TECHNICAL UNIVERSITY OF CLUJ-NAPOCA

ACTA TECHNICA NAPOCENSIS

Series: Applied Mathematics, Mechanics, and Engineering
Vol. 61, Issue I, March, 2018

THE PROCESS OF ELIMINATING PORES FROM TUNGSTEN HEAVY ALLOYS BY LIQUID PHASE SINTERING OF W-Ni

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Abstract: *The study concerns the processes taking place in the liquid phase sintering of W-Ni. When we sinter fine powders of W and Ni, at first, Ni concentrates in the center of the sample, forming a high density area. This phenomenon occurs without any local density variation in raw and may be attributed to the decreasing tendency of the area of total liquid-vapors interface. After continuing the sintering, the dense area and Ni are dispersed towards the exterior, sometimes leaving behind isolated pores. The process of eliminating pores was studied using the large spherical pores artificially created by sintering the mixtures of Ni spherical particles and fine W powder. After prolonged sintering, these spherical pores are filled with liquid from the surrounding dense area. We found out that the liquid phase sintering of W-Ni takes place by concentrating the dispersed liquid from the dense area towards the exterior and by eliminating pores due to liquid flow.*
Key words: *Tungsten heavy alloy; liquid phase sintering; porosity.*

1. INTRODUCTION

Heavy alloys of W from the W-Ni-Fe or the W-Ni-Cu system were considered as model systems for liquid phase sintering, due to the almost spherical form of the grains. Surprisingly, however, there is no detailed study of the microstructure, by powder mixtures, that is close enough to those used in production. The difficulty of observation is due to a rapid densification, when using fine powders.

The purpose of this study is to observe the processes taking place during the liquid phase sintering of the W-Ni system. Since a liquid can easily flow, it is expected that it fills small capillaries and acquire a minimum configuration of the total surface energy. In real compacts, liquid is rarely distributed uniformly among the solid grains, as it is supposed in the models consisting of two particles [1] and [2]. As a consequence, liquid flowing and distribution presents a real interest within this study.

Within the experiments we used W particles of 10µm in size for the purpose of slowing down the densification process. We observe That during the sintering, the liquid is concentrated in a certain area of the sample, usually around the center, forming a densified area, which is clearly distinguished from the surrounding area with higher porosity. Then, the densified area expands, when Ni is dispersed towards the exterior [3], .

Following this initial study, a few isolated pores still remain. The process of eliminating such isolated pores is important, because after a liquid phase sintering it is often desired to reach a full densification. The process of eliminating isolated pores in “real” powders mixtures is often difficult to observe due to their small sizes, uneven distribution of their size and form [4].

In this study we performed a model of large size spherical pores, by mixing fine powders of W with spherical particles of Ni having quite an even size. By observing the evolution of these pores, created artificially, during long-term sintering, we could study the process of eliminating pores in the end stage of liquid phase sintering.

2. EXPERIMENTAL PART

The samples were divided in two groups: the first one is a mixture of fine powders of W and Ni, and the second one is a mixture of W powder and large spherical particles of Ni. The size of the W powder particle was: 10 ; 5.4 and 1.23 µm, while the size of the Ni fine powder was 4.6 µm. The size of the large spherical particles of Ni was 30 and 125µm.

The Ni content varied between 2 and 8 wt.%. The powders were homogenized and bilaterally pressed in a rigid die, with a pressure of 50 MPa resulting in samples with a section of 1 cm^2 and height approximately 1 cm.

Fig.1. Horizontal cross-section through the sample with W particles of 10µm in size and 2wt% fine Ni sintered at 1550ºC "0" hours.

The compacts were sintered in hydrogen atmosphere, in tubular oven with Mo electrical resistivity. The liquid phase sintering temperature was 1550ºC, and the solid phase sintering was performed at temperatures between 1200 and 1440ºC. The temperature was maintained with a precision of +10ºC. The sintering time varied from 0 to 24h. The average heating velocity up to the sintering temperature was approximately 50ºC/min. The sintering time "0" means that the sample was pushed to the center of the oven and then taken out. It is estimated that the sample was over the melting point of Ni for about 5 min during the heating and cooling phase.

