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# Influence of Addition of Al and Ti Solutes and Variable Processing Conditions on Mechanical and Electrical Properties of Cu-Cr Alloys

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**Abstract:** To apply the electric component with high efficiency, the softening problem of material should be improved. Cu-Cr alloys are recognized to be proper materials to be applied. However, the softening problem has not been solved yet. In this study, the effect of Ti and Al on mechanical property and electrical conductivity in Cu-Cr alloy was investigated. Cr content is designed to up to 0.25 wt.% in order to be expected to improve electrical conductivity. During fabrication processing, microstructure identification, Cr concentration, lattice parameter, and micro-hardness in copper matrix were measured. Then, aging condition was investigated. Resistance about over-aging is increased compared to Cu-Cr and Cu-Cr-Al alloy added to Ti. The hardness and electrical conductivity are discussed by the working conditions and heat treatment, such as rolling, solid-solution treatment, and aging process, which improve the formability also at the optimum condition. Moreover, the role of Ti contents is studied. In the Cu-Cr alloys, the addition of both Al and Ti contents keeps the hardness from being reduced. As a result, the precipitation of Cr particles and the obstacle by Al and Ti contents to be softened are observed in this study.

**Keywords:** Cu-Cr alloy; electrical conductivity; hardness; softening; processing condition; precipitation; formability; Al and Ti contents

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## 1. Introduction

In the electric industries, to apply the electrical parts of electrical appliance and vehicle, which are improving performance, such as electrical power transmission and electrical conductor, both high strength and good electrical conductivity should be achieved [1–8]. The certain required properties can be obtained by controlling the chemical composition in alloys [9,10]. Although there are requirements of electrical industry, these electrical and mechanical properties cannot be achieved generally. If the mechanical strength increases, the electrical conductivity decreases, such as trade-off relationship. Therefore, many researchers have studied to improve both strength and electrical conductivity widely [1–8].

To solve or minimize the problem of trade-off relationship of material, Cu alloys have been used and studied widely, for its high electrical conductivity. Figure 1 shows the variation of Cu alloys by their electrical and mechanical properties. In general, the pure Cu has high electrical conductivity, but low strength and bad resistance of softening at heating condition. Therefore, the Cu-Cr based alloys are recognized as the key to solve the problem. These alloys are widely used to high current connectors and integrated circuit lead frames, because of their outstanding mechanical and electrical properties [11–15]. The Cu-Cr alloys have high strength, because of the precipitation hardening effect [11–15], so the aging time and temperature are important factors to control precipitation. Moreover, the addition of other chemical composition can control the size and fraction of precipitation [16], such as Mg content. The addition of other chemical composition, to improve the softening resistance, is spotlighted. Likewise, controlling the chemical composition of Cu-Cr alloys

by addition of other composition can be useful to improve its mechanical and electrical properties.

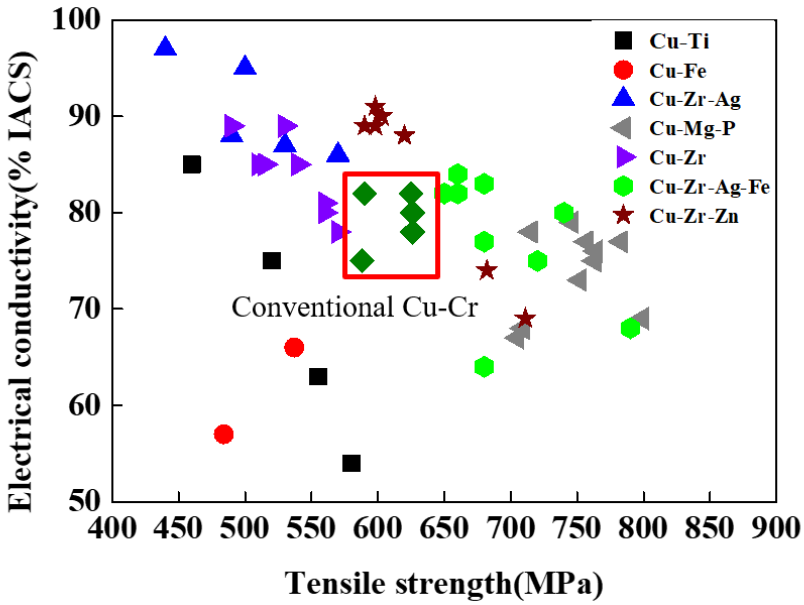



**Figure 1.** Conventionally used and recently developed Cu-Cr alloys and their properties.

Cu-Cr alloys exhibit precipitation hardening response, because the supersaturated Cr in the Cu-matrix creates a high degree of thermodynamic meta-stability. As a result, high strength is obtained by the distribution of nano-sized Cr precipitates in the Cu-matrix. In addition, their electrical conductivities are similar level with pure copper due to the low concentration of Cr in Cu matrix after aging [17]. Severe working environments such as downsizing and thinning are needed to couple high strength with high electrical conductivity. In order to obtain improved properties, there are a lot of investigations on the effects of alloying elements such as Fe, P, Zr, etc. However, Cu-Cr alloy containing Fe shows a small age hardening response compared to Cu-Cr alloy. Additionally, it is reported that P addition retards the growth of precipitates. Additionally, Zr attribute to low stacking fault energy and to improve softening resistance. However, mechanical property and electrical conductivity is similar to Cu-Cr alloy [18,19]. It is reported that Al and Ti can attribute to reducing the concentration of Cr in Cu matrix [20]. Additionally, it is expected to form Al3Ti phase in these quaternary alloy systems. Additionally, while P.C. Zhang's report is studied only about effect of Ti except to Al, addition of Al and Ti can be expected to improve mechanical properties [21,22]. In this study, the effect of Ti and Al on mechanical property and electrical conductivity in Cu-Cr alloy were investigated. Especially, Cr content is designed to up to 0.25 wt.%, which is lower than previous reported Cr content in order to be expected to improve electrical conductivity.

