Surface modification of titanium subjected to hydrostatic extrusion

INTRODUCTION

The basic technology for producing large volumes of bulk nanocrystalline metals is based on the Severe Plastic Deformation (SPD) methods, which enable refining the structure from the micrometric to the nanometric level. The microstructural changes are accompanied by a significant improvement of the mechanical strength and hardness [1÷7]. One of the SPD methods is hydrostatic extrusion, employed in the present experiments, which has been proven to efficiently refine the grains in aluminum, copper, nickel and steel [8÷12].

Our earlier studies have shown that hydroextrusion can also be used for producing nanostructured titanium [13÷15] with an equivalent diameter of grains about 60 nm and substantially increased strength and hardness.

The chief difficulty in producing nanocrystalline titanium by HE is that the reduction of the rod cross-section must be realized gradually in many passes. Other disadvantageous effects that occur during HE of Ti are: the high extrusion pressure, the recurrent elastic deformation, the stick slip effect (stepwise displacement of the material) and a considerable wear of the die resulting from the tribological conditions.

The aim of the study was to reduce or eliminate these adverse effects, and thereby to optimize the hydroextrusion process of titanium. To this end the surface of titanium was subjected to various modifications. The beneficial effect of Ti-Al coatings was shown in our earlier publications [15, 16]. Even though the extrusion pressure was reduced, the Ti-Al layers underwent wear in a short time. In the present study, the Ti billets were covered with Al coatings of various thicknesses. The effect of these coatings on hydroextrusion process, the surface topography of the Ti products and their microstructure after the extrusion were investigated. The maximum strain was also determined that could be applied in a single and two extrusion passes.

MATERIALS AND PROCESSING

The material examined was pure polycrystalline technical grade 2 titanium (Ti>99.4) in the form of rods 12 mm in diameter. Prior to extrusion, the titanium rods were covered with aluminum coatings 5 and 10 μm thick using magnetron sputtering (MS). Then the rods were subjected to hydrostatic extrusion which was conducted with various numbers of passes and strains (Tab. 1).

The extrusion processes were conducted at the Institute of High Pressure Physics, Polish Academy of Sciences, Warsaw, within the framework of a project coordinated by the Faculty of Materials Science and Engineering, Warsaw University of Technology.

The microhardness of the rod-shaped samples before and after the extrusion, was measured on their surface and cross-sections using the Vickers method under loads of 20 and 200 g. EDS linear and point analyses of the chemical composition were conducted using 2600N and TM1000 scanning electron microscopes (SEM). The nanohardness (nanoindentation) was determined with a Hysitron triboindenter under a load of 100 mN. A scratch test was used to estimate the adhesion of the coatings to the titanium substrate (CSM Instruments equipment with a steel ball under a linearly increasing load).

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RESULTS AND DISCUSSION

Titanium in the as supplied state

The assumption underlying the experiments was to produce on titanium billets aluminum coatings that adheres very well to the substrate and exhibit better plasticity. The coatings were investigated via scanning electron microscopes (SEM), EDS analyses, nanoindentation and microhardness and finally using „scratch test" method.

The examinations performed showed that the Al coatings are homogeneous and uniformly distributed (Fig. 1). They have adhesive character and contain no transition (Ti-Al)-based intermetallic regions of the diffusive character (EDS analysis – Fig. 2). Examinations of the nanoindentation conducted under a load of 100 mN (0.01 g) proved that thinner coatings have high nanohardness (Tab. 2), due to the effect of the titanium substrate. Although the penetration depth of the indenter was the same in both cases, the influence of the substrate was certainly stronger in the sample with the thinner coating.

