A New Laminate Composite System: Metallic-Intermetallic Laminate Material

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Abstract: Metallic-intermetallic laminate (MIL) composites consisting of alternating layers of Ta, Al and the intermetallic Al_3Ta have been fabricated by reactive foil sintering in open atmospheric furnace. In this study, tantalum and aluminum foils with initial thicknesses of 250 µm were used. Sintering process has carried at 850-900 and 950 ºC for 5 and 7.5 hours under 2 MPa pressure. The aluminium foil was consumed by forming a tantalum aluminide intermetallic compound. Thus, the final microstructure consists of alternating layers of intermetallic compound and unreacted Ta metal. Microstructural characterizations of produced composites have conducted by using scaning electron microscope. Hardness values of test samples have also measured by vickers indentation method.

Key Words: Intermetallic, tantalum aluminide, laminate composite, sintering

Introduction

It is well known that the widespread engineering application of ceramics and other highly brittle materials, e.g. intermetallic compounds, is severely limited by their low toughness. A number of toughening strategies have been proposed to improve the critical stress intensity required for crack propagation. One of the most effective toughening techniques is to introduce a ductile phase, which remains intact and bridges the crack faces in the wake of a growing crack. Under such a circumstance, the crack tip is shielded by the closure traction imposed by the plastic deformation of ductile ligaments. In particular, when layered ductile phases are incorporated, laminate composites can be formed, which enhance the toughness (Oktay, 2010, p.1043-1050, Peng, 2004, p.243-248).
Metallic-intermetallic composites can be designed for structural use to optimize the unique properties and benefits of the constituent components, resulting in materials that have the high strength and stiffness of the intermetallic phase and the high toughness of the metal. Pro intermetallics have been reinforced with particles, rods, or layers of ductile metals in efforts to increase toughness. Ductile phase reinforcement of brittle materials utilizes crack-laminate interactions to generate a zone of bridging ligaments that restrict crack opening and growth by generating closure tractions in the crack wake and utilize the work of plastic deformation in the ductile metal phase to increase fracture resistance of the composite (David, 2005).

Laminate composites are being intensively studied for a number of potential applications: electronic devices, structural components, armor, etc. Ceramic–ceramic, metal–ceramic, metal–metal, metal–ceramic–intermetallic and metal–intermetallic systems have shown desirable properties (Tiezheng, 2004, p.10-26). In particular, the Ti–Al_3Ti system has a great potential for structural applications because of its low density and excellent specific mechanical properties (Rohatgi, 2003, p.2933-2957, Price, 2011, p.1334-1346)

Titanium–titanium tri-aluminide (Ti–Al_3Ti) metallic–intermetallic laminate (MIL) composites have been produced from elemental titanium and aluminum foils by a novel one-step process utilizing a controlled reaction at elevated temperature and pressure. Of the various possible aluminides in the Ti–Al system, the formation of the intermetallic Al_3Ti is thermodynamically and kinetically favored over the formation of other aluminides when reacting Al directly with Ti. This preferential formation of Al_3Ti is fortuitous as its Young's modulus (216 GPa) and oxidation resistance are higher, and the density (3.3 g/cm^3) lower than that of the other titanium aluminides such as Ti_3Al and TiAl **(**(Rohatgi, 2003, p.2933-2957, Zeytin, 2008).

Intermetallic compounds based upon aluminides of several transition metals such as iron, nickel, titanium, cobalt, niobium, and tantalum have been recognized as potential candidates for the high-temperature structural applications. Among various aluminides of transition metals, the compounds in the Ta–Al system have been of great interest for their excellent mechanical properties and high degree of structural complexity. The Ta–Al phase diagram depicted in Fig. 1 shows the existence of four aluminides, including Ta_2Al, TaAl, $TaAl_2$, and $TaAl_3$. (Yeh, 2010, p.153-158).

They are stable, refractory and reflective, and have been proposed as mirror coatings for use in the IR. Melting point of 1400°C and the density is 7.02 g/cm^3. Because of this aspects, tantalum aluminides are can be used in high temperature applications (http://en.wikipedia.org/,2012)

Tantalum-aluminum system is one of the most well known in terms of the formation of intermetallic phase. This system is also in the priority among in laminate composite systems. The objective of the present research is to synthesize tantalum-tantalum aluminide metallic-intermetallic composites and its microstructural characterization.