3. RESULT AND DISCUSSIONS

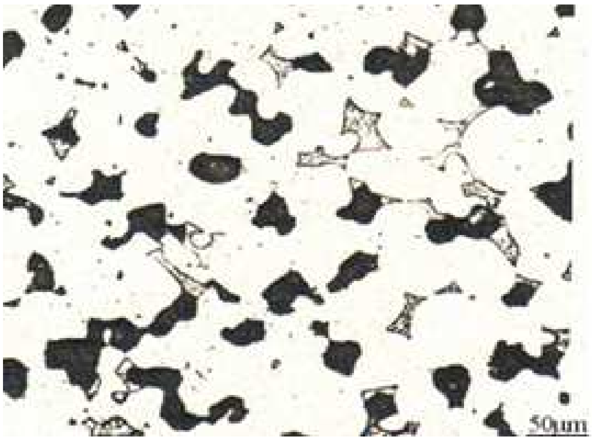

Fig.2. The same sample as in fig 1, with liquid concentration in the inner area.

We studied first of all the microstructural changes during heating and sintering of the mixtures of fine powders of W and Ni. The most marked phenomenon in the initial stage consisted of the formation of an area of high Ni content and large density around the center of the samples. Figure 1 presents a horizontal cross-section of a sample with 98wt% W measuring 10µm in size and 2wt% fine Ni, sintered at 1550ºC, time "0". The inner and exterior area of this sample is shown in figures 2 and 3. The variations of the porosity and content of Ni in radial direction are shown in figures 4 and 5.

Fig.3. The same sample as in fig 1, with liquid concentration in the exterior area.

Results clearly show that the Ni concentrated in the center, forming an area of low porosity.

At further sintering, the dense central area has grown towards the exterior until the entire sample was almost completely densified. The modifications that occurred after 1h are shown in figures 4 and 5. The distribution of the density was now almost even, and Ni flowed

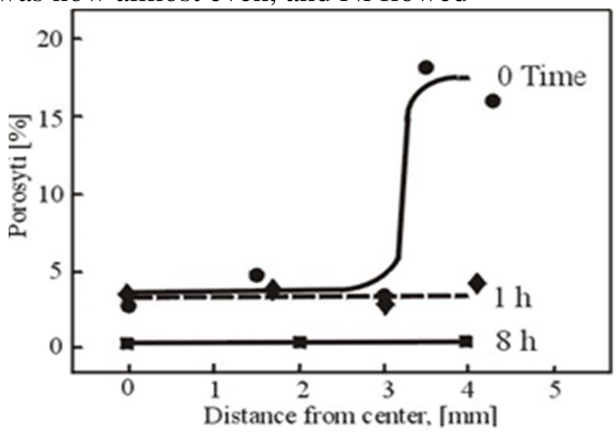

Fig.4. Variation of the porosity in radial direction of the sample shown in figure 1 after different sintering times.

towards the exterior. In this stage, only isolated pores remained in the sample, and the pores of the exterior area were slightly larger than those in the center. After 8h, the isolated pores were eliminated.

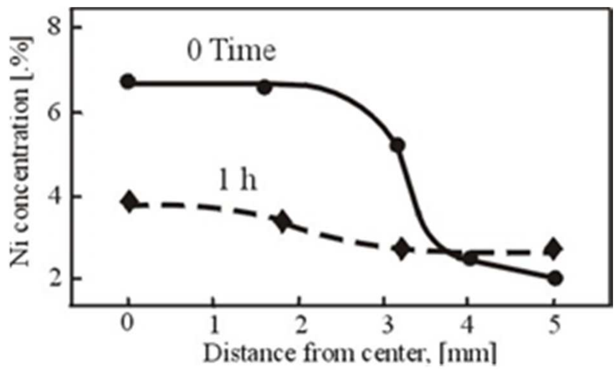

Fig.5. Variation of the Ni content in radial direction of the sample shown in fig. 1 after different sintering times.

