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## 主 論 文 の 要 旨

論文題目 Improvement of osteoconductivity of valve metals and their alloys by hydroprocessing  
(湿式表面改質によるバルブメタル・合金の骨伝導性向上)

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## 論 文 内 容 の 要 旨

### ABSTRACT

Anti-corroded valve metals (Ti, Nb, Ta, and Zr) and their alloys are widely used as biomedical materials because of their excellent biocompatibility, excellent corrosion resistance and appropriate mechanical properties. They also have recognized as non-toxic elements and do not contribute any harmful reaction in human body. However, they show a low direct bone bonding (osseointegration) because of bioinertness. Thus, many researchers tried to concentrate on surface modification to increase their bioactivity, thereby shortening the healing period and increasing clinical success of valve metals and their alloys, especially in areas of poor bone quality. Surface roughness and hydrophilicity are important factors of biomaterials which related with protein adsorption and cell adhesion after implantation into the body, consequently their osseointegration. In recent studies, various methods such as, anodizing, UV light irradiation, plasma irradiation and hydrothermal treatment in order to create hydrophilic surface in improving implants osseointegration and osteoconductivity has been investigated.

For this purpose, I tried to improve the osteoconductivity of valve metals and their alloys by applying above methods. *In vivo* test was conducted to evaluate the osteoconductivity of valve metals and their alloys. Firstly, four surface modification methods (anodizing, UV light irradiation, plasma irradiation, and hydrothermal treatment) were applied

to pure Ti. Although UV light irradiation and plasma irradiation processes created the hydrophilic surfaces on pure Ti as well as anodizing and hydrothermal treatment, they are difficult to apply for complex-shaped substrates of implant materials with complex topographies, because of the line-of-sight nature of the methods. Super-hydrophilic surface (water contact angle lower than 10 degree) with osteoconductivity higher about 50% than as-polished Ti was provided on hydrothermaled pure Ti after stored in x5 PBS(-).

Subsequently, I continued my research to improve other valve metals (such as, Zr, Nb, and Ta), which pure Ti was used as a reference. Anodizing and hydrothermal treatment were applied to those valve metals. The surface hydrophilicity of Zr, Nb, and Ta samples were not enhanced by anodizing as well as pure Ti. Their surfaces were still in hydrophobic conditions with water contact angle higher than 60 degree. In contrast, the improvement of surface hydrophilicity was detected after the hydrothermal treatment was implemented to all valve metals. This condition became more valuable when the hydrothermaled valve metals stored in x5 PBS(-) where their osteoconductivity increased up to 55 % compared than those of as-polished samples. It means that, hydrothermal treatment that continue by storing in x5 PBS(-) is not only effective to improve the osteoconductivity of pure Ti, but also effective for Zr, Nb, and Ta.

Hydrothermal treatment was conducted to Ti alloys (such as, Ti64, Ti67, TNTZ, and TCFA) in order to improve their osteoconductivities. Anodizing was used for comparison. It was found that anodizing only successful to produce hydrophilic surfaces on Ti, Ti64, and Ti67 and improved their osteoconductivities, but it was not useful for TNTZ and TCFA alloys. The hydrophobic surfaces were still formed on the surface of these two alloys. Consequently, the osteoconductivities of anodized TNTZ and TCFA were low and almost similar to those of as-polished conditions. On the other hand, all the Ti alloys surfaces became-hydrophilic after hydrothermal treatment. When the hydrothermal Ti alloys were continue stored in x5 PBS(-), the superhydrophilic surface were formed on all Ti alloys and improved their osteoconductivities significantly. It can be said that a combination process between hydrothermal treatment and storing in x5 PBS(-) became a simple and effective way of enhancing the osteoconductivity of not only for pure Ti, but also for Ti alloys.

Then, the effect of hydrothermal treatment on osteoconductivity of Zr alloy (Zr-9Nb-3Sn) was examined. Anodizing was also used as comparison. Similar to its pure metal alloys (Zr and Nb), anodizing did not provide a significant enhancement of the hydrophilicity of Zr-9Nb-3Sn alloy. However, this condition became different when

hydrothermal treatment was applied to Zr-9Nb-3Sn alloy. Its surface became hydrophilic after hydrothermal treatment. In addition, higher osteoconductivity was produced on hydrothermally treated Zr-9Nb-3Sn alloy after stored in x5 PBS(-). This suggests that hydrothermal treatment become one of the promising surface treatments which improve the osteoconductivity of implant materials such as Zr, Nb, and Zr-9Nb-3Sn alloy.