## 2. Materials and Methods

For this study, the experimental alloys with the compositions as shown in Table 1 were cast as 59 × 32 × 190 $mm^3$ plate structure with iron mold in a vacuum induction furnace (Ar atmosphere + degree of vacuum: 7.5 × $10^{-2}$). The cast alloy is scalped, solid solution treated at 1000 °C for 6 h in a NaCl salt bath to prevent oxidation and quickly quenched into water. They were cold-rolled to 80% reduction ratio in thickness. Cold-rolled strips were aged at 420, 450 and 480 °C for 1, 3 and 5 h in NaNO3 salt bath and then carried out final cold-rolled to a 99.5% reduction. Analysis and evaluations were performed with FactSage program (ThermFact Inc. & GTT-Technologies, Montreal, Canada), Spectrometer (Agilent, Santa Clara, CA, USA), SEM/EDS, XRD, Electron probe micro-analyzer (EPMA, AMETEK, Berwyn, PA, USA), Hardness, Electrical conductivity. Microstructure identification of

as-cast and solid solution treated specimens were observed by SEM/EDS (Quanta 200F FE-SEM, FEI, Hillsboro, OR, USA). Lattice parameter is measured by X-Ray Diffraction (D8 Discover, Bruker, Billerica, MA, USA). Measure condition is 20°~90° (2θ angle), 0.06°/s (scan speed) and 1.5406λ (wavelength). The mechanical properties for solid solution and aging treatment were evaluated by hardness test (FM–7E, FUTURE-TECH CORP., Kanagawa, Japan). Micro-hardness in matrix condition is 10 gf, 5 s, and hardness condition is 100 gf, 10 s. Electrical properties were measured by SMP-10 equipment (Fisher Scientific, Schwerte, Germany) after calibration with a pure Cu sample. Specimen for microstructure is polished by sandpaper (#100, #600, #1000, #2000, #2400 grade), 1 μm diamond suspension and 0.05 μm Silica suspension and etched by solution ($FeCl_3$ 5 g + HCl 20 mL + $H_2O$ 100 mL). The other specimen is polished by SiC paper (#100, #600, #1000, #2000, #2400 grade). Cr concentration using EPMA (SX-100, AMETEK, Berwyn, PA, USA) is conducted on 15 kV, 60 nA, and peak analysis is conducted on 7 points in 2 grains and averaged.

**Table 1.** List of chemical compositions of Cu-Cr alloys by using spectrometer.

| Wt.% | Cr | Al | Ti | Cu |
|---|---|---|---|---|
| Cu-0.25Cr | 0.2677 | 0.0011 | 0.0004 | Bal. |
| Cu-0.25Cr-0.06Al | 0.2655 | 0.0548 | <0.0004 | Bal. |
| Cu-0.25Cr-0.06Al-0.015Ti | 0.253 | 0.0565 | 0.0189 | Bal. |
| Cu-0.25Cr-0.06Al-0.03Ti | 0.2604 | 0.0577 | 0.0264 | Bal. |
| Cu-0.25Cr-0.06Al-0.05Ti | 0.2438 | 0.0620 | 0.0507 | Bal. |

## 3. Results and Discussions

*Experimental Results and Discussions*

Figure 2 shows the microstructures of Cu-Cr and Cu-Cr-x alloys, which were cast in this study. The dependence of alloying components about microstructure, such as the addition of Ti or Al, is not observed. However, the Cr particles are identified by SEM/EDS result. These particles are distributed to grain boundary. Moreover, the size of particles is less than 1 μm. The shape of particles is formed spherically, but not uniformly. The EDS result of Cr particle is examined by TEM analysis in Figure 3. The Cr solubility in Cu matrix decreases with temperature. Therefore, it seems that the excessive Cr atoms are precipitated from Cu matrix. The Cr-rich particles are formed, due to relative slow cooling rate during solidification. However, the solid solution treatment affected the microstructure and formed Cr particles of Cu-Cr alloys. This effect is shown as Figure 4. The solid solution treatment affects grain size higher. Moreover, the size and distribution of Cr particles change. Comparing with Figures 2 and 4, the contrast decreases. In other words, the distribution of particles decreases. It seems that the Cr particles are solutionized in Cu matrix.

