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Heat transfer from a chemically oxidized or anodized copper plate to liquid helium

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Abstract

Heat transfer characteristics between a copper plate (50 mm × 10 mm) and liquid helium have been measured for two kinds of surfaces, i.e. the surfaces were chemically oxidized and anodized. The experimental results showed the critical heat fluxes, q_{CHF}, do not change between the different oxidization samples. But the minimum heat fluxes, q_{MHF}, have a variation, which may have been caused from the growth of other components than copper oxide on surface. The formation of matrix –COOH in chemically oxidized sample became clear by X-ray photo-electron spectroscopy (XPS), and a different compactness on surface was indicated by scanning electron microscopy (SEM).

Keywords: Heat transfer characteristics; Chemical oxidization; Anodization; Heat flux

1. Introduction

HT-7U, a superconducting Tokamak, is under construction at the Institute of Plasma Physics (IPP), Chinese Academy of Sciences (CAS), supported by the National Development Planning Commission of China. The toroidal field (TF) coils of the superconducting Tokamak once have been designed as the pool-cooled superconducting magnets. It is well known [1,2] that for the cryogenic stability of the magnets, the heat transfer characteristic from the surface of superconductors to liquid helium plays a very important role. Many methods have been attempted to improve the heat transfer between these surfaces and liquid helium. For example, during the development of helical coils of the Large Helical Device (LHD) at the National Institute for Fusion Science (NIFS), Japan, and the Large Coil Task (LCT) at Japan Atomic Energy Research Institute, the surfaces of the magnets for LHD or LCT were treated by roughening or oxidization to improve the heat transfer performance [3–5]. Because the experimental results indicated the heat transfer in the liquid helium for a wide oxidized copper plate was significantly improved, the roughing technology for the LCT and the oxidized surface for LHD were then successfully used during the superconducting coil fabrication process, respectively [6,7]. Iwamoto and his co-workers noted in papers [8,9] the samples were prepared by chemical oxidization. It is known that the method of anode oxidization is a convenient and easily controlling process, especially

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for large coils. However, the heat transfer characteristics of anodized copper samples are unknown. In this paper, we investigate the heat transfer characteristics of a copper surface treated by chemical oxidization or by anodization to liquid helium, and analyze the surface structure of the two kinds of samples to clarify the relationship between surface structure and heat flux.

2. Experimental

The dimensions of used copper plate are 50 mm in length, 10 mm in width and 4 mm in thickness (schematically shown in Fig. 1) to simulate large scale superconductors. Before oxidizing, the sample was cleaned ultrasonically in acetone to ablate the oil and dust from surfaces, and then put horizontally in the chemical/electrolytic solution to oxidize/anodize the surface. The whole processing is performed in a bath heated by constant temperature water with ±1 °C error. During the chemical oxidization process the samples were just immersed in the solution. But in the anodization process, the direct current (DC) power was supported in $\approx 0.5\ \mathrm{dA/cm^2}$ current density. A stainless steel plate was used as cathode and a sample was the anode. After oxidizing, samples were rinsed using pure water and then dried. To measure the heat transfer characteristics, the experiments were carried out in a glass Dewar with a viewing window to observe visually the condition of the heat transfer surface and the behavior of helium vapor. During the measurement, the pressure in the glass Dewar was constant at one atmospheric pressure. In experiments, the whole sample body was sealed with a fiber reinforced plastic (FRP) holder except for the heat transfer surface. A Fe–Ni foil as a heater was attached on the rear side of the copper plate to heat it uniformly. The temperature difference between the surface and the liquid helium was measured by AuFe–Cr thermocouples. Five pair of thermocouples was mounted under the heat transfer surface with 1 mm in depth to confirm a uniform surface temperature. It was confirmed that the thermal leak from FRP to liquid helium was small and neglectable. In comparison, the heat transfer from polished copper surface was measured, too. X-ray photoelectron spectroscopy (XPS) spectra were obtained by using a monochromatic hybrid mode Al Kα. The C1s and Cu2p spectrums were decomposed by fitting a Gaussian–Lorentzian mixture function to the experimental curves.

