

RESEARCH ARTICLE | JUNE 12 2012

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AIP Conf. Proc. 1435, 133–139 (2012)

<https://doi.org/10.1063/1.4712089>



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EFFECT OF CRYOGENIC TREATMENT ON THERMAL CONDUCTIVITY PROPERTIES OF COPPER

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ABSTRACT

Copper exhibits high thermal conductivity properties and hence it is extensively used in cryogenic applications like cold fingers, heat exchangers, etc. During the realization of such components, copper undergoes various machining operations from the raw material stage to the final component. During these machining processes, stresses are induced within the metal resulting in internal stresses, strains and dislocations. These effects build up resistance paths for the heat carriers which transfer heat from one location to the other. This in turn, results in reduction of thermal conductivity of the conducting metal and as a result the developed component will not perform as per expectations. In the process of cryogenic treatment, the metal samples are exposed to cryogenic temperature for extended duration of time for 24 hours and later tempered. During this process, the internal stresses and strains are reduced with refinement of the atomic structure. These effects are expected to favourably improve thermal conductivity properties of the metal. In this experimental work, OFHC copper samples were cryotreated for 24 hours at 98 K and part of them were tempered at 423K for one hour. Significant enhancement of thermal conductivity values were observed after cryotreating and tempering the copper samples.

KEYWORDS: Cryogenic treatment, Thermal conductivity, Residual stress, OFHC copper

INTRODUCTION

Cryogenic treatment

The practice of cryogenic treatment on metals has been extensively employed for many years mainly for improving life of cutting tools. In the recent years, this practice is being used to improve dimensional stability of machined parts and gauges, removal of residual stresses, enhancement of mechanical and physical properties, etc.[1] The cryotreatment process consists of mainly three phases where the test samples are gradually cooled to cryogenic temperature, held for extended length of time and gradually warmed to room temperature. A typical cryotreatment cycle takes around 38 hours with 5 hours of cooling to cryogenic temperature, 24 hours of soaking and 9 hours of warming to room temperature. Liquid nitrogen is popularly used as the cooling medium. Cryogenic treatment is generally followed by tempering processes to further enhance the changed properties which are produced during cryotreatment. A typical cryotreatment cycle is show in figure 1.

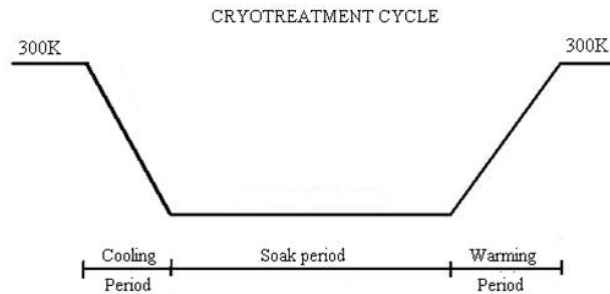


FIGURE 1. Cryotreatment cycle

Thermal conductivity of Copper

Oxygen free high conductivity (OFHC) copper is extensively used in many of the cryogenic applications. This is produced by the direct conversion of selected refined cathodes under carefully controlled conditions to prevent any contamination of the pure oxygen-free metal during processing with copper content in the range of 99.996% with oxygen less than 10 ppm. With negligible content of impurities, elemental copper exhibits high inherent properties. Characteristics are high ductility, high electrical and thermal conductivity and high impact strength. At room temperature, it has the thermal conductivity of 395 W/mK. Copper has the face centered cubic (FCC) lattice structure with coordination number of 12.

The thermal conductivity (K) is an intrinsic property of a material and depends on its properties like density, specific heat, average particle velocity and its mean free path. The value of thermal conductivity varies with temperature as the above properties are temperature dependent. For heat conduction in solids, thermal conductivity is expressed by the equation (1).

$$K = (1/3) \rho c_v v \lambda \quad (1)$$

where ρ is the density of the material, c_v is the specific heat at constant volume, v the average particle velocity and λ the mean free path of the particles.