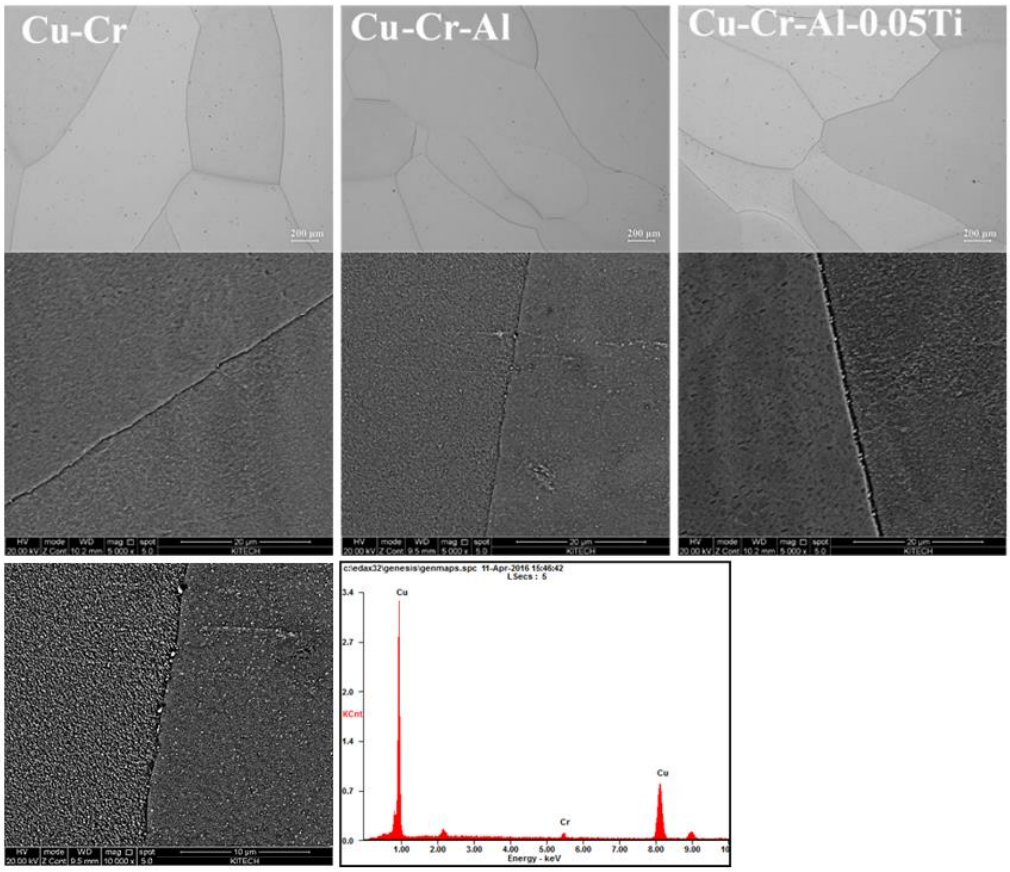



**Figure 2.** Microstructures of Cu-Cr alloys as cast by SEM.

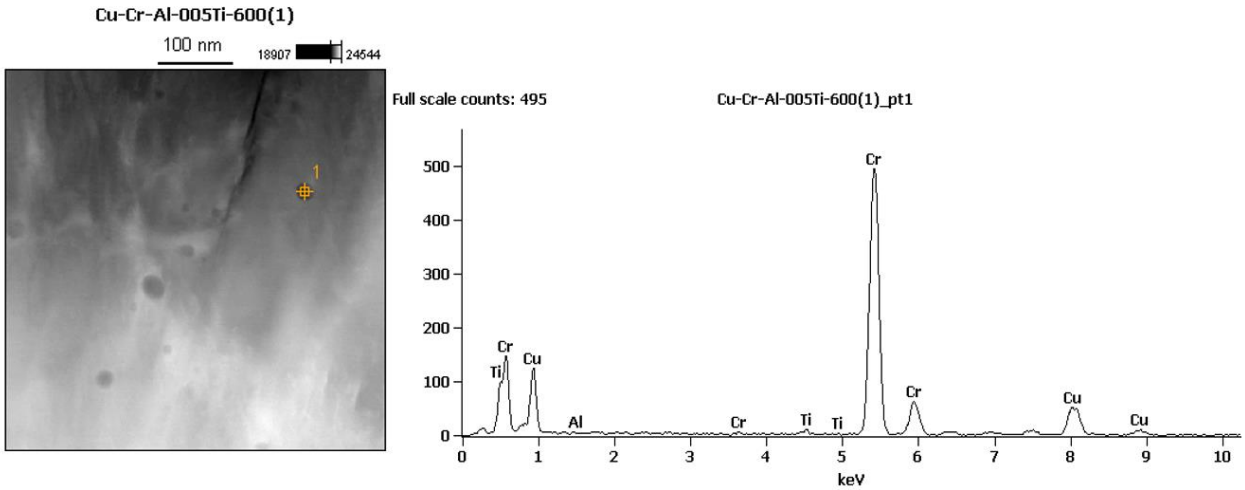



**Figure 3.** Observation and identification of precipitation of Cu-Cr alloys by TEM and EDS (the point indicating "1" means the point to be investigated by EDS).

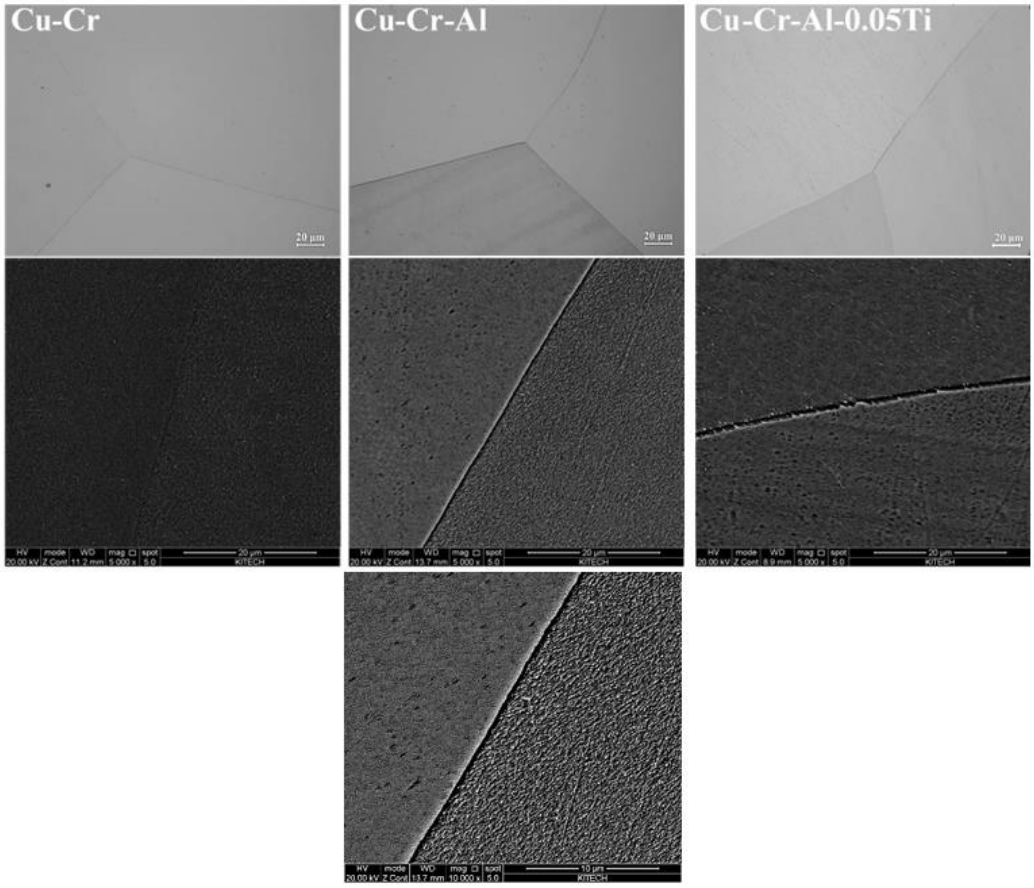



**Figure 4.** Microstructures of Cu-Cr alloys after solid solution treatment.

To observe the effect of solid solution treatment for mechanical and electrical properties of Cu-Cr alloys, the micro-hardness and electrical conductivity of alloys, which are cast and solid solution treated, are shown as Figure 5. At the as-cast state, the Cr particles are detected (Figure 2), so micro-hardness, lattice parameter and electrical conductivity are affected by the behavior of Cr particles. The precipitated Cr particles obtain the strength and hardness of material higher, following the precipitation hardening effect, in general [23]. At the as-cast state, because of the Cr particles and their distribution, the hardness is high, and the electrical conductivity is high. The hardness was detected by Rockwell hardness. At this state, the micro-hardness is low, and the lattice parameter is low. However, after solid solution treatment, the tendency becomes reversed. The solutionized Cr particles affect the increase in the lattice parameter, so that the stress field by precipitation decreases, but the stress field by solid atoms increases. Moreover, in case of Rockwell hardness, the decrease in hardness in solid solution treatment is seemed to be affected by the high temperature heat treatment and measurement of grain and grain boundary. The measurement of micro-hardness is examined at the grain and matrix, not particle existence. Therefore, the micro-hardness is caused by solutionizing effect more. However, macroscopically, the Rockwell hardness is caused by all microstructure changing effects, such as solutionizing and decrease in precipitation particles. Therefore, the hardness decreases, and electrical conductivity decreases also. As a result, although the locally employed Cr atoms show a solid solution strengthening effect, which is shown the micro-hardness change, overall, the solid solution strengthening effect does not seem to exceed the extinguishing the precipitation strengthening effect. Moreover, the electrical conductivity decreases, not trade-off. After solid solution treatment, the solutionized Cr atoms seems to hinder the electron path.