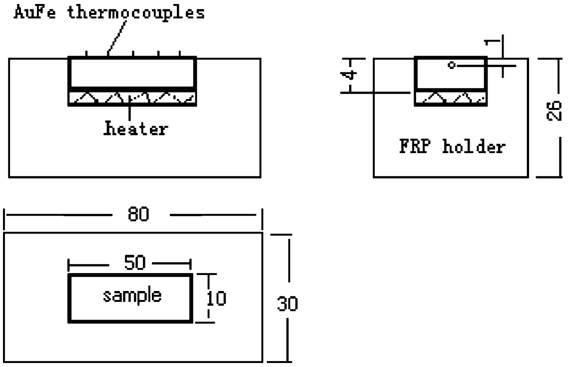

Fig. 1. Schematic of the sample and the FRP holder (unit in mm).

3. Results and discussion

3.1. Heat flux

Fig. 2 shows the influence of the orientation on the heat fluxes for chemically oxidized surface. It notices that the critical heat flux q_{CHF} was increasing with increasing the oriented angle, but decreasing at ≈40°. The maximum q_{CHF} appeared in 30° degree where the homo-, heterogeneous nucleate boiling all were ignited completely and the bubbles in the surface escape quickly from the surface.

The dependence of critical heat flux (denoted by q_{CHF}) and minimum heat flux (q_{MHF}) on the heat transfer surface treated by chemical oxidization and anodization is shown in Fig. 3. It results that the critical heat flux q_{CHF} increased by 32.3% for chemically oxidized sample and 32.7% for anodized sample, respectively. For minimum heat flux q_{MHF}, moreover, the data was improving greatly. It increased 134.2% for chemically oxidized sample and 145.7% for anodized sample in comparison with a polished copper. Comparing with Iwamoto's results in [8,9], the increase rates of q_{CHF}, q_{MHF} are in the same level as they showed but not q_{CHF}

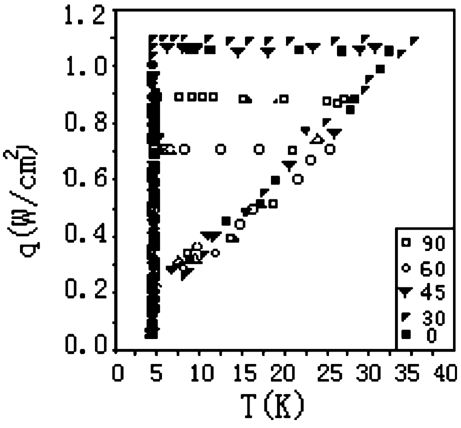

Fig. 2. Relationship of the heat fluxes with the orientation of chemically oxidized samples (the data were the average of five samples).

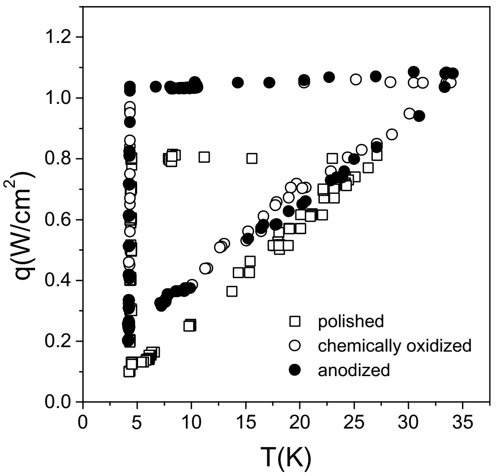

Fig. 3. Heat flux versus surface temperature for samples with different surface treatment (in horizontal orientation (0°), the data were the average of five samples).

or q_{MHF} which is quite difference due to the large size of samples in their experiments. It gets the same conclusion as Iwamoto [8,9] that the copper surface oxidized can remarkably increase the heat transfer characteristic, q_{CHF}, q_{MHF}. Besides that, the chemical oxidization and anodization have the similar efficiency to improve the interface heat transfer characteristics: the q_{CHF} shows a relatively small variation for two kinds of samples but the q_{MHF} has a big difference. It is not clear why are different q_{MHF} but the assumedly reason might be from the surface morphology and chemical compounds which were investigated as following.

