

## Active Brazing of Tantalum – to – Tantalum

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### Abstract

The aim of this study is to start a work about the bonding mechanism of tantalum -to- tantalum brazed by active filler metal alloy which basely stand on using copper or silver with active element such as titanium. Brazing will happen under inert gas (argon). Ag4Wt% Ti,Cu8wt %Ti , and the eutectic Ag 26wt% Cu 4 wt % Ti are the active filler metal which are used , and the bonding phases at the filler metal / tantalum interface line almost contain ( Ti Ta<sub>2</sub>O<sub>7</sub> ) and ( Ti<sub>0.51</sub> Ta<sub>0.49</sub> O<sub>2</sub> ) . Which means that active element ( titanium ) shares oxygen with tantalum to make this bond as a cheap filler .

### لحام المونة الفعال للتاليوم

#### المستخلص

ان هدف هذه الدراسة هو البدء في معرفة الية الربط لوصلة لحام التنتاليوم باستخدام سبيكة المونة الفعالة والتي تعتمد على عناصر مثل النحاس ، الفضة والعنصر الفعال ( التيتانيوم ) . يجري اللحام بالمونة في ظروف خاملة باستخدام غاز الاركون . المونة المستخدمة هي الفضة مع نسبة وزنية 4% من التيتانيوم ، النحاس مع نسبة وزنية 8 % من التيتانيوم ، وايوتكتك الفضة والنحاس ( Ag 26wt% Cu 4 wt % Ti ) مع نسبة وزنية للتيتانيوم 4 % ، اما طور الربط فقد وجد بأنه دائماً يكون على شكل ( Ti Ta<sub>2</sub> O<sub>7</sub> ) أو ( Ti<sub>0.51</sub> Ta<sub>0.49</sub> O<sub>2</sub> ) حيث ان التيتانيوم يتقاسم ارتباطه مع التنتاليوم مع الاوكسجين . وفي النهاية فإن هذه الوصلة تعتبر قليلة الكلفة .

## 1. Introduction

From literature survey between 1960 to 1997 as shown in Table (1) [ 1 – 6 ] it was pointed that , refractory metals didn't have the data base that exists in the literature . Because of specific details on brazing equipment , such as vacuum levels , type of vacuum equipment , quality of inert gases and heat brazing cycles are not reported in most of the development work on the refractory metals [5] . Tantalum can be brazed with nickel-base filler metals , such as ( Ni-Cr-Si ) alloys . However , tantalum and nickel form brittle intermetallic compounds , such as ( Ni<sub>2</sub>Ta ) in brazed joint [ 1 , 2 , 3 ] . In 1985 F.M Hosking studied sodium compatibility of refractory metal alloys – to – A ISI 304L stainless steel joint [7] . He brazed ( Mo , Re , Ta , and W ) alloy to AISI 304L stainless steel with nickel base filler metal in two forms ( metallic glass and powder ) by vacuum furnace . Silver – copper filler metals can produce joints with useful strength at room temperature [8] . The active-metal modifications ( Ti additions ) to ( Ag – Cu ) and ( Au – Ni ) alloys may be useful for tantalum joining particularly if residual oxide on the surface presents a wetting problem [5] .

Ta – V – Ti and Ta – V – Cb alloys given in Table (2) [3] , may be suitable for certain high temperature applications . Brazing should be done in vacuum of  $10^{-4}$  torr or better , but there is affinity to vaporize titanium and vanadium from molten filler metal during the brazing cycle because of their high vapor pressure [3] . Also pure titanium and titanium alloys might be suitable for brazing tantalum . In 1977 , Scientific Oak Ridge National Laboratory ( ORNL ) has been instrumental in the development of brazing filler metals for applications at temperature above ( 1000 °C ) [6] . The filler metals and brazing temperatures employed in the development of these alloys are presented in Table (3) [6] . They used tantalum as refractory , aluminum oxide samples as ceramic were bonded with vacuum of  $10^{-6}$  torr . The excellent flowability of these alloys on tantalum was recorded . The aim of this study is to recognize the Ta – to – Ta joint characteristics by usual brazing filler containing titanium depending on optical microscoping ( OM ) , and X-ray diffraction ( XRD ) testing .