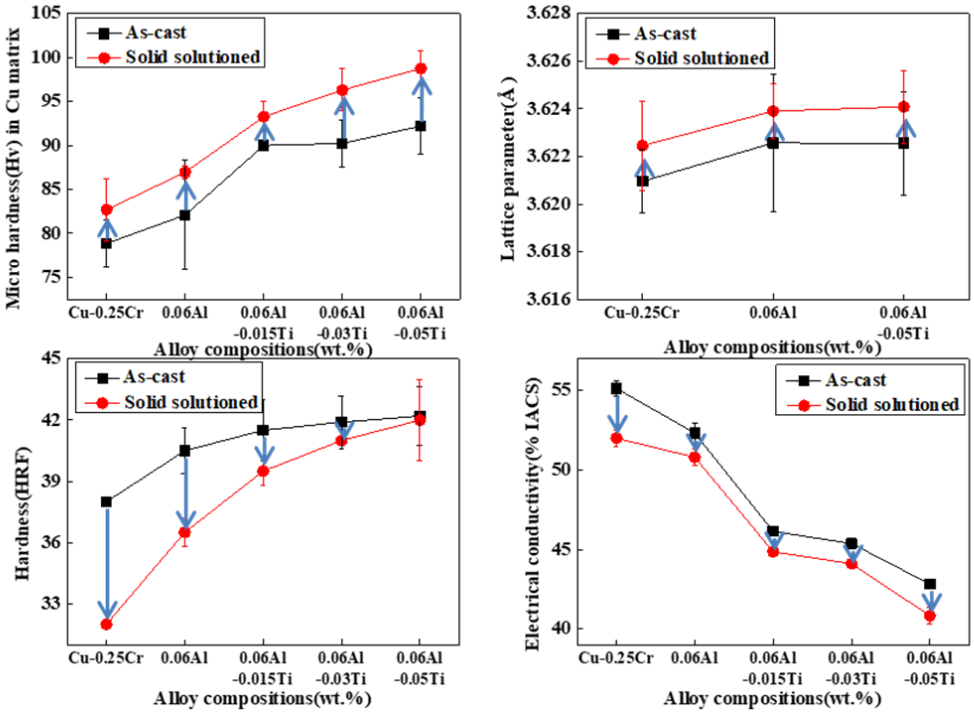



**Figure 5.** Variation of properties of Cu-Cr alloys as-cast and solid solution treatment.

In this result, the Al and Ti contents reduce the electrical conductivity, but these contents keep the hardness from high decrease. Additionally, other solid solution effect is minimized by the addition of Al and Ti contents. The addition of Al and Ti contents keeps the mechanical properties from being poor, but the solid effect to reduce the electrical conductivity cannot be controlled.

Figure 6 shows the electrical conductivity and lattice parameter and hardness of solid solution treated and cold-rolled Cu-Cr alloys. The cold-rolled specimen with 80% reduction ratio (from 30 to 6 mm) has higher value of hardness, but the lattice parameter and electrical conductivity value remain similar. The electrical conductivity is little lowered than solid solutionized state. It seems that the work hardening effect increases the hardness of the cold-rolled state. The formed dislocations by rolling process strengthen the Cu-Cr alloys. However, the lattice parameter is not affected by formation of dislocations. It seems that the dislocation density is not the main factor to decrease electrical property than Cr particles, which are precipitated. In these Cu-Cr alloys, the Cr particles are the main factor to control the electrical conductivity.

Figure 7 shows the variation of electrical conductivity of Cu-Cr alloys after aging treatment. As the aging time is longer, the electrical conductivity value gets higher, in all Cu-Cr alloys. Moreover, the electrical conductivity increases, as aging temperature increases. Especially, as the aging temperature increases, the time to reach the peak point decreases. However, the hierarchy of each alloys is maintained. The alloy, which has a higher content of alloying component, such as Al and Ti, has a lower value of electrical conductivity value. After 3 h of aging time, the electrical conductivity is maintained to be the highest value. In this result, it can be figured out that the precipitation rate and amount are the main factors to control the electrical conductivity; for the high aging, time and temperature make the electrical conductivity high.