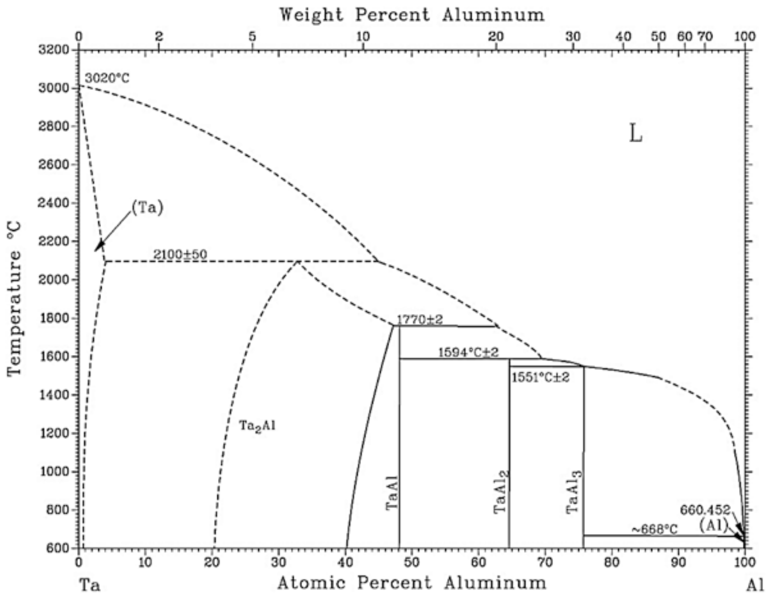


Figure 1: Ta-Al phase diagram (Yeh, 2010, p.153-158)

Materials and Method

The MIL process consists of stacking commercial purity aluminum and tantalum aluminide foils in alternating layers. The foils were provided from Alfa Aesar Company and their properties are shown in the table 1.

Table 1: Properties of foils used in experiments

Foil	Thickness, μm	Purity, %
Ta	250	99,5
Al	250	99,0

The foil dimensions were initially selected to completely consume the aluminum in forming the intermetallic compound with alternating layers of partially unreacted Ta metal. The dimensions of the processed samples are in the shape of platelets as 10mm×10mm. Samples were cleaned with alcohol and dried before stacking process. In each stack was consisting of 4 tantalum and 3 aluminum foil as in shown Fig.2a. An initial pressure of 2.0 MPa is applied at room temperature to ensure good contact between foils (Fig.2b). Sintering process is applied in an open air, electrical resistance furnace at 850-900-950 ºC for 5 and 7.5 hours for each temperature.

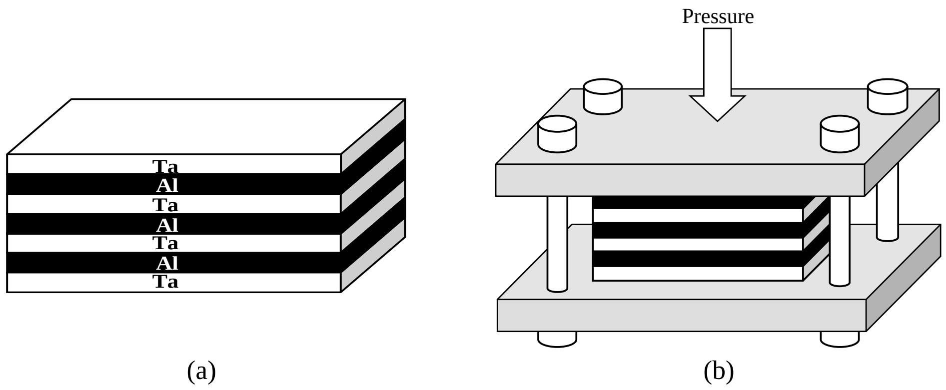


Figure 2:(a) Tantalum-Aluminum stack, (b) Schematic diagram of the synthesis apparatus. Image is not to scale and does not show the load frame (İpek, 2008, p.1262-1268)

Samples were ground and polished using standard metallographic techniques. Microstructure analyses of composites were performed with a JEOL JSM-5600 model scanning electron microscope (SEM). The presence of phases formed within the sintered samples was determined by Energy Dispersive Spectroscopy (EDS). Microhardness of composites were determined using a Leica WMHT-Mod model Vickers hardness instrument under an applied load of 300 g for intermetallic zone, and 100 g for metallic zone.