**Table ( 1 ). Filler metals used in tantalum brazing between 1960 to 1997[1-6].**

| <b>Filler Metal</b>   | <b>Service Temperature<br/>(°C )</b>  | <b>The References</b>   |
|---|---|---|
| <u>Nickel – Base</u><br>Ni-Cr-Si alloys   | 981 ≥   | [1,2,3]   |
| <u>Copper –Base</u><br>Cu-Au<br>Cu-Sn<br>Cu-Ti  | Low Temperature<br>Not Specified<br>Not Specified   | [4,1,2,3]<br>[3]<br>[3]   |
| <u>Silver –Base</u><br>Ag-Cu<br>Ag-Cu-Ti  | Room Temperature<br>Not Specified   | [3]<br>[5]  |
| <u>Gold –Base</u><br>Au-Ni<br>Au-Ni-Ti<br>Au-Cu   | Low Temperature<br>Not Specified<br>Low Temperature   | [5,1]<br>[5]<br>[5,1]   |
| <u>Tantalum –Base</u><br>Ta-V-Ti<br>Ta-V-Nb   | 1482 ≥<br>1482 ≥  | [5,3]<br>[5,3]  |
| <u>Titanium-Base</u><br>Pure Ti<br>Ti-Cr<br>Ti-V<br>Ti-V-Be<br>Ti-Zr-Be<br>Ti-V-Cr<br>Ti-Zr-Ge<br>Ti-Zr-Ta<br>Ti-Zr-Nb<br>Ti-Zr-Cr<br>Ti-Zr-B<br>Ti-V –Nb<br>Ti- V-Mo | Not Specified<br>1927 ≥<br>Not Specified<br>1927 ≥<br>Not Specified<br>1000 <<br>1000 <<br>1000 <<br>1000 <<br>1000 <<br>1000 <<br>1000 <<br>1000 <<br>1000 < | [2]<br>[3]<br>[5]<br>[3]<br>[2]<br>[6]<br>[6]<br>[6]<br>[6]<br>[6]<br>[6]<br>[6]<br>[6] |
| <u>8-Platinum-Base</u><br>Pure platinum<br>Pt-Ir<br>Pt-Rh   | Not Specified<br>Not Specified<br>Not Specified   | [2]<br>[2]<br>[2]   |
| <u>9-Zirconium-Base</u><br>Zr-Nb-Be<br>Zr-Ti-V  | Not Specified<br>Not Specified  | [2]<br>[5]  |
| <u>10-Palladium-base</u><br>Pure Palladium  | Not Specified   | [2]   |

**Table (2). Typical brazing filler metal for tantalum alloys for service temperature up to 1482c° [3].**

| Filler metal composition<br>weight percent | Temperature (°C ) |         |
|--|-------------------|---------|
|  | Brazing           | Remelt  |
| 10Ta-40V-50Ti                              | 1760              | 2381.10 |
| 20-Ta-50V-30Ti                             | 1760              | 2381.10 |
| 25Ta-55V-20Ti                              | 1825.5            | 2204.40 |
| 30Ta-65V-5Ti                               | 1825.5            | 2381.10 |
| 5Ta-65V-30Nb                               | 1815.6            | 2281.11 |
| 25Ta-50V-25Nb                              | 1871.1            | 2481.90 |
| 30Ta-65Vv-5Nb                              | 1871.1            | 2281.11 |
| 30Ta-40V-30Nb                              | 1926.7            | 1981.90 |

**Table (3) .Ternary system in which filler metal have been developed and their recommended applications [6].**

| Filler<br>Metal<br>System | Approximate<br>Brazing<br>Temperature<br>(°C ) | Materials            |          |                                |
|---------------------------|--|----------------------|----------|--------------------------------|
|                           |  | Refractory<br>metals | Graphite | Al <sub>2</sub> O <sub>3</sub> |
| Ti-V-Cr                   | 1550-1650                                      | X (a)                | X        | X                              |
| Ti-Zr-Ta                  | 1650-2100                                      | X                    | X        | -                              |
| Ti-Zr-Ge                  | 1300-1600                                      | X                    | X        | -                              |
| Ti-Zr-Nb                  | 1600-1700                                      | X                    | X        | -                              |
| Ti-Zr-Cr                  | 1250-1450                                      | X                    | - (b)    | -                              |
| Ti-Zr-B                   | 1400-1600                                      | X                    | -        | -                              |
| Ti-V-Nb                   | 1650   | X                    | -        | -                              |
| Ti-V-Mo                   | 1650   | X                    | -        | -                              |

(a) ( X ) =Applicable for joining .

(b) ( - ) = Not applicable for joining .