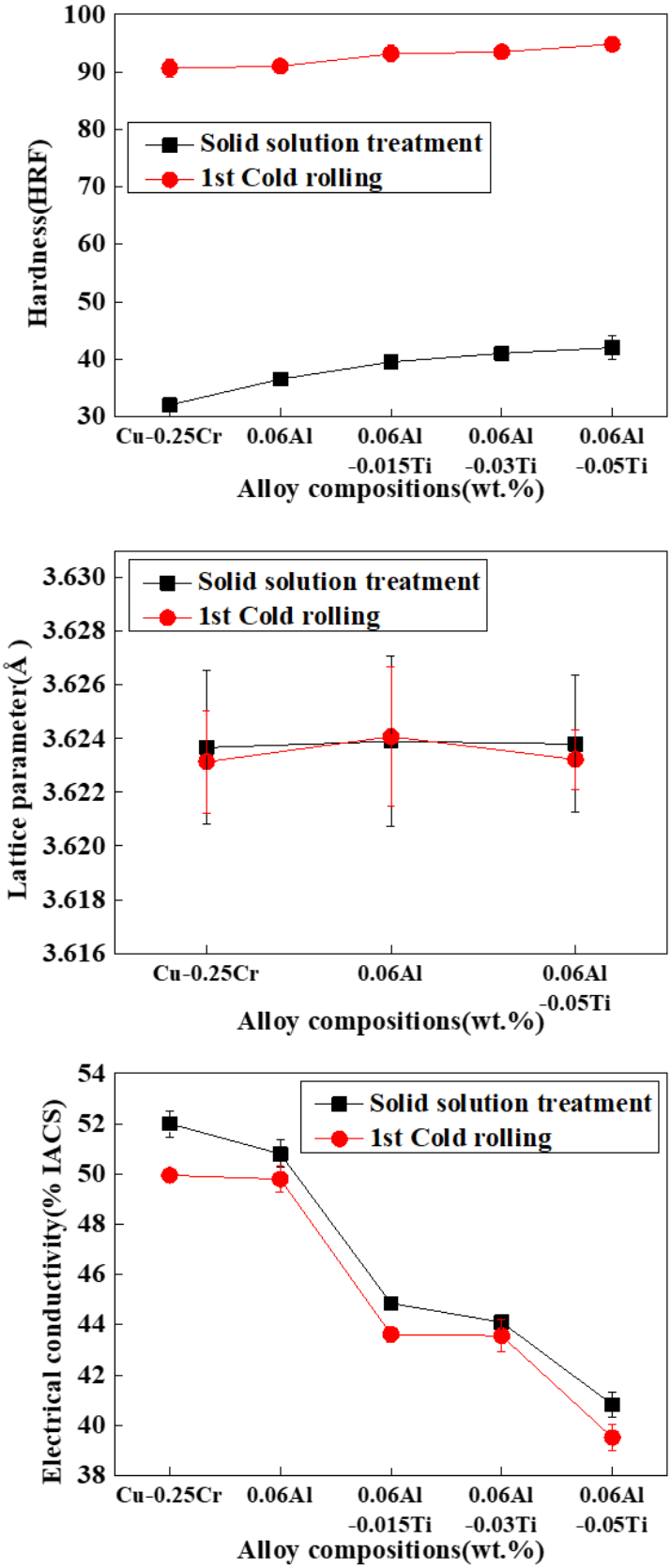



**Figure 6.** Electrical and mechanical properties and lattice parameter variation after 1st cold-rolled Cu-Cr alloys.

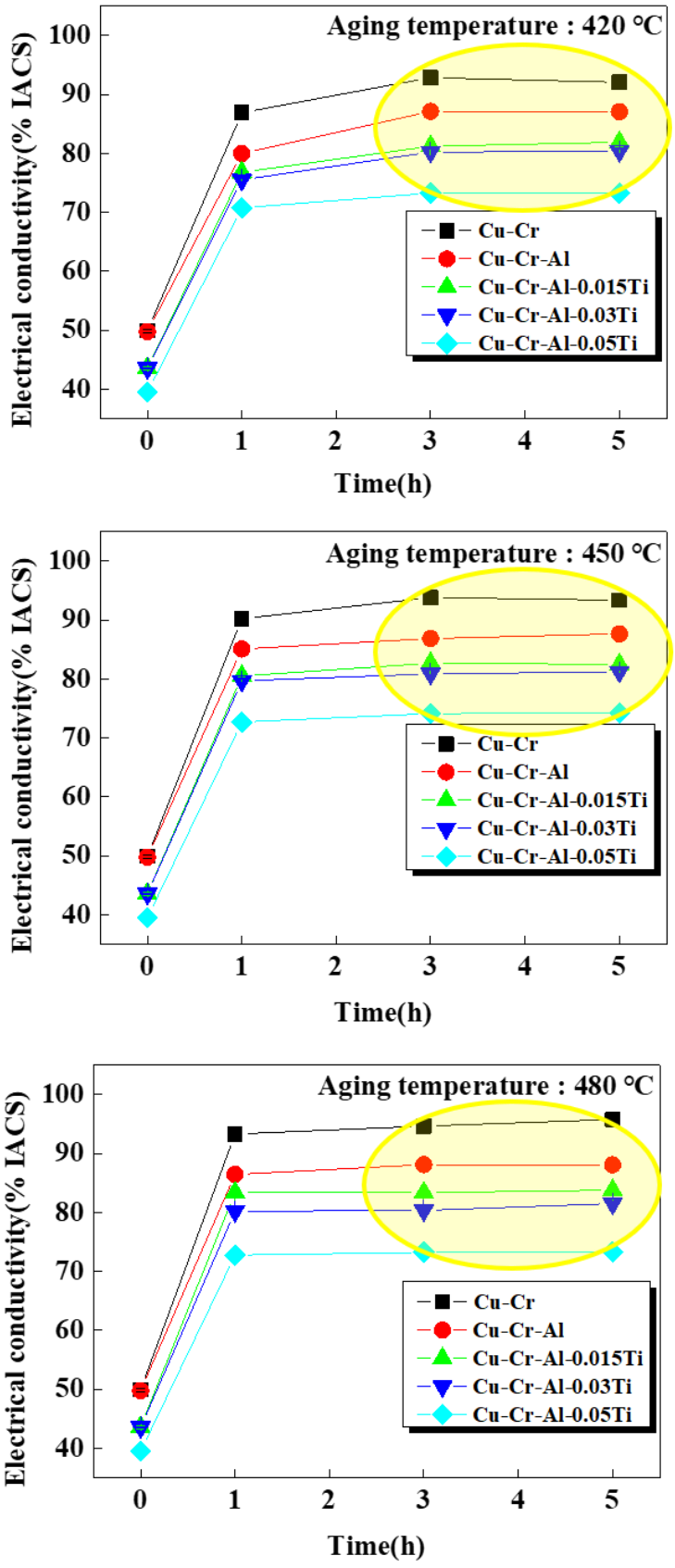



**Figure 7.** Variation of electrical conductivity of Cu-Cr alloys after aging treatment.

However, the hardness tendency by aging time and temperature is different. Figure 8 shows that the decrease in hardness of Cu-Cr alloys is observed at 480 °C of aging temperature. Furthermore, in Cu-Cr and Cu-Cr-Al alloys, the decreased rate of hardness is sharp at the aging temperature of 480 °C, then the Cu-Cr-Al alloys added the Ti content. It seemed that Cr precipitates are coarsened by over-aging. The decrease amount of hardness value in Cu-Cr-Al-Ti alloys is little; whether the Ti content is small or high. Even at 420 °C of aging temperature, the hardness value of Cu-Cr-Al-0.05Ti increases. As a result, when the results of Figures 7 and 8 are discussed, the Cr precipitates size and amounts improve the electrical conductivity, but the coarsened Cr precipitates reduce the hardness of material, so that the softening effect occurs.