Table 1. Process parameters and the HE passes applied to titanium coated with 5 and 10 μm layer of aluminum

Tabela 1. Parametry i schemat procesów HE tytanu z powłokami aluminium o grubościach 5 oraz 10 μm

HE parameters	Ø12→Ø7.5	Ø12→Ø6	Ø12→Ø5	Ø12→Ø4	Ø12→Ø3
Coating 5 μm	extruded	extruded	extruded	extruded	extruded
Coating 10 μm	not extruded	not extruded	extruded	not extruded	extruded
Accumulated strain ε	0.94	1.38	1.75	2.19	2.77
Numer of passes	1	1	1	2	2

Table 2. Nanoindentation results obtained for the Al coatings and Ti substrate before HE

Tabela 2. Wyniki pomiarów nanoindentacji dla powłok Al oraz podłoża Ti

Materials / Hardness	Al 5 μm	Al 10 μm	Ti
HV0.01	246	202	342

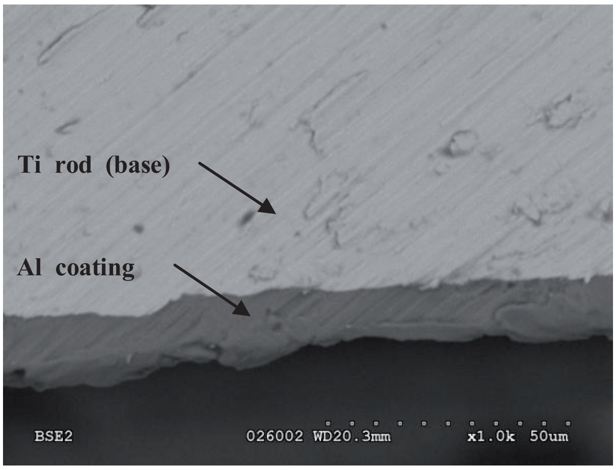

Fig. 1. SEM image of an Al coating (10 μm thick) formed on a Ti substrate before HE

Rys. 1. Obserwacje SEM powłoki Al (grubość 10 μm) na podłożu Ti przed HE

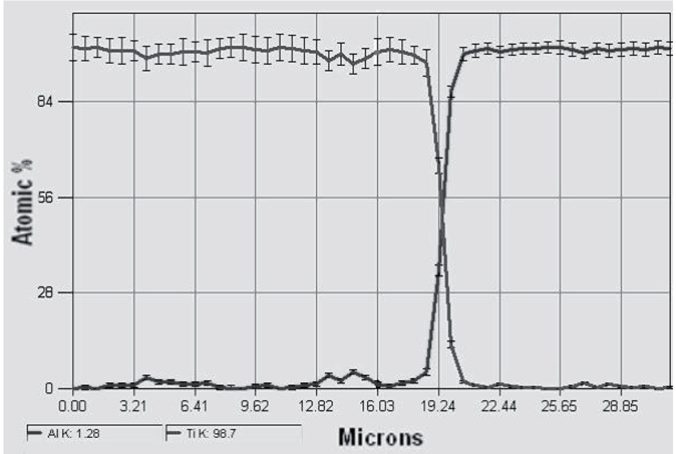

Fig. 2. EDS analysis of an Al coating (10 μm thick) and its Ti substrate before HE

Rys. 2. Analiza EDS dla powłoki Al (grubość 10 μm) i podłoża Ti przed HE

The dynamic scratch tests (i.e. conducted under increasing load) revealed very good adhesion of MS-deposited Al coatings to the Ti substrate. No cracking or spalling was observed. The coatings showed high plasticity. The good adhesion of the coatings was also proved by the acoustic emission.

HE processed samples

During the extrusion process, the Al coating and Ti substrate were subjected to plastic deformation, bringing about reduction of the diameter. Even though the extrusion was carried out at high strain, the Al coatings maintained good adhesion to the substrate with no visible cracking or spalling. During extrusion, the coatings functioned as a lubricant which separated titanium from the die reducing the friction and protecting titanium from adhering to the die. The coating on the billet extruded with a diameter change from 12 to 7.5 mm ($\varepsilon = 0.94$) in a single operation, is continuous on the entire surface of the rod (Fig. 3). Also, the surface of the extruded product is decidedly less rough than in the case of the titanium extruded with no coating under the same strain.

Only for the highest diameter reduction (Ø12→Ø5 mm – the highest of those applied in the present experiments) the Al coating degradated and the disadvantageous tribological conditions appeared at the billet/die contact (Fig. 4). This has led to the increased roughness of the titanium surface, and scratches on the extruded products.