These processes happened extremely fast when the Ni content was greater or the W powder was finer. In the samples with W of 10μm and 6 or 8wt% Ni, the Ni concentrated after 10 min. of sintering at 1350ºC below the Ni melting point. The samples were completely densified by heating at 1550ºC. With finer wolfram (1.23μm) and 4 or 6wt% Ni we obtained an almost complete densification when heating at 1200ºC. Therefore, when we used fine powders to produce W-Ni-Fe alloys, the Ni concentration and the densification in the center would occur during the heating phase, and the moment of reaching the liquid phase sintering temperature is not observed.

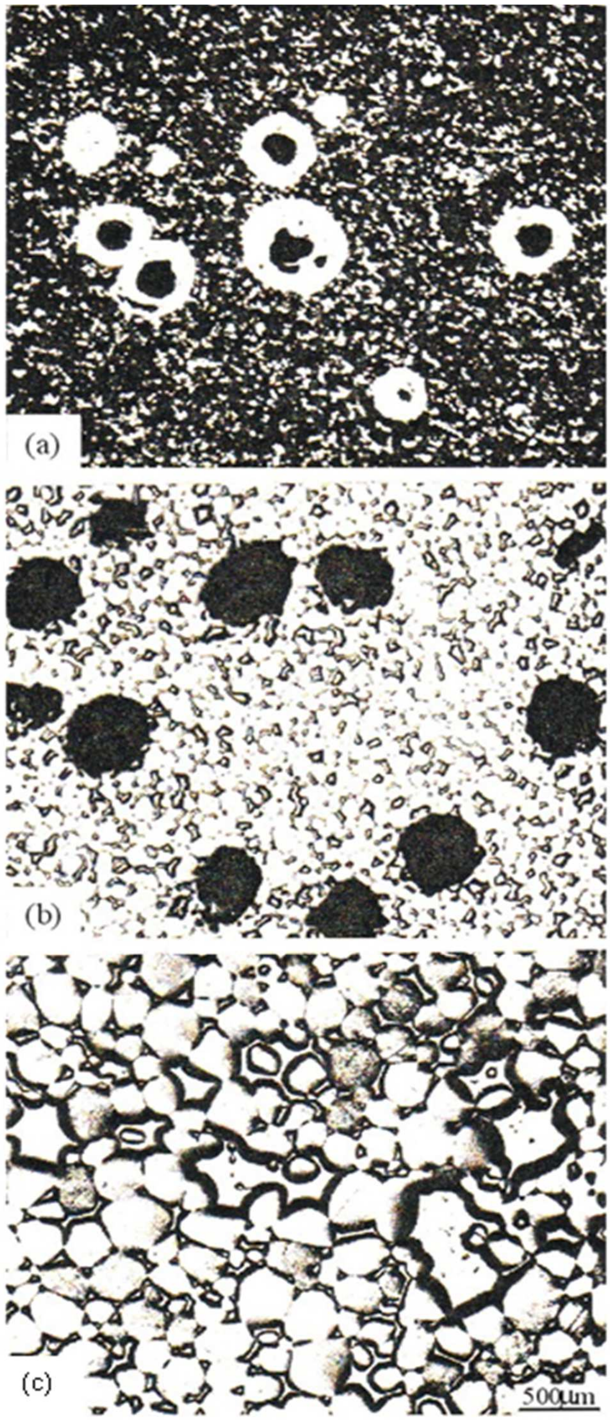

Fig.6. Microstructure changes during sintering a sample with 4wt% Ni with particle size of 125μm, in various stages: (a) - 1400^{0}C, 10 min ; (b) - 1550 ^{0}C, 30 min ; (c) - 1550 ^{0}C, 8h.