3.2. Surface morphology and chemical compounds

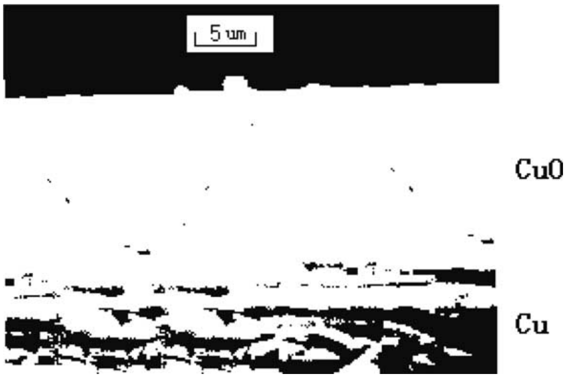

Fig. 4. SEM cross-section image of the chemical oxidization sample.

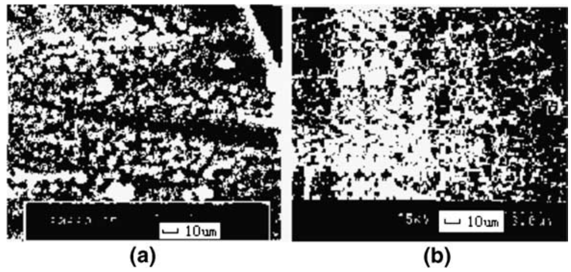

Fig. 5. SEM image of sample surface treated by (a)—chemical oxidization and (b)—anodization.

The sample surface morphologies are shown in Figs. 4 and 5. The cross-section image of the scanning electron microscope (SEM) of the chemically oxidized sample in Fig. 4 indicates clearly that the copper oxide on the surface is ≈15 μm in thickness after 10 min of chemical reaction time, and shall be responsible for the increase of the heat transfer.

From Fig. 5 it finds that that the chemically oxidized surface was apparently loose and rough by more aggregated clusters on surface but the anodized surface was relatively compact and smooth. This may result from the reaction rate occurring in the oxidization processing because the chemical oxidization reaction was faster than the anodization process: to achieve the same thickness of copper oxide, 15 μm for instance, the chemical oxidization reaction needed ≈10 min, but the anodization reaction would need 30 min. It is known that the rough surface will improve the heat transfer characteristics, but Fig. 3 , relationship of heat flux with the surface temperature on different surface treatment samples, did not reveal that.

By XPS, the surface components of oxidized samples were analyzed. Fig. 6(a) and (c) revealed that there were four kinds of elements O, C, Cu and H in both surfaces, and compounds on the surfaces mainly consisted of Cu, $Cu^{+}$and Cu^{2+} as shown in Fig. 6(b) and (d).

Deep investigation of the surface component in Fig. 7 shows the C1s spectra at 285.83 eV assigned to C–H, C–O groups, the possible compounds HCO_3^{-1} formed on the both surfaces of chemical oxidization and anodization samples. However, bond energy at 288.5 eV assignment to –C=O, O–C–O groups, i.e. the matrix of –COOH radicals only appeared obviously in the surface of chemically oxidized sample. It may be one of the possible reasons causing the minimum heat flux q_{MHF} in chemical oxidization sample lower than that in the anodization one. The –COOH group is assumedly formed from post-reaction: the copper oxide film deposited quickly and grown loosely in chemical oxidization process will incorporate H_2O and CO_2 from air and formed matrix of –COOH in the surface, which influence heat transfer characteristic, q_{MHF}.

