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Copper produced from powder by HIP to encapsulate nuclear fuel elements

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COPPER PRODUCED FROM POWDER BY HIP TO ENCAPSULATE NUCLEAR FUEL ELEMENTS

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COPPER PRODUCED FROM POWDER BY HIP TO ENCAPSULE NUCLEAR FUEL ELEMENTS.

Lars B Ekbom and Sven Bogegård.*

Abstract:

In the Swedish nuclear waste mangement program, nuclear fuel elements are proposed to be encapsulated in copper canisters. To fill the space between the fuel elements two methods have been proposed. Originally lead was proposed to be cast into the canister. According to a second method the space between the fuel rods is filled with copper powder and hot isostatic pressed (HIP) to seal the canister lid and to densify the powder to homogenous copper. This latter method has the advantage that each fuel rod is individually encapsulated in a very corrosion resistant material.

This investigation was performed to find out to what extent pure copper powder can be hot isosatic pressed to full density and to achieve properties comparable to that of the oxygen free high conductivity (OFHC) copper of the canister.

OFHC copper was molten under helium gas protection and atomized to a fine spherical powder in a pilot plant. The powder was transfered to a glove box with an argon atmosphere. The powder was filled into a steel container, which was evacuated and sealed. HIP was done at 550°C and 200 MPa for one hour. The resulting copper was found to have a good ductility and mechanical properties comparable to that of ordinary copper. The constant strainrate stress corrosion test used to test the canister copper showed that the HIP-ed copper has the same good properties as OFHC copper.

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INTRODUCTION.

The Swedish program for Nuclear fuel waste management includes several barriers for waste products to reach the enviroment (1). The fuel elements are placed in a copper canister. Outside the canisters blocks of compressed bentonite will form a diffusion barrier to copper oxidants carried by the ground water. The rock surrounding the canisters will be the next barrier through which ground water in cracks will flow at a very low rate. Realeased radioactive substances will have to pass through 500 m rock to reach the enviroment.

The inner barrier consists of copper canisters produced from high purity oxygen free high conductivity copper (OFHC). The nuclear fuel elements are placed inside this canister. The inner space is filled and the lid is seeled by welding and/or hot isostatic pressing. Originally lead was proposed to be used to fill the inner space. However, considerable improvements in corrosion resistance would be gained if the fuel elements could be encapsulated in a solid copper matrix. It was therefor proposed to use copper powder as filling material and through a final hot isostatic pressing (HIP) of the canisters transform this powder into solid copper.

At the initial tests using copper powder to fill the inner space, the powder was atomized, reduced by hydrogen and HIP-ed. It had full density and a suitable microstructure, but was found to be rather brittle after heat treatment. At this state there was some doubt about whether the HIP-ed copper filling could be a complementary barrier for the corrosion penetrating the canister.

This investigation was undertaken to examine the possiblities to improve the quality of the atomized and HIP-ed copper so that its corrosion resistance could be regarded as equal to that of the canister itself.

Earlier investigations of the properties of atomized and hot isostatic pressed copper powders (2) (3) have shown that the conditions during the fabrication and filling of the powders into the containers are of fundamental importance for the quality of the HIP-ed material. Much care was thus used during the production.

EXPERIMENTAL.

High purity, oxygen free copper, OFHC, was melted in a laboratory

atomization plant at the Swedish Institute for Metals Research (3). Helium was used as a protective atmosphere. Charges of around 4 kg copper powder is produced. The powder container attached to the plant was taken directly over to an argon filled glove box without exposure to the air. Coarse powder particles and flakes (>80µm) were screened from the powder. The remainder was filled into small steel canisters of the type shown in Fig. 1. The canisters were seeled and taken out of the glove box for evacuation using a mechanical vacuum pump (0.01 torr). Sqeezing and welding was used to seal the containers before HIP.
HIP was done in laboratory plant at 200 MPa and 550°C for one hour.