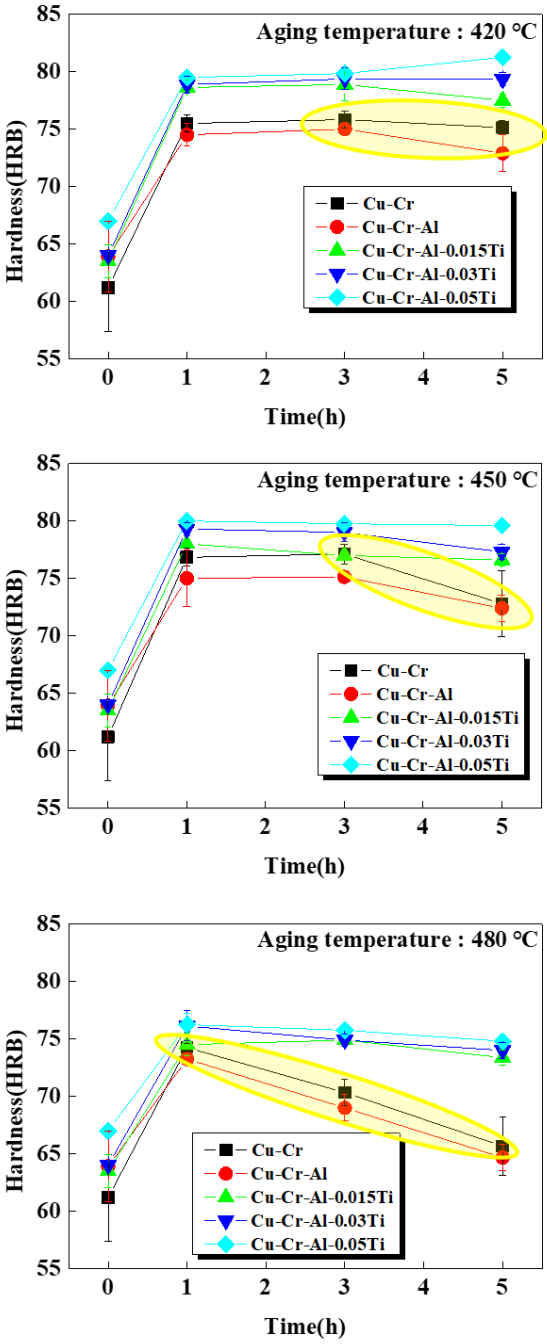



**Figure 8.** Variation of hardness of Cu-Cr alloys after aging treatment.

The tendency of the processes of all Cu-Cr alloys, which are investigated in this study, is shown as Figure 9. In this Figure 9, the dramatic increase in hardness is shown, while the first cold-rolling process (80% reduction ratio) is conducted. The work hardening effect is more powerful to make hardness higher than precipitation hardening effect. However, at the viewpoint of electrical conductivity, the precipitation effect is stronger than other effects, such as the solid solution effect and work hardening. Though the electrical conductivity of Cu alloys decreases by solid solution hardening and work hardening effect, the increase in conductivity by precipitation effect is more powerful, and the increment of electrical conductivity value is much larger at the aging treatment. In these alloys, the dislocation formation and its density are main factors to make the hardness high, but the electrical conductivity is highly affected by precipitation proportion and rate. As a result, both of the electrical and mechanical properties can be adopted by controlling the processes, such as rolling and aging condition.

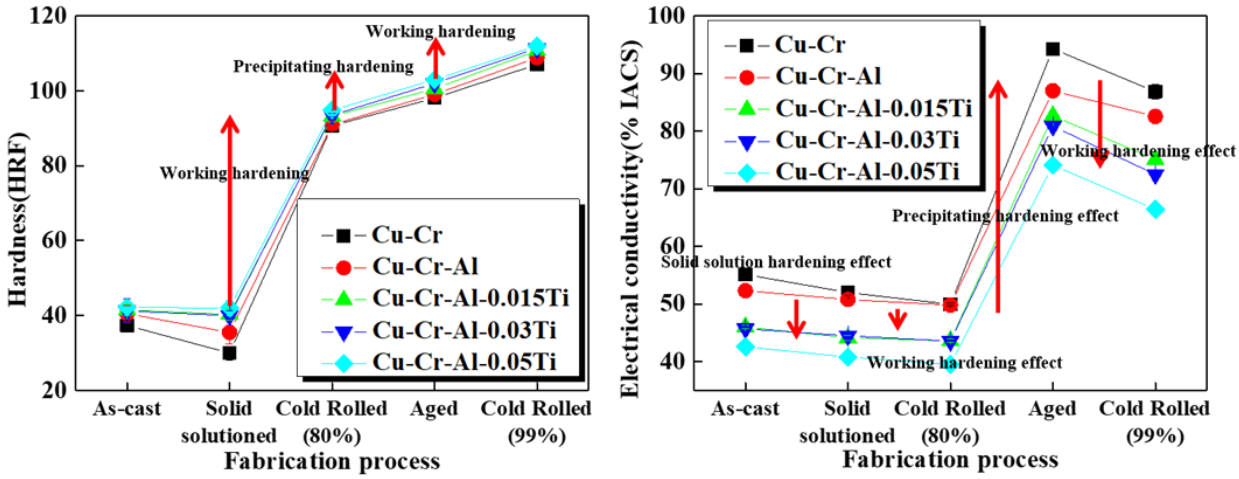



**Figure 9.** Variation of mechanical and electrical properties of Cu-Cr alloys.

Figure 10 indicates the variation of resistance to softening effect by the alloy's components. The Cu alloys with higher other alloying components have higher value of micro hardness, in general, because of the solid solution and precipitation hardening effects. However, as the annealing temperature increases at the annealing processes, after second rolling process (99% reduction ratio), the hardness decreases, and softening occurs. The decrease in hardness of Cu-0.25Cr and Cu-0.25Cr-0.06Al alloys occurs linearly. However, the value of hardness of Cu-0.25Cr-0.06Al with Ti contents alloys is maintained to 400 °C of the annealing temperature and decreases from that temperature to 600 °C. In other words, the addition of both Al and Ti contents is important to make the maintenance of hardness at the annealing process (~400 °C).

The prediction of solubility and crystallization phase of Cu-Cr alloys by FactSage is expressed as Figure 11. In Figure 11, the change in solvus line is insignificant, depending on the increase in Al and Ti contents. Moreover, the crystallization phase cannot be found, except the formation of Cu4Ti at 310 and 330 °C. These results can help to understand the behavior of alloying elements and improving resistance to softening effect.