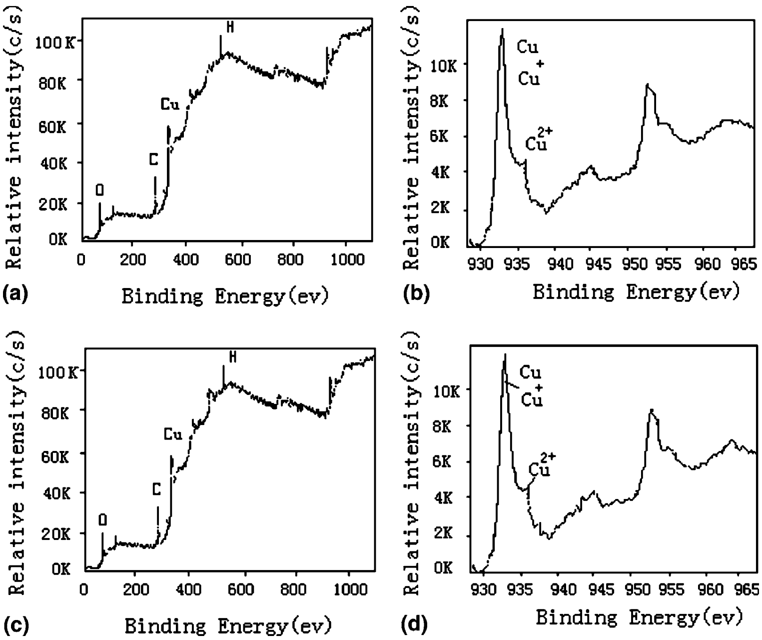

Fig. 6. XPS spectra of the sample surface treated by (a), (b)—chemically oxidization and (c), (d)—anodization.

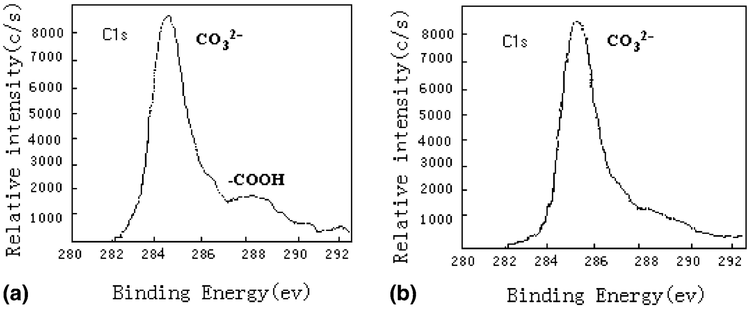

Fig. 7. C1s XPS spectra of the sample surfaces treated by (a)—chemical oxidization and (b)—anodization.

The critical heat fluxes q_{CHF} for both kinds of samples, however, was not affected by the surface compounds. This can be explained by the mechanism of liquid helium nucleate boiling igniting by homo- and heterogeneous nucleate boiling. The possible reason is that the size of pores on solid surface was still smaller than that of sites for homogeneous nucleate boiling and cannot initiate liquid helium boiling.

4. Conclusion

Copper sample surfaces were oxidized by two kinds of methods: chemical oxidization and anodization. The heat fluxes from the heated copper surfaces to liquid helium were measured. Meanwhile SEM and XPS were employed to analyze the surface structure. This led to the following results:

The surface copper oxide, growth from chemical oxidization or anodization, can remarkably increase the heat transfer characteristics, q_{CHF}, q_{MHF}. So in order to improve the stability of superconducting coils in liquid helium, the surface of conductor should be treated by oxidization.

It is found that the minimum heat fluxes, q_{MHF}, have a variation between the chemical oxidization and the anodization samples. It may be caused by the different components grown on the surface, possibly due to –COOH in chemically oxidized surfaces.

The critical heat fluxes, q_{CHF}, were approximately the same for two kinds of samples though the surface of chemically oxidized samples was relatively rough. The possible reason was that the sizes of pores formed in the loose oxidization layer were still smaller than the sites for homogeneous nucleation boiling, and cannot initiate liquid helium boiling on surfaces.

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