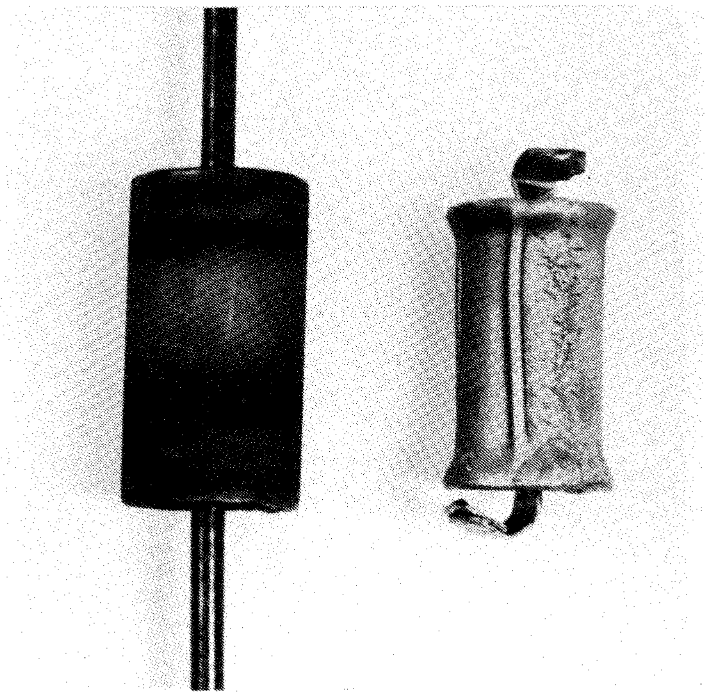

Fig. 1. Steel containers used for HIP of the copper powder.

The copper quality after HIP was preliminary controlled by a simple bending test, which showed that the copper could be bent 180° without cracks. Samples for mechanical tensile tests and for constant strainrate stress corrosion tests were taken from the HIP-ed copper cylinders.

The stress corrosion test were performed by Parkins (4) at University of Newcastle upon Tyne. Small rods of the material was pulled at a slow tensile elongation rate in a solution containing $NaNO_2$ at room temperature. The area reduction at fracture was determined.

Metallographic investigation was done to control the microstructure. Some of the samples were heat treated at 700°C for one hour. This is done to control that grain growth can occure and is not inhibited due to oxide skins on the atomized grains. Ion microprobe (At Chalmers Inst. of Technology) was used to determine the amount of impurities in the HIP-ed copper.

EXPERIMENT RESULTS.

The microstructure after HIP and after heat treatment is shown in Fig. 2. The spherical particles obtained at the atomization are clearly visible after HIP. This shows that small amounts of impurities, mainly oxides, are obstacles to a full joining of the grains. A heat treatment partly overcome these obstacles and gives a microstructure more like pure copper.

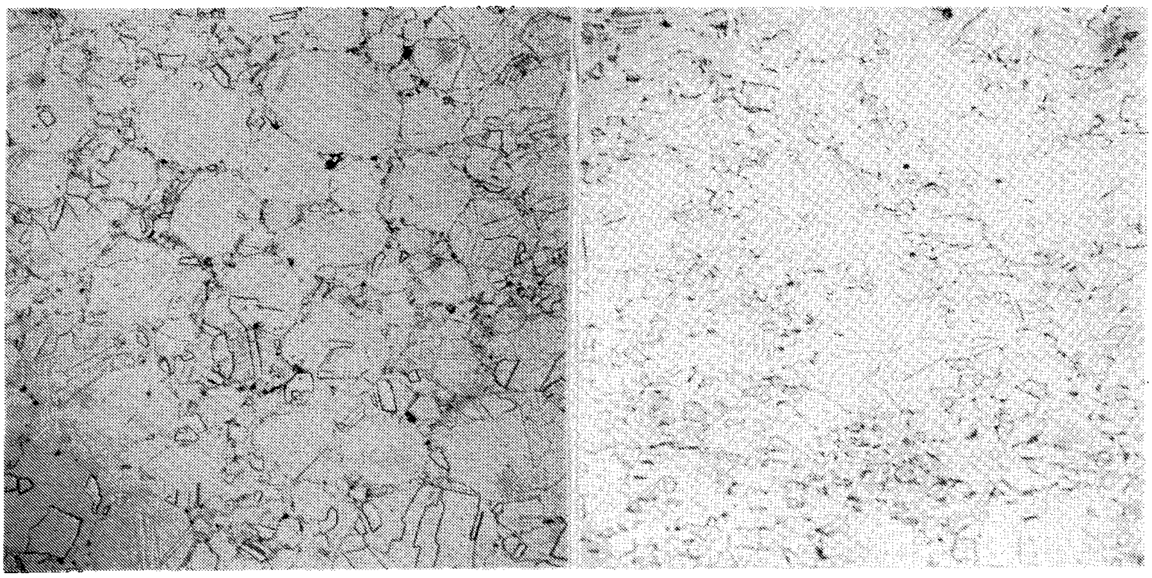

Fig. 2. Microstructure of the HIP-ed copper before and after heat treatment. LOM 300 x.

The very fine grain microstructure obtained at the atomization, is not changed essentially by HIP. The hardness is still high. The post-HIP heat treatment to some extent leads to a growth of the fine atomized microstructure.

The Ion microprobe analysis gave impurity distribution micrographs as is shown in Fig. 3. As can be seen impurities like sodium and calcium can be found at the particle surfaces. Inspite the very carefull handling of the powder these impurities, probably from dust, could not be avoided.

