critical grain size D_c , superparamagnetic characteristic will appears and the remanence and coercivity of the ribbons tends to zero, and need a high magnetization field to make the ribbons approaching the saturation magnetization.

The critical grain size D_c of the α -FeSi grains in the FeCuNbSiB ribbons at different T are calculated and shown in Figure 7. In the Figure the D_a is the average grain size of α -FeSi calculated from the X-ray diffraction structural analysis. It can be easily seen that the average size of α -FeSi grain is much smaller than the grain size of the critical grain size D_c , indicating that the average grain size of α -FeSi has to meet the superparamagnetic presentation condition. In fact, the FeCuNbSiB ribbons at room temperature ($T=25$ oC) does not appear the superparamagnetic characteristic for above results, the superparamagnetic characteristic were suppressed, which is directly related to the exchange coupling mechanism in nanocrystalline soft magnetic materials [16].

Figure 7. The critical grain size D_c of the α Fe (Si) grains in the FeCuNbSiB ribbons

Analysis of the Exchange Coupling Mechanism of α Fe-Si Nanocrystals

Using the amorphous random anisotropy Alben model, Herzer [17] successfully explained the origin of the excellent soft magnetic properties in the nanocrystalline soft magnetic material. He pointed out that when the α -FeSi grain size is less than the exchange coupling critical length D_e , the adjacent crystal grains will have exchange coupling effect, so that the local anisotropy of the crystal grains is effectively averaged out, and make the effective magnetic anisotropy of the nanocrystalline FeCuNbSiB ribbons to an extreme low level, therefore, the FeCuNbSiB ribbons possess high soft magnetic properties. For the nanocrystalline soft magnetic material, the D_e can be regarded as the critical exchange coupling length of about 35nm, greater than α -FeSi grain size D_a of 10-20nm (table 1). The D_e can be calculated by the following formula 2:

$$D_a < D_e = \sqrt{A_a / K_a} \quad (2)$$

The theory exchange coupling critical length proposed by Herzer is establish on the basic of Alben's single phase amorphous random anisotropy model, which is different with bipolar structure of nanocrystalline soft magnetic materials. In the nano-crystalline soft magnetic materials, the α -FeSi exchange coupling interaction between grains, in fact, are carried out and transferred by the amorphous phase between α -FeSi grains, as shown in the figure 8. So researching the exchange coupling interaction of the α -FeSi grains should consider the affect of the magnetic properties of the α -FeSi phase, in addition to considering the affect of the amorphous phase.

Indeed, for biphas structure, R. C. O. Handiey[18] expound in its monograph that when the distance between grains (t) is less than the exchange-coupled grains critical size Lex , the magnetization from one grain to the other grains is carried out by penetrating through the grain boundary phase, so the exchange coupling interaction between the α -FeSi grains must penetrate through the intergranular amorphous phase [19].

Compared with the exchange coupling critical length De , for the nanocrystalline soft magnetic ribbons, the Lex not only is relate to the A and K of α -FeSi phase, but also associated with the difference of saturation magnetization (δM_s) between the α -FeSi phase and the amorphous phase, which relate to the duplex microstructure parameters of nanocrystalline FeCuNbSiB ribbons. When δM_s reduce to zero, namely the saturation magnetizations of α -FeSi phase are equal to that of the amorphous phase, the Lex reach to the maximum, and the ribbons exhibit excellent magnetic properties. K.Y He consider [20] that when the saturation magnetization is equal in the grain phase and the amorphous phase, the equal induce the origin of excellent magnetic properties in the nanocrystalline FeCuNbSiB ribbons, for which there is need for further research.

Figure 8. Schematic diagram of nanocrystalline soft magnetic materials exchange coupling[21]

The Lex can be calculated by the formula 3:

$$Lex = \sqrt{A / [K_u + 2\pi(M_s^\alpha - M_s^A)^2]} \quad (3)$$

Where A is exchange integration constant; K_u is the magnetic anisotropy constant; M_s^α is the saturation magnetizations of α FeSi phase; M_s^A is the saturation magnetizations of amorphous phase. According to the formula 3, the Lex of nanocrystalline FeCuNbSiB ribbons at different temperature are calculated and shown in Figure 8. It is observe that the thickness t of the soft magnetic amorphous phase is smaller than the exchange-coupled grains critical size Lex , indicating that the exchange coupling interaction between the α -FeSi grains penetrate through the intergranular amorphous phase in the nanocrystalline FeCuNbSiB ribbons, which play important roles to improve the performance of the soft magnetic materials.