Results and Discussion

The typical microstructures of the MIL composites are shown in Fig.3-5. The presence of different regions indicates that different phases in the composites. In relatively low magnification (Fig.3.a) it is shown that foils are stacked properly. Furthermore, the laminated composites are well-bonded. There is a symmetric change from a light colored phase to another. Following the light colored phase there is a grey colored formation, subsequently, darker grey colored phase and in the center line a black colored phase. The light colored layers consist of unreacted Ta, which are separated by the apparently darker Al_3Ta-layers, as was identified by quantitative EDS analysis (Fig. 6). In metallic-intermetallic composites, interaction initially occurs at the interface then progress along the center of aluminum foil as well as along tantalum foil. But this progress reaction along tantalum is slower than that of in aluminum. As tantalum and aluminum react with each other, the impurities in aluminum or interface of Ta-Al, shifts in the front of the reaction. If the reaction ended before aluminum consuming these impurities deposited in aluminum, otherwise in centerline of the intermetallic. Because of this it is understood that the centerline is a problematic zone. As evidence in laminate composites centerline is one of the preferred zones for the formation of cracks.

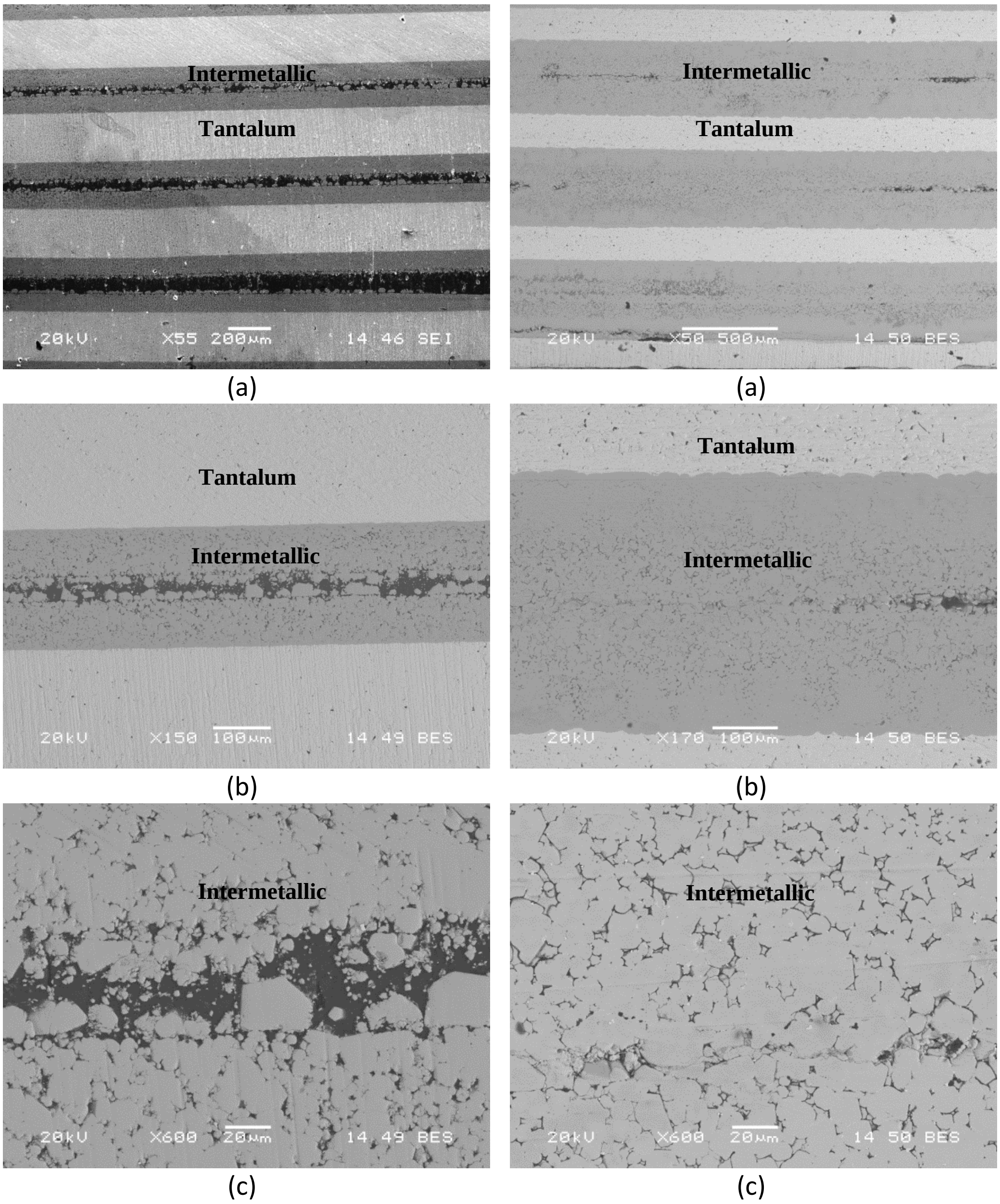
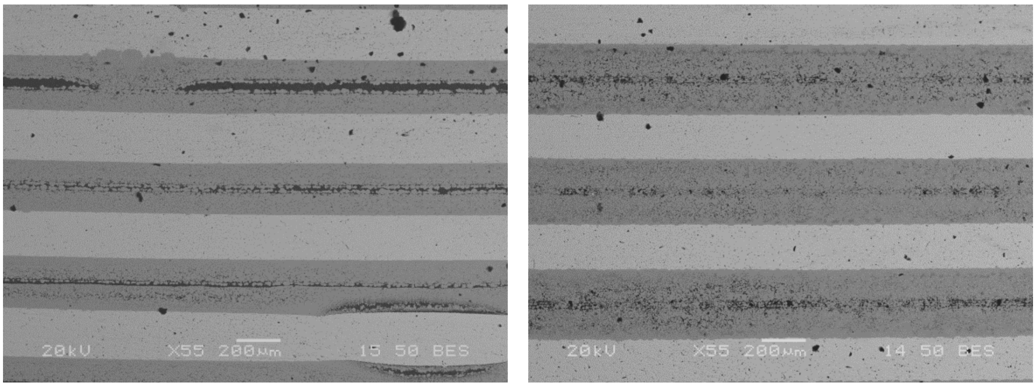