Heat is transported in metals by both electronic motion and phonon motion. For temperatures above liquid nitrogen, contribution of electronic motion is more significant

for the heat transfer compared with the phonon motion. The electrons are scattered within the metal by the phonons and impurities, dislocations, strains and lattice defects. As the temperature is decreased, the scattering of electrons by surrounding phonons decreases and hence the thermal conductivity starts rising. This increase is continuous till the mean free path of electrons is restricted by dislocations and strains within the metal. Thus, reducing such resistances like impurities, dislocations, stresses and strains contribute favorably to enhance the thermal conductivity properties of the metal. [2,3]

OFHC copper is extremely pure and exhibits high thermal conductivity properties. The nominal pure copper is expected to possess good atomic structure with minimum internal stresses and strains and with minimum crystal defects. As it is subjected to any work hardening process, the internal stresses are developed resulting in dislocated atomic structure with lattice defects. These combined effects will influence the scattering effect and result in reduction in thermal conductivity at very low temperatures. In the experimental work carried by G.K.White, the thermal conductivity values of work hardened copper wire could be improved substantially by annealing in vacuum furnace for 3 hours at 823K. In this study, the thermal conductivity values were experimentally determined over the entire temperature range from room temperature down to deep cryogenic temperatures. The study indicated the possibility of improving the thermal conductivity properties of a metal by reducing the internal stresses (and strains) by proper heat treatment process like annealing or tempering. [4]

The lattice structure of newly procured metal will be not in the natural condition and possesses distorted lattice structure within. When this metal undergoes machining, more stresses and strains are induced resulting in further distortion. This results in dislocation in the atomic structure and adds to the scattering effect. When the metal is exposed to cryogenic temperature during the cryotreatment process, the strength of the atomic bonds starts to diminish due to very low internal kinetic energy. During this state of zero motion molecular state of mass, there is thermal compression. As the temperature is slowly increased to room temperature, thermal expansion takes place resulting in release of stresses and hence strains. This combined effect reduces the scattering effect and hence the thermal conductivity is expected to increase. [5]

The metals subjected to cryotreatment are normally tempered at moderate temperatures for a few hours. During this process, the internal stresses are further reduced resulting in still decreased scattering effect. The individual and combined effects of cryotreatment and tempering are expected to enhance the thermal conductivity properties of copper. [6,7]

EXPERIMENTAL PROCEDURE

In this experimental study, 12 samples were machined out of a single OFHC copper rod with each sample measuring 8 mm in diameter and 4 mm in thickness. The samples were properly polished to ensure that the top and bottom surfaces are flat and parallel. This condition is essential so that the heat flux passes through the sample uniformly from the hot surface to the cold surface.

A cryotreatment system has been specially designed and developed to cryotreat the samples. This system incorporates a unique technical feature of cryotreating the samples with circulating cold nitrogen gas. This avoids direct contact of liquid nitrogen with the samples and hence effects of thermal shocks are avoided. The control of the temperature of the system is carried out using a programmable PID controller which switches the solenoid valve on and of depending on the temperature of the samples inside the cryotreatment

chamber. Out of the total 12 OFHC copper specimens, 8 were cryotreated at 98K for 24 hours with 5 hours of cooling and 9 hours of warming up to room temperature. The process indicator diagram of the cryotreatment system is shown in figure 2.