The next series of experiments referred to the elimination of isolated residual porosity during the final study of liquid phase sintering. We prepared samples containing large spherical pores with even size, by sintering mixtures of large spherical Ni particles and W fine powder.

Figure 6 presents changes of the microstructure during the heating and sintering phases of a sample containing 4wt% Ni with particle size of 125µm. After 10 min at 1400ºC small pores began to form in the center of the Ni particles. These small pores that would correspond to the "natural" pores found in mixtures of fine powders were eliminated after 30 min of sintering.

The process of filling the large pores clearly manifested after 8h of sintering, all the pores were filled and turned into accumulation reservoirs for liquid.

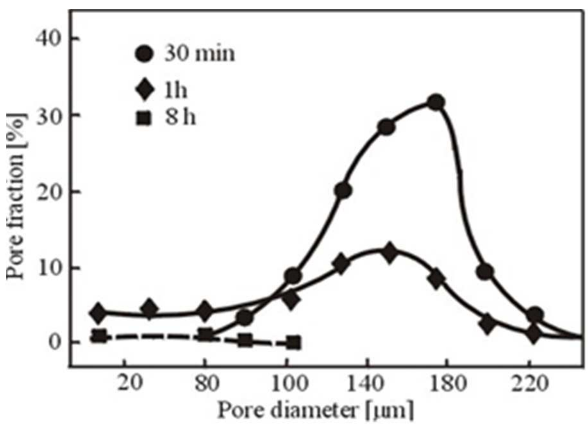

Fig.7. Distribution of the pores size (in relation with the same number of initial pores from each cross-section) in the sample presented in figure 6.

The result is also represented graphically in figure 7. We measured the apparent diameter of all the pores in each sample in cross-section. This figure also presents a few pores filled after 1 h, as well as an almost complete densification after 8 h.

4. CONCLUSIONS

- The experimental results prove that the densification processes during the liquid phase sintering are not even in the entire sample, because the liquid can flow freely and can form configurations that correspond to a lowest level surface energy.
- The processes taking place during the liquid phase sintering of W-Ni are: concentration of liquid, liquid flow towards the exterior and expansion of the dense area and filling of the isolated pores with liquid.
- In stages two and three, the pores are filled by the liquid flowing from the dense areas, where the grains are dipped in liquid. A negative pressure on the liquid would produce the accommodation of the grains form and the release of a quantity of liquid.
- It is expected that the accommodation of the grains form which are surrounded by liquid takes place by dissolution and re-precipitation into liquid, rather than diffusion, a mechanism that is probably the most important in the liquid phase sintering.

5. REFERENCES

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Procesul de eliminare a porilor din aliajele grele pe baza de wolfram prin sinterizarea cu faza lichida a W-Ni

Rezumat: Sunt luate in studiu procesele care au loc la sinterizarea cu fază lichidă a W-Ni. Când se sinterizează pulberi fine de W şi de Ni, la început, Ni se aglomerează în centrul probei, formând o zonă cu densitate mare. Acest fenomen apare fără vreo variaţie locală a densităţii la crud şi poate fi atribuit tendinţei de descreştere a ariei interfeţei totale lichid-vapori. După continuarea sinterizării, zona densă şi Ni se propagă spre exterior, lăsând în urmă uneori, pori izolaţi. Procesul de eliminare a porilor a fost studiat cu ajutorul porilor sferici mari care au fost creaţi în mod artificial prin sinterizarea amestecurilor din particule sferice de Ni şi pulbere fină de W. După o sinterizare prelungită aceşti pori sferici sunt umpluţi cu lichid din zona densă înconjurătoare. S-a constatat că sinterizarea cu fază lichidă a W-Ni are loc prin aglomerarea lichidului propagat din zona densă înspre exterior şi prin eliminarea porilor datorită curgerii lichidului.

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