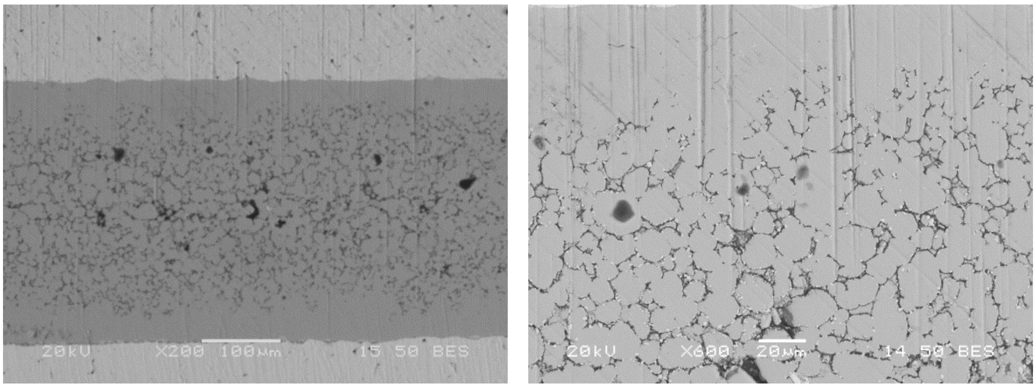
Figure 3:SEM micrographs of Ta-Al metallic–intermetallic composites in the left sintered at 850 °C,5 h (a)55X, (b)150X, (c)600X, in the right sintered at 850 °C ,7.5 h (a)50X ,(b)170X, (c)600X

It is clearly seen in metallic intermetallic Ta-Al composites sintered at 850 °C for 5 hours, the middle grey-black zone essentially formed by various phases such as residual aluminum, $TaAl_3$. As the sintering time increases from 5 hours to 7.5 hours, residual aluminum layer at the intermetallic centerline decreases. Moreover sintering process is better at relatively high temperature and time.