## 2. Experimental Work

A 0.15 mm sheet thickness of pure tantalum manufactured by Pansee Metalwork GmbH was used as a base metal in this work . High purity powders of copper , silver , and titanium metals were used to prepare the active paste filler metal alloy . The paste prepared by manual mixing

( for one minute ) , the suitable mass of filler with about (  $0.5 \text{ mm}^3$  ) of glycerin and (  $1 \text{ mm}^3$  ) of alcohol ethanel to form the paste , which is suitable for one butt joint assembly . The suitable gap size is about 0.1 mm , and the overall mass is about 0.6 gram , which was used for all testing samples . The dimension of tantalum samples are  $25 \times 25 \times 0.15 \text{ mm}$  . More than nine tantalum to tantalum butt joint assembly were used , the joining process done by three fillers , metal alloy types ; Ag4wt%Ti ( Ag4Ti ) , Cu 8 wt%Ti ( Cu 8Ti ) , and eutectic Ag 26 wt%Cu4wt% Ti . ( AgCuTi ) . For each filler, three joint assemblies were carried out , one joint for microstructure testing , and the others for X-ray diffraction testing . Furnace brazing with argon , as a protection gas used to complete the brazing process . A muffle furnace type with a controlled-argon-atmosphere is prepared with gas flow rate of about 1-3 L/min in a stainless steel retort . The brazing temperatures are ,  $950 \text{ }^\circ\text{C}$  ,  $1050$  , and  $1100 \text{ }^\circ\text{C}$  for the fillers AgCuTi , Ag4Ti , and Cu8Ti respectively . The holding time at brazing temperature is about five minutes . Grinding and polishing processes were carried out to prepare the microstructure testing samples , while the broken samples at the bonding interface will be used for XRD testing after grinding process to the two broken sides , to be examined by Philips-PW1840 X-ray diffractometer . The X-ray tube target was copper (  $\lambda$  for  $\text{Cu}\alpha = 1.5405 \text{ \AA}$  ) , the filter is nickel . The 2 $\theta$  ranges taken was from  $10^\circ - 90^\circ$  , in the speed of 3 degree/min . Identification of phases was based on matching with powder diffraction files ( JCPDS ) [9] . The welded samples are fixed by simple two plates of molybdenum (  $50 \times 50 \times 0.25 \text{ mm}$  ) with two screws to maintain the joint in proper dimensions .

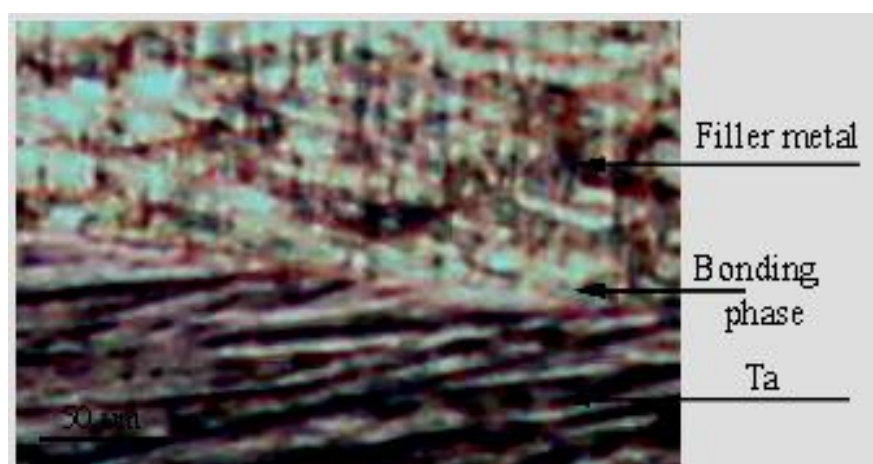
### 3. Results and discussion

Optical micrographs for a cross section brazed by Ag4Ti filler metals in Figure (1) show mutual dissolution of tantalum and filler metal in a wave form . This may be explained by the solubility of B-Ti in tantalum [9] . Closer scrutiny revealed formation of a distinct layer adjacent to the base metal and migration of separated particles from this layer to central region of filler metal as in Figure (2) . This morphology may represent the aggression and disruption of the bonding layer by active filler metal . Also dark points were observed in the braze joint , this may refer to the titanium oxide formation duo to the bonding mechanism by active filler metal alloy [10,11] .