Similar results were obtained with the rods 3 mm in diameter, extruded in two passes: Ø12→Ø6→Ø3 mm at $\varepsilon = 2.77$ (Fig. 5, 6). In this case the aluminum was deformed into the irregularities of the surface. The remaining Al filled cavities of the titanium surface, thereby improving the lubrication conditions. This effect occurred irrespective of the thickness of the Al coating. In the case of the 10 μm coatings, in which the amount of aluminum is higher, the amount of Al that remaining at the titanium surface after extrusion is only slightly higher.

Technological advantages

Our earlier studies [15] have shown that the coating of Ti rods with layers of Al permit reducing considerably (by about 30÷50%) the maximum extrusion pressure. This in turn makes possible the increasing of the strain (i.e., increasing the reduction of the rod diameter) applied in a single extrusion pass and thereby decreases the number of passes necessary to produce nanocrystalline titanium. The aim of the present studies was to estimate the maximum strain (reduction), possible to apply during the one pass of extru-

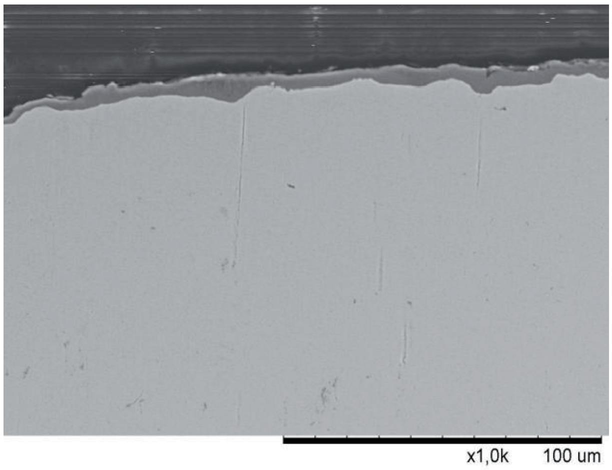

Fig. 3. SEM image of a 5 μm Al coating after HE; Ti rod diameter reduction from Ø12 to Ø7.5 mm

Rys. 3. Powłoka Al 5 μm na podłożu Ti po HE. Średnica Ø7,5 mm uzyskana po wyciskaniu Ø12→Ø7,5 mm

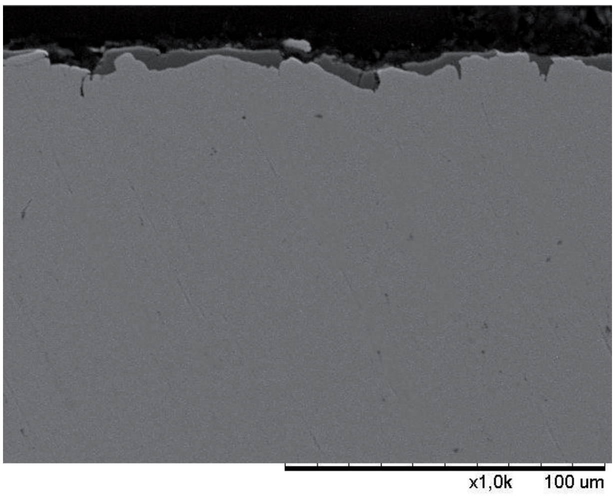

Fig. 4. SEM image of a 5 μm Al coating after HE from Ø12 to Ø5 mm

Rys. 4. Obraz SEM powłoki Al (5 μm) na podłożu Ti po WH. Średnica Ø5 uzyskana z wyciskania Ø12→ Ø5 mm