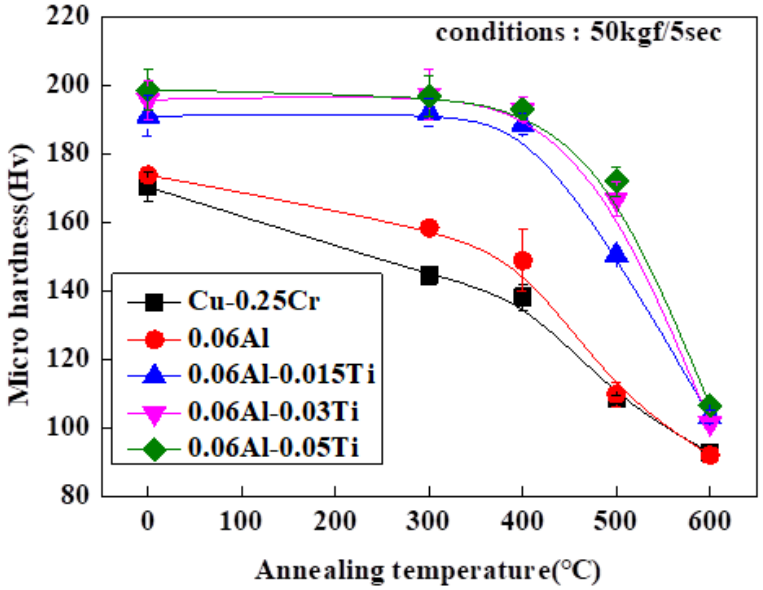



**Figure 10.** Variation of resistance to softening characteristics of Cu-Cr alloys.

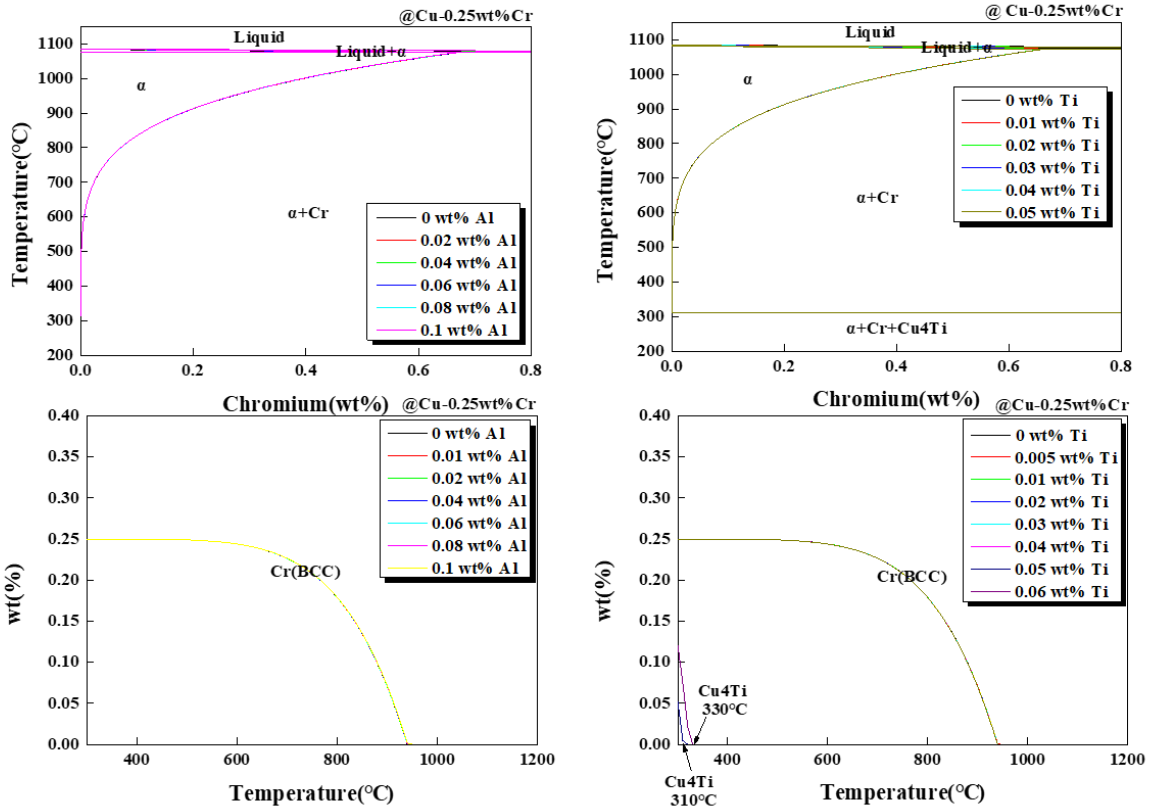



**Figure 11.** Prediction of solubility and crystallization phase of Cu-Cr alloys by FactSage®.

To compare Figure 11, the solute concentration variation in Cu matrix is detected by EPMA analysis (Figure 12). Figure 12 represents the concentration of alloying elements at all processing conditions. In this result, the amount of Al and Ti in matrix does not change in all process. In other words, at all processes, the loss or addition of certain elements is not shown, except the Cr amount increase after solid solution treatment. This tendency does not change, whether the chemical composition changes, such as the Ti contents

increase. This result is supported by the FactSage result, which is shown in Figure 11. Figure 13 also supports these results (Figures 11 and 12). In the Cu-Cr-Al-Ti alloys, at the heat treatment, the Cr particle size is examined as 5.24 nm, smaller than 8.41 nm of the Cu-Cr alloy. Moreover, the displacing in the diffraction pattern of Cu-Cr alloy (0.19) is smaller than one (0.21) of the Cu-Cr-Al-Ti alloy. This result indicates that the addition of Al and Ti in Cu-Cr alloy makes the lattice parameter lower. The addition of Al and Ti elements seems to affect the precipitation and release of strain by observing the diffraction pattern changes. In other words, the loss or addition of the Cr amount is the evidence of the precipitation amount.