(a) (b)

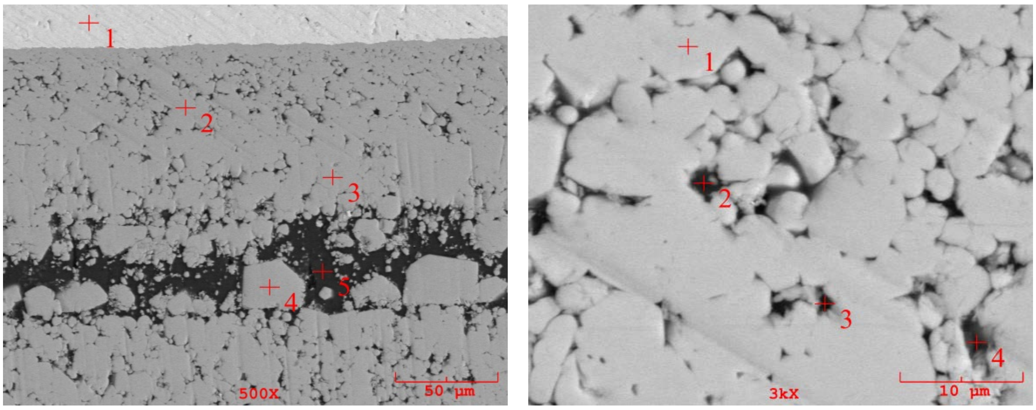
Figure 4: SEM micrographs of Ta-$TaAl_3$ metallic–intermetallic composites 900 °C,(a) 5 h-55X, (b) 7.5 h-55X



(a) (b)

Figure 5: SEM micrographs of Ta-$TaAl_3$ metallic–intermetallic composites 950 °C, 5 hour, (a)200X, (b)600X

From SEM micrographs for 850-900 and 950°C (Fig:3-5) it is seen that interface of metallic tantalum and tantalum-aluminum intermetallic is quite smooth. To form Ta-Al intermetallic, reaction between tantalum and aluminum initiate with nucleation in a number of different regions. These nucleuses grow and merge with each other. A whole aluminide layer form in this way. Stable metallic-intermetalic structure occurs when suitable temperature and time conditions are provided.



%Wt.	1	2	3	4	5
Ta	100	16.991	14.567	17.078	0.512
Al	0	83.991	85.433	82.922	99.488

(a)

%Wt	1	2	3	4
Ta	14.991	3.289	6.173	24.857
Al	85.009	96.711	93.827	75.143

(b)

Figure 6: SEM-EDS analysis of Ta-Al metallic-intermetallic composite sintered at 850ºC for 5 h.(a) 500X, (b) 3000X.

As in shown in Fig 6.a, spot 1 is located in the unreacted Ta layer and spots 2, 3 and 4 in the Al_3Ta layer with different positions. Some dark particles, identified as residual Al are also observed in the Al_3Ta layers. Furthermore, the laminated composites are well-bonded and remain some residual porosity in the final microstructure due to the relatively low temperature and time. Also in this EDS analyses show that, formation of intermetallic hasn't completed yet.

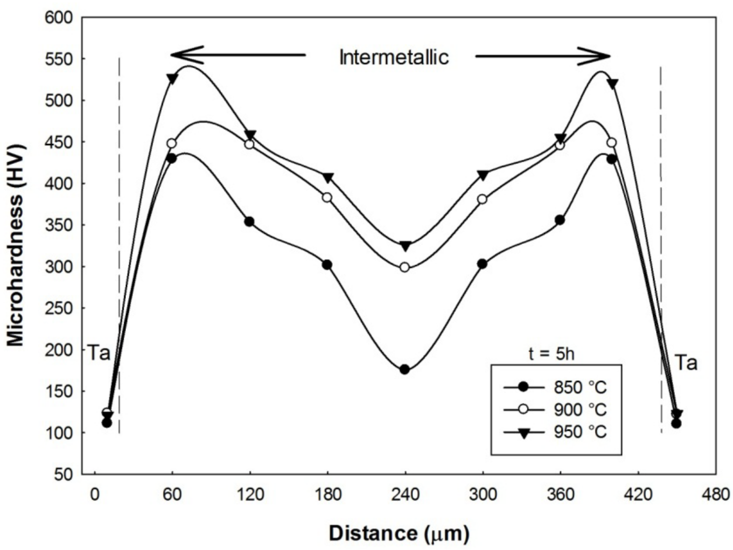


Figure 7: Hardness graphic of Ta-Al metallic-intermetallic composites sintered at 850, 900, 950 ºC, 5 hours.