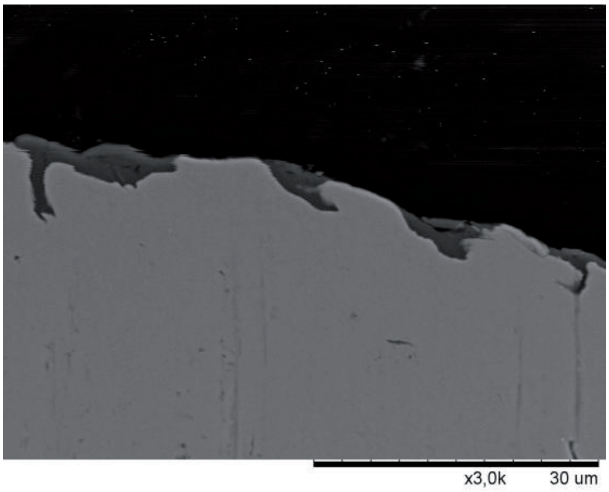

Fig. 5. A 5 µm Al coating after subjecting the Al-coated Ti rod to two stage HE; rod diameter reduction Ø12→Ø6→Ø3 mm
Rys. 5. Powłoka Al (5 µm) na podłożu Ti po WH. Średnica Ø3 mm uzyskana po wyciskaniu Ø12→Ø6→Ø3 mm

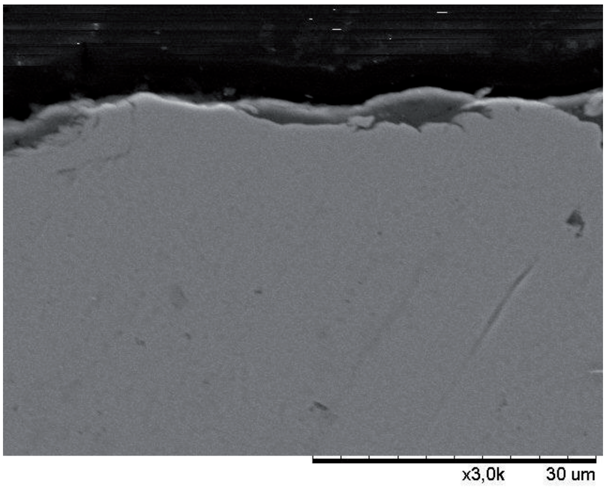

Fig. 6. A 10 µm Al coating after subjecting the Al-coated Ti rod to two--stage HE; rod diameter Ø12→Ø6→Ø 3 mm
Rys. 6. Powłoka Al (10 µm) na podłożu Ti po HE. Średnica Ø3 mm uzyskana po wyciskaniu Ø12→Ø6→Ø3 mm

sion. Also, the study provided an estimate of the minimum number of HE passes which are necessary to obtain the Ø3 mm rods from Ø12 mm billets.

The HE processes applied in this work were analysed in terms of the maximum extrusion pressure, wear of the die, "spring back" and "stick-slip" effects.

The results of experiments have shown that the reduction of the diameter from Ø12 to Ø3 mm of Ti rods without coatings requires from 5 to 10 extrusion passes. On the other hand rods with the Al coatings can be extruded in 2 HE passes (Fig. 7). By using coatings, it is possible to increase the strain applied in each cycle from 0.94 to 1.75. This is because of the pressure decrease. The reduction of the rod diameter (*D*) as a function of strain and the corresponding values of the maximum extrusion pressure are shown in Figure 7.

Therefore, it can be concluded that by coating Ti rods with the Al several technological benefits can be achieved such as:
- reduction of the frictional wear of the die,
- decrease of the surface roughness of the extruded rods,
- no "spring back" effect (recurring elastic deformation),
- elimination of "stick slip" effect,

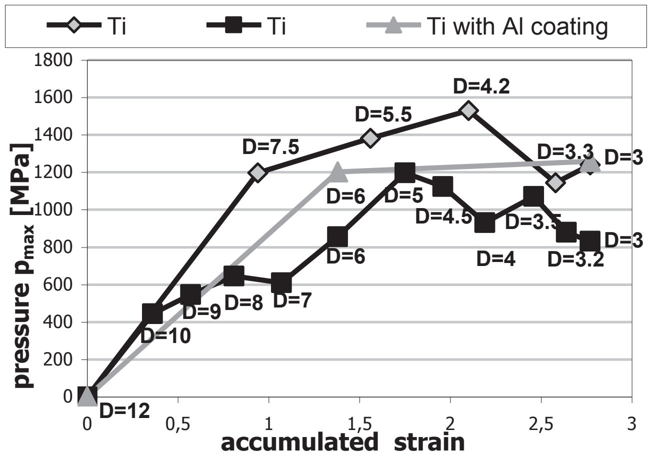

Fig. 7. Reduction of the number of extrusion passes achieved thanks to the presence of Al coatings
Rys. 7. Zmniejszenie ilości operacji HE dzięki powłokom Al