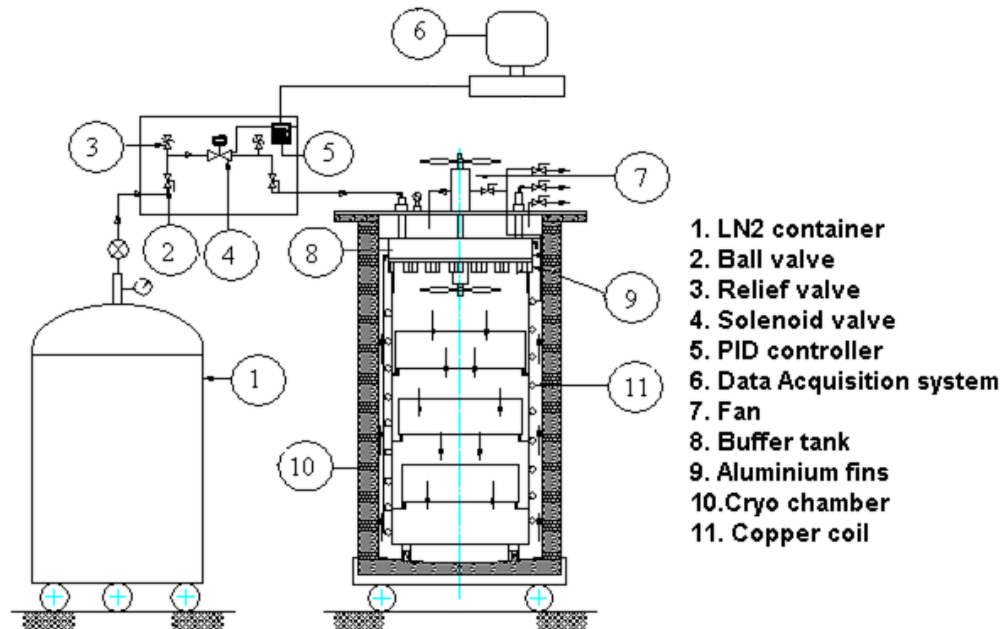


FIGURE 2. Process indicator diagram of the cryotreatment system.

Out of the 8 cryotreated samples, 4 samples were tempered at 423K for 1 hour in a furnace filled with inert gas (Nitrogen) to avoid oxidation of the external surface. [5,6]. The temperatures of the samples were gradually raised to avoid thermal shocks.

The thermal conductivity of all the specimens were experimentally determined using the laser flash apparatus as per the ASTM standard E 1461-01. In this equipment, a short pulse of energy is injected on one face of the copper disc which is maintained at constant temperature initially. Due to the input energy, the front face of the disc increases and propagates to the back surface through the material. The temperature increase in the back surface is dependent on the physical properties of the metal viz thermal diffusivity, thermal conductivity, specific heat and density. The thermal conductivity is measured using the equation 2.

$$\alpha = K / (\rho c_p) \quad (2)$$

where α is the thermal diffusivity, K is thermal conductivity, ρ is the density and c_p is the specific heat. [8,9]

The laser flash apparatus consists of a normal oscillation type ruby laser system, vacuum pumping system, a cryostat equipped with a double walled shroud for liquid nitrogen circulation along with a heating system for cooling/heating the sample space, sample assembly, temperature measuring system along with a PID controller to control the sample temperature and a data acquisition system with high speed transient memory. The schematic of the laser flash method is shown in the figure 3.

The specific heat is determined after measuring the temperature rise of the specimen at the back surface as a result of the imposed energy on its front surface. The principle of

specific heat measurement is based on the relative measurement technique. The temperature rise of the standard sample (sapphire) and glassy carbon combination whose specific heat and mass details are known is subjected to the laser pulse and the temperature rise is measured.

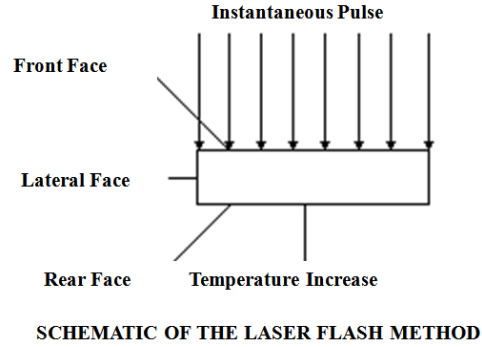


FIGURE 3. Schematic of the laser flash method to determine thermal conductivity.

Sapphire being a transparent material, a glassy carbon disc is thermally coupled to the sapphire for absorbing the incident energy from the laser pulse. Under the similar conditions, rise of the temperature of the test sample is measured. Under similar experimental conditions, with the known value of specific heat value of the standard, the unknown specific heat of the test sample can be determined, using the relation given in the equation 3.

$$[(m C_p)]_{\text{sapphire}} (\Delta T_1) + [(m C_p)]_{\text{glassy carbon}} (\Delta T_1) = [m C_p]_{\text{copper}} (\Delta T_2) \quad (3)$$

where m is the mass, C_p is the specific heat and ΔT is rise in temperature of the sample for sapphire, glassy carbon and copper respectively.

After determining the value of specific heat of the copper sample, the thermal diffusivity is measured by irradiating the copper specimen using laser pulse and recording the response of the temperature sensor fixed at the back surface. The value of the thermal diffusivity (α) is determined by the equation 4.

$$\alpha = (0.138L^2) / (t/2) \quad (4)$$

where L is the thickness of the copper specimen and $t/2$ is the time required for the back surface to reach half of the maximum temperature rise.