The result of the mechanical testing is shown in Table 1. The yield stress is high due to fine grains after atomization and HIP. The elongation to fracture is normal for a copper with the high yield stress but is low compared to soft copper. The area reduction shows, however, that the copper has a good ductility. After the heat treatment the yield stress has increased while the elongation to fracture is about the same.

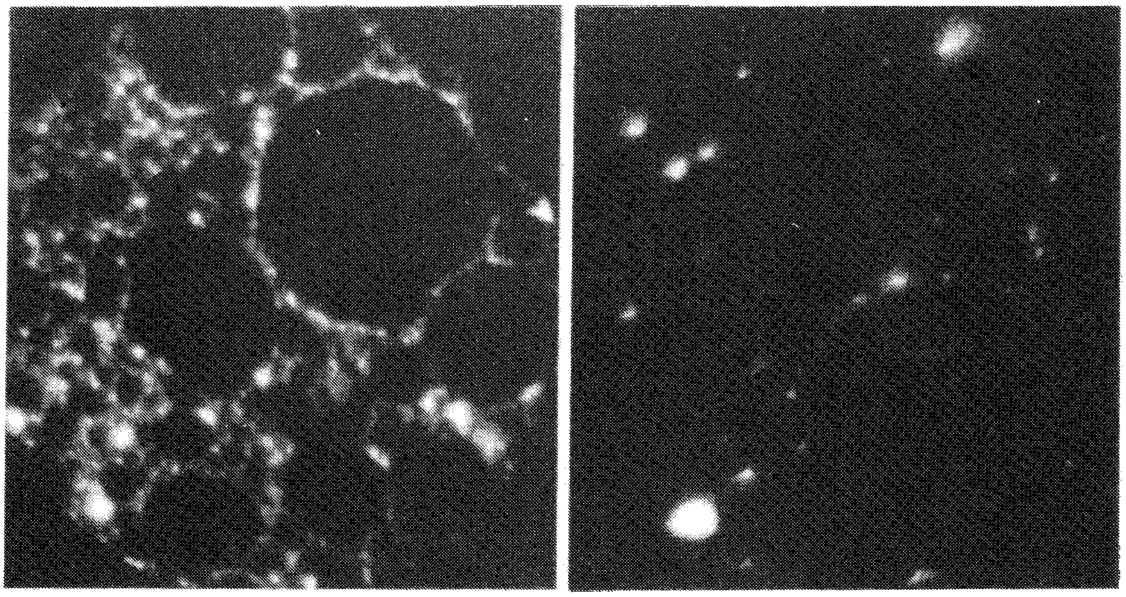

Fig. 3. Ion microprobe micrographs showing impurity distribution.

Table 1. Mechanical properties of HIP copper before and after heat treatment.

Sample	Yield stress MPa	Fracture str. MPa	Elongation %	Area red. %
After HIP	142	234	31	70
Earlier test*	151	231	49	x
Heat treated	202	252	29	70
Earlier test*	73	218	53	x

* In Ar, See (2).

The constant strainrate stress corrosion test gave values of the area reduction at failure as shown in Table 2. This is the same values as was measured for OFHC copper to be used in the canisters. They sho

Table 2. Results from the constant strainrate stress corrosion test (4).

Potential mV (SCE)	HIP copper powder	OFHC
0.1M $NaNO_2$		
+100	28%	28%
+ 50	32%	30%
0	69%	90%
- 50	63%	86%

there is no tendency to stress corrosion cracking of the material.

DISCUSSION OF THE RESULTS.

This investigation shows that there are good possibilities to fill the inner space of the canisters for nuclear fuel element waste with high quality copper. A proper handling of the copper powder to be HIP-ed gives the material properties comparable to those of the canister OFHC copper. A high quality of the copper filling can contribute to the corrosion resistance of the canister and thus to the time to break-through of an improbably deep pitting attack. Alternatively, the canister wall thickness can be reduced.

The ductility of the copper was good as can be seen from the area reduction, Table 1. However, the yield strength and hardness was high. This can be due to helium which was used at the atomization. Probably helium has gone into solution in the copper. A heat treatment at 700°C increases the yield strength, which can be due to reactions caused by the helium in solution. (Grain refinement, dislocation locking). At an earlier investigation (2) argon was used at the atomization. As can be seen from Table1 a low yield strength was achieved at that time.

The ion microprobe analysis, Fig. 3, shows that although the screening and filling procedure was performed in a sealed box, impurities apparently coming from dust, have stuck to the powder surfaces. These impurities have not influenced the ductility or stress corrosion resistance, but have an influence on the grain growth during heat treatment. Such impurities can probably be avoided at a closed room serie production for powder filling and HIP-ing of cannisters in the future.

Acknowledgement.
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