- significant reduction of the maximum extrusion pressure (p_{max}),
- decrease in number of the extrusion passes needed for grain refinement.

Microhardness analysis

The microhardness of the Ti rods before and after extrusion was measured on cross-sections of the rods and on their circumference. The values obtained on the rod cross-sections and surface are similar (Tab. 3). This is an evidence that no hard diffusion-type surface layers have formed. On the surface of the extruded rods there is only a soft thin Al coating (occasionally discontinuous) which does not affect the hardness measurements conducted under a load of 200 g.

It should be noted that the strain (and hence diameter reduction) applied in the extrusion of the Al-coated rods was considerably higher than that of uncoated billets. This resulted in a reduction of the number of extrusion passes and in an increase of the temperature of the extruded material. Nevertheless, the HE markedly increased the microhardness of the Al-coated rods to the values obtained for uncoated rods after HE (Tab. 3, Fig. 8).

It can be noted that by reduction of the initial diameter (Ø12 mm) to the final (Ø3 mm), an increase of microhardness for samples with coatings was by 35% and for samples without coatings 45% . This is an evidence that in both cases the degree of material hardening was the same. Microhardness of the uncoated Ti and the Al-coated Ti extruded at the same or similar accumulated strain is shown in Figure 8.

Table 3. Composition of results of the microhardness (HV0.2) testing
Tabela 3. Zestawienie wyników pomiarów mikrotwardości (HV0.2)

Diameter		Ø12	Ø7.5	Ø6	Ø5	Ø4	Ø3
Thickness 5 µm	section	175	220	225	223	235	237
	surface	180	207	226	228	-	-
Thickness 10 µm	section	175	-	-	218	-	240
	surface	180	-	-	228	-	237

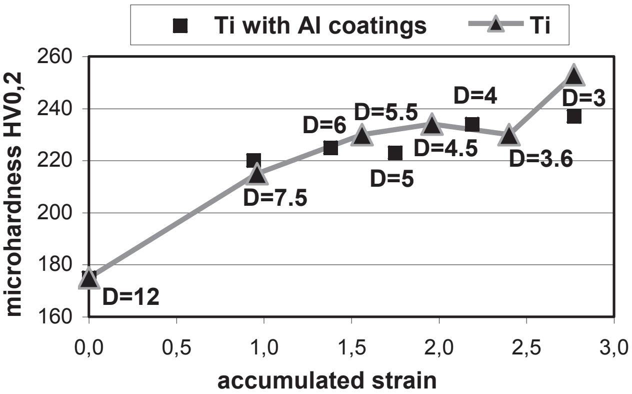

Fig. 8. Comparission of the microhardness of titanium uncoated and coated measured after HE

Rys. 8. Porównanie mikrotwardości Ti po HE z powłokami oraz bez powłok

CONCLUSIONS

The results reported here lead to the following conclusions:

1. Coating of the surface of Ti samples with Al gives numerous technological advantages, in the first place the reduction of the maximum extrusion pressure and the minimization of the die wear.
2. The hardening of the material during HE is the same for uncoated and Al-coated titanium.
3. With the Al coatings 5 and 10 μm thick, it is possible to conduct a single HE passes using strain up to 1.75, or two HE cycles conducted at a total strain of 2.77.
4. The surface coating of Ti with the Al enables reducing the number of HE cycles necessary to produce nanocrystalline titanium.
5. Within the range of the rod diameter reduction and the thickness of the Al coatings investigated in the present study, no effects of the coatings on the course or the results of the extrusion were observed.

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