With the prior data of the density (ρ) of the copper samples and experimentally determined values specific heat and diffusivity, the thermal conductivity values of the samples were evaluated using equation 2 over the temperature range from 173 to 323K.

RESULTS AND DISCUSSIONS

The thermal conductivity values were experimentally determined using the laser flash apparatus in the temperature range from 323K down to 173K. Over the entire temperature

range, the K values have increased as the temperature is lowered. The test results for all the cases are shown in figure 4.

In the case of pure metallic conductors like OFHC copper, the thermal conductivity is a function of its specific heat (at constant volume), density, average particle velocity and mean free path of particles of the material. Even though phonon motion and electron motion play pivotal role in heat transport, the effect of phonon motion becomes significant as the temperature is lowered. In this temperature zone, the thermal conductivity becomes proportional to T^{-2} . This tendency of increased thermal conductivity property is observed in the untreated specimens.

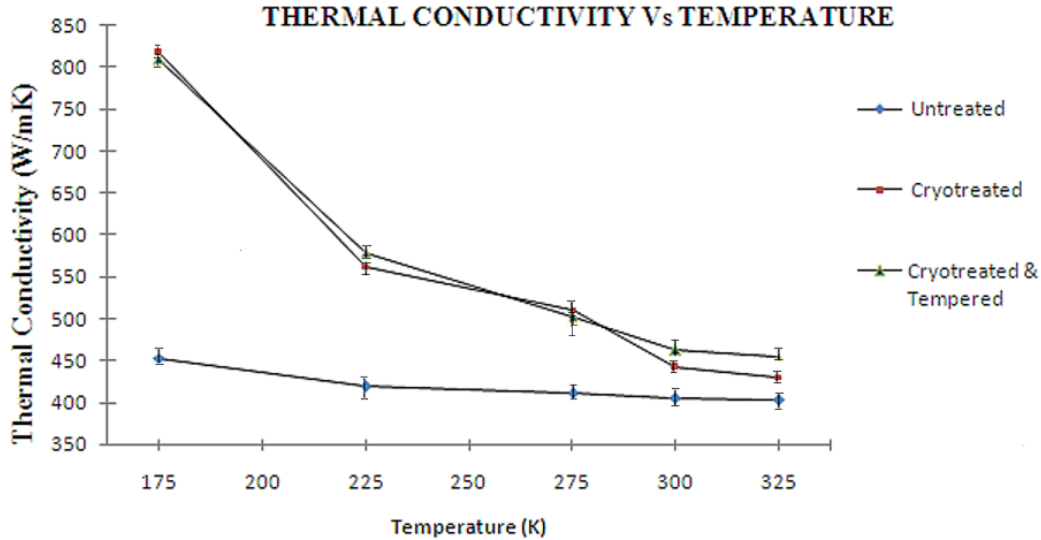


FIGURE 4. Test results of thermal conductivity experiment

As mentioned earlier, any metallic conductor will have developed internal stresses, strains and distorted lattice structure during the machining process. During the cryogenic treatment, the stresses are released resulting in reduced strains. The atomic structure gets refined. These combined effects act favourably for the heat carriers due to reduced scattering effect. This results in an easier path for the heat carriers inside the metal and hence thermal conductivity property gets enhanced. In this experimental study, significant increase in K values has been observed over the entire temperature range when compared with untreated specimens.

Tempering is a heat treatment process normally carried after the cryogenic treatment. During this process, the internal stresses are further relieved, resulting in further reduction in strains. These effects in turn reduce the scattering effect and increase the thermal conductivity values over the entire temperature range. Significant increase in K values were not observed when compared with the cryotreated samples. This result indicates the need for further studies to optimize the tempering parameters.

CONCLUSIONS

From the experimental study, it could be seen that cryogenic treatment can significantly improve the thermal conductivity properties of OFHC copper. While

cryotreatment could enhance thermal conductivity, tempering could marginally improve this effect. The results offer scope for further optimization studies with regard to cryotreatment and temper parameters like temperature and time. There is possibility for further studies by extending the temperature range of the thermal conductivity measurements down to liquid helium temperature. The study can be extended to other conducting metals like aluminum, silver and other alloys which are extensively used in cryogenic application.

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