In the formula 3, note that the A , K , M_s^α , M_s^A all are the temperature variables, indicating the Lex is a temperature variables too, which will affect the thermal behavior of the nanocrystalline FeCuNbSiB ribbons. So further analyzing the relation between the Lex and the temperature is very significant to explore its effect on the magnetic thermal behavior of the ribbons. If the $K \ll M$, the formula 3 can be simplified to the formula 4:

$$Lex = \sqrt{A / 2\pi\Delta M^2} \quad (4)$$

Where ΔM is the difference of the M_s^α , M_s^A . By the magnetic theory, the $A = Ms$, and in order to maintain consistency with the test magnetic thermal behavior, the Ms is instead by Bs , and then the formula 4 can be further simplified to the formula 5:

$$Lex \propto \Delta M \sqrt{M_s / 2\pi} \quad (5)$$

Figure 9. Lex and t of the nanocrystalline FeCuNbSiB ribbons at different temperature

The Lex is calculated with formula 5 and shown in Figure 9. It can be found the Lex has a peak or valley during increasing temperature, and the temperature of peak or valley close to the characteristic temperature of μ , H_c , α . when Lex decreases with increasing temperature, the H_c and α exhibit an opposite change with increasing temperature, indicating the decrease of Lex induce the distance between grains (t) is larger than the exchange-coupled grains critical size Lex. Therefore, the exchange coupling interaction between the α -FeSi grains are weak, and the effective anisotropy are strong relatively. At the lower temperature area, the H_c and α change slightly with increasing temperature, indicating the exchange coupling interaction between the α -FeSi grains are sustained at steady state; but at the higher temperature area, the exchange coupling interaction between the α -FeSi grains are broken or weakened, inducing the decrease of the H_c and α with increasing temperature.

Through the above analysis, it is clear the exchange coupling interaction is a very important aspect specific for the nanocrystalline FeCuNbSiB ribbons. In the ribbons exchange coupling interaction between the α -FeSi grains mainly occurs via the amorphous phase. The amorphous phase has a much lower Curie temperature than the α -FeSi grains [22]. Approaching the Curie temperature of the amorphous phase, the exchange coupling interaction is largely reduced, and the soft magnetic properties degrade correspondingly. The temperature dependence of the magnetic properties in the nanocrystalline FeCuNbSiB ribbons demonstrates that it is important to maintain an efficient exchange coupling interaction between α -FeSi grains and the residual amorphous matrix, and it is necessary to sustain the higher Curie temperature for the application of the FeCuNbSiB ribbons in high temperature.

Conclusions

The microstructure and thermal magnetic behavior of the FeCuNbSiB ribbons annealed at different temperature are investigated in detail. All the annealed samples have crystallized partially, and their microstructure of the ribbons consists of the residual amorphous phase, the α -FeSi crystalline phase and a little Fe₃Si crystalline phase. The content of crystalline phase increases accordingly with the increase of annealing temperature. The α -FeSi grains in the annealed ribbons are octahedral shape, which is confirmed by the morphology simulation and the AFM method. The thermal behaviors of the original permeability of the ribbons are complicated, can be divided into a single Hopkinson peak of α -FeSi and double Hopkinson peaks of amorphous phases and α -FeSi phase. The B_s and M_s decrease with increasing temperature. Increasing the anneal temperature, which induces the increase of grain volume fraction, the decrease of the M_s became slower until to the Curie temperature. For

single-phase soft magnetic amorphous ribbon, the coercive force H_c and remanence ratio α have shown a continuous monotonic decline with increasing temperature. For biphasic nanocrystalline ribbons, the coercive H_c and remanence ratio α firstly increased to a max value or a peak and then decreased to zero with the increase of temperature. The temperature at the peak of H_c is higher than that of α , implying that there exist different mechanisms in the thermal behavior of H_c and α .

The thermal behavior of magnetic properties are controlled by the exchange coupling interaction between the α -FeSi grains and the intergranular amorphous phase, so researching the exchange coupling interaction of the α -FeSi grains should consider the affect of the magnetic properties of the α -FeSi grains, in addition to considering the affect of the amorphous phase. If the distance between α -FeSi grains is less than the exchange-coupled grains critical size L_{ex} , the magnetization from one grain to the other grains was carried out by penetrating through the grain boundary phase. It is important to maintain an efficient exchange coupling interaction between α -FeSi grains and the residual amorphous matrix, and it is necessary to sustain the higher Curie temperature for the application of the FeCuNbSiB ribbons in high temperature by controlling the size and crystallization degree of α -FeSi grains.

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