The ceramic-like aluminide phases (Al_3Ti or Ni_3Al) give high hardness and stiffness to the composite, while the unreacted Ti or Ni provides the necessary high strength, toughness and ductility for the system to concurrently be flexible. The multi-layered structure of the composite allows for variations in the layer thickness and phase volume fractions of the Al and Ti or Ni components simply through the selection of initial thickness, which consequently allows for the optimization of mechanical and thermal management properties for practical application (Peng, 2005, 309-318).

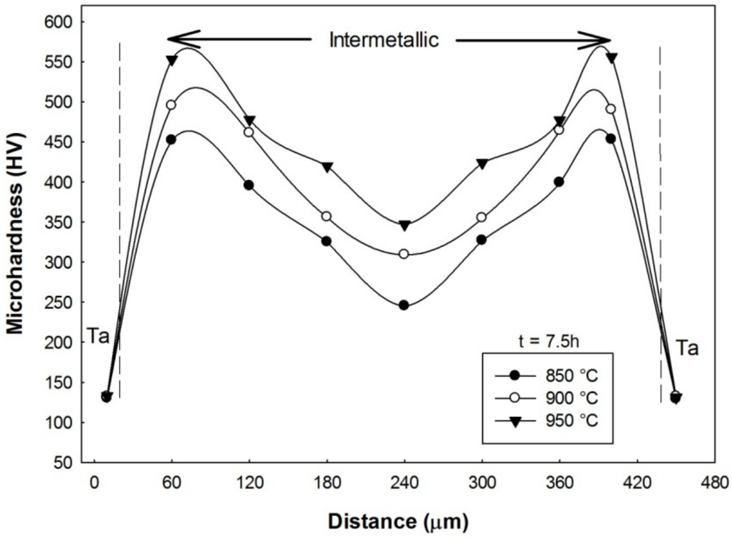


Figure 8: Hardness graphic of Ta-Al metallic-intermetallic composites sintered at 850, 900, 950 ºC, 7,5 hours.

The microhardness profile measured for several layers of the composite is shown in Fig.7, 8. As known, hardness value of metallic tantalum and its intermetallic structure is quite different. The average microhardness values for tantalum, aluminum and $TaAl_3$ are 120, 475 and 45 HV, respectively. While the hardness of the intermetallic layer is highest at the border $TaAl_3$-Ta, it gradually decreases toward the border $TaAl_3$-Al.These results are in good agreement with the literature. In addition, relatively high temperature and time is also very effective on sintering metallic-intermetallic laminate composite. As time and temperature increase, the hardness values raise because of bonding in the interface improved and residual aluminum started the consuming.

Conclusions

The conclusions of this research can be summarized as follows:

- By controlling the duration of the reactive foil sintering process, composites can be fabricated in which a tailored amount of residual aluminum remains at the intermetallic centerline. The result is a well-bonded composite with a high degree of microstructural control.
- Ta–Al_3Ta metal–intermetallic laminate (MIL) composites have been successfully synthesized by reactive foil sintering technique in open air at 850-900 and 950 ºC for 5 and 7.5 hours under 2 MPa pressure. The laminated structure is well-bonded, nearly fully dense.
- Microstructural characterization by, SEM and EDS indicates that Al_3Ta is the only intermetallic phase. Tantalum aluminide phase occurs due to the thermodynamics of the reaction between Ta and Al. The existence of liquid Al phase plays important roles in the nucleation and growth of Al_3Ta particles and the eventual formation of continuous alternative Al_3Ta intermetallic layers.
- The mechanic properties of hardness of the fabricated laminated composites were examined. Whereas the hardness of metallic aluminum and tantalum respectively is about 45, 120 HV, hardness of intermetallic zone is approximately 450-500 HV. The results showed that as time and temperature increase, the hardness values raise because of bonding in the interface improved and residual aluminum started the consuming.

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