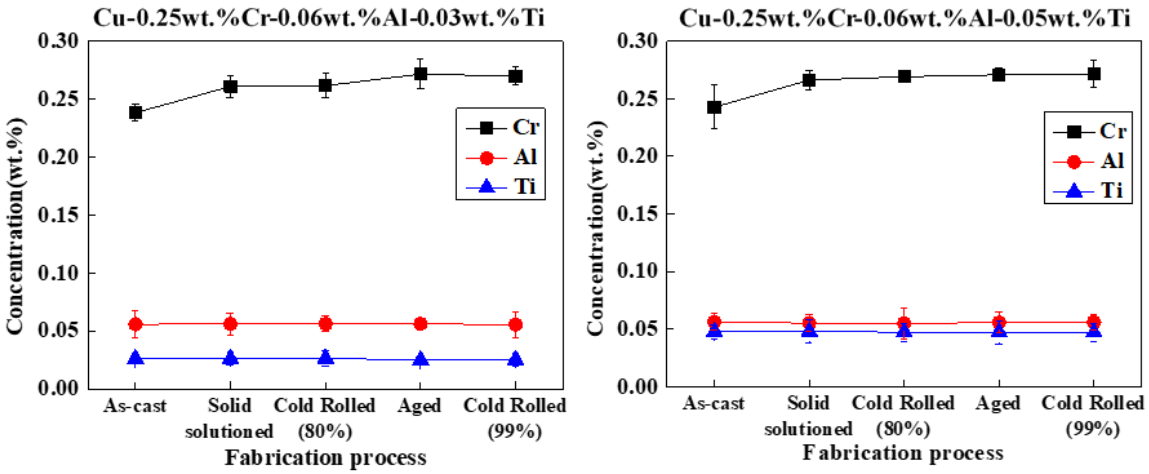



**Figure 12.** Solute concentration variation of Cu matrix by EPMA analysis.

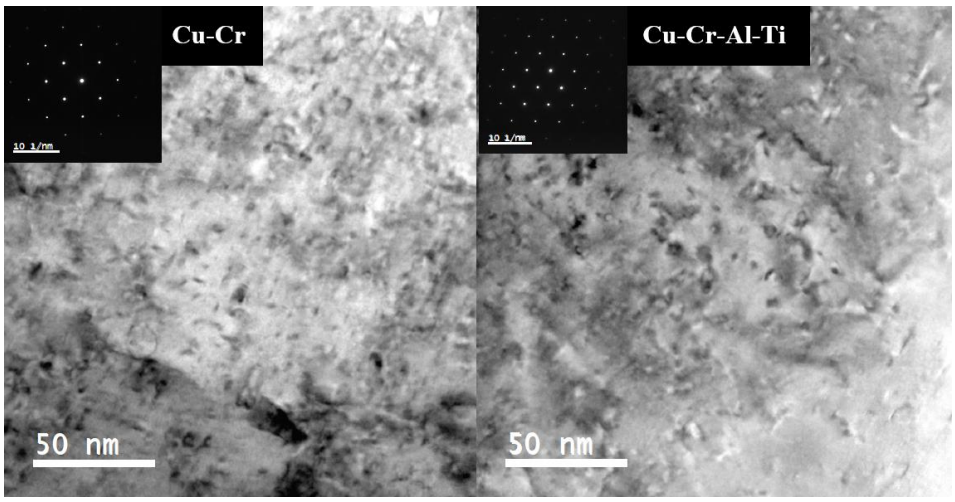



**Figure 13.** TEM images and diffraction pattern of Cu-Cr and Cu-Cr-Al-Ti alloys after solid solution heat treatment.

To visualize the distribution of Cr particles, TEM analysis was conducted. Then, this result is shown as Figure 14. The black region of TEM images indicates the Cr particles. The Cu-Cr alloy added to the Al and Ti contents has a lot of nano-sized Cr particles (<10 nm), and the particles are randomly distributed. It seems that the addition of Ti contents on Cu-Cr-Al alloys affects to block the release of strains, so that the softening effect is not efficient at aging process (Figure 7). However, the further research should be processed more to confirm the Ti addition effect on Cu-Cr alloys.

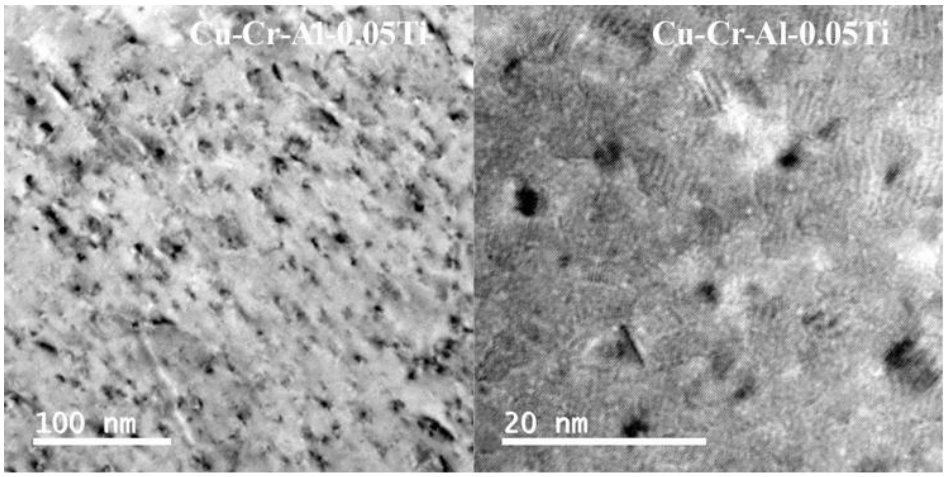



**Figure 14.** Observation and identification of precipitation of Cu-Cr alloys by TEM.

In this study, the alloying components and its amount effects are discussed in Cu-Cr alloys. Then, the processing condition effects on electrical and mechanical properties are discussed. In all processing conditions, the solid solution treatment, cold-rolling, aging processes increase the hardness by their strengthening mechanisms, such as work-hardening, solid solution-hardening, and precipitation-hardening effects. However, in these conditions and strengthening mechanisms, the precipitation hardening effect increases the electrical conductivity, only. The Cr particles have a role to improve both mechanical and electrical properties. Even, the softening occurs at the annealing process. In this process, the Ti alloying component acts as the obstacle to be softened to maintained the lattice parameter. These results can help the electrical industries needing to apply high-efficient electric components, which should have both the high strength and electrical conductivity.

## 4. Conclusions

In this study, Cu-Cr alloy with Al and Ti is investigated by microstructure identification, electrical and mechanical properties, and lattice parameter.

(1) Solid solution treatment is proper at 1000 °C for 6 h through analysis such as SEM/EDS microstructure identification, Cr concentration through EMPA, and variation of lattice parameter and micro-hardness.

(2) The optimizing aging condition differs for each alloy. Cu-Cr, Cu-Cr-Al, Cu-Cr-Al-Ti (0.015,0.03, and 0.05 wt.%) alloy is, respectively, 450 °C for 3 h, 450 °C for 2 h, 420 °C for 3 h, 420 °C for 5 h, and aging condition at 480 °C more than 1 h is over-aged.

(3) Cu-Cr alloy with Al observe a slight increase in mechanical properties, but electrical properties also decrease fairly. Cu-Cr-Al alloys with Ti observe a proper increase in mechanical properties and a severe decrease in electrical properties. Cu-Cr-Al alloys with Ti are shown resistance to over-aging compared to Cu-Cr and Cu-Cr-Al alloy.

**Author Contributions:** Conceptualization, C.-H.C. and H.C.; methodology, C.-H.C. and H.C.; validation, C.-H.C., J.S. and D.K.; investigation, C.-H.C., J.S. and D.K.; resources, C.-H.C., J.S., D.K. and H.C.; writing—original draft preparation, C.-H.C.; writing—review and editing, H.C.; visualization, C.-H.C.; supervision, H.C.; project administration, H.C. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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