The XRD results for Ta to Ta assemblies brazed by Ag4Ti as in Figure (3) show that ; the bonding phases are (  $\text{Ti Ta}_2 \text{O}_7$  ) and (  $\text{Ti}_{0.51} \text{Ta}_{0.49} \text{O}_2$  ) , which refer to the oxide layer at the surface of tantalum , and the bond created by the action of tantalum sharing its passivity oxide

film before combined with titanium [12] . The presence of Ag and Ti phases refers to (Ta-Ti) solid solution . While the separated phases of Ti and Ta refer to the limited solubility of Tantalum in B-Ti and tend to formation of intermetallic phases .

The brazing of Ta to Ta with silver – 4wt% Ti filler metal alloy results in an improved joining with low cost as compared with that brazed by expensive filler metal as in Tables 2 and 3 and part of Table (1) .



**Figure (1) .Optical micrograph for ( Ta – to – Ta ) assembly brazed by Ag4Ti filler metal .**

Figure (4) shows the optical micrographs for Ta – to Ta joint which was brazed by Cu 8 Ti , in which a mutual dissolution of tantalum and active filler metal was remarked as a waved interface like what was happened with the joint by Ag 4 Ti . However the bonding mechanism completed by the existing of  $Ti Ta_2 O_7$  phase only as shown in Figure (5) .

Using of eutectic AgCuTi filler metal alloy to braze Ta – to – Ta joint results in a good joining . Optical micrographs for AgCuTi filler / tantalum interface as in Figure (6) , shows a formation of a distinct layer adjacent to base metal . This layer has expanses in roots form , were expanded in eutectic braze metal as in figure 6 – a . This may refer to the formation of more than one bonding phase . The closer scruting of interface line shows separation part of roots and many particles from bonding phase . This morphology may represent the dissolution or erosion of tantalum by active filler . The XRD chart for this joint Figure( 7) shows that the bonding phases are ;  $Ti Ta_2 O_7$  ,  $Ti_{0.51} Ta_{0.4}$  ,  $O_2$  ,  $Ti Ta_{13} O_{47}$  , and  $Ti O$  phase . The last phase explains the good bonding structure which means that the active element ( titanium ) forms a good bonding layer [11] .



Figure (2) . (Ta – to - Ta ) assembly brazed by Ag<sub>4</sub>Ti filler metal .

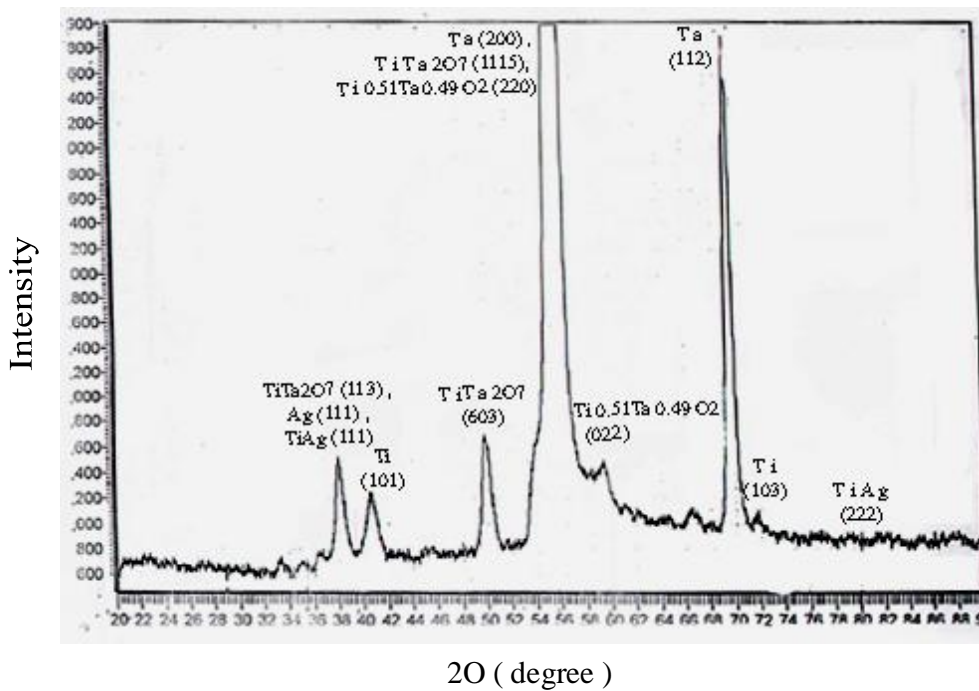


Figure (3) . X-ray diffraction chart for ( Ta – to – Ta ) assembly brazed by Ag<sub>4</sub>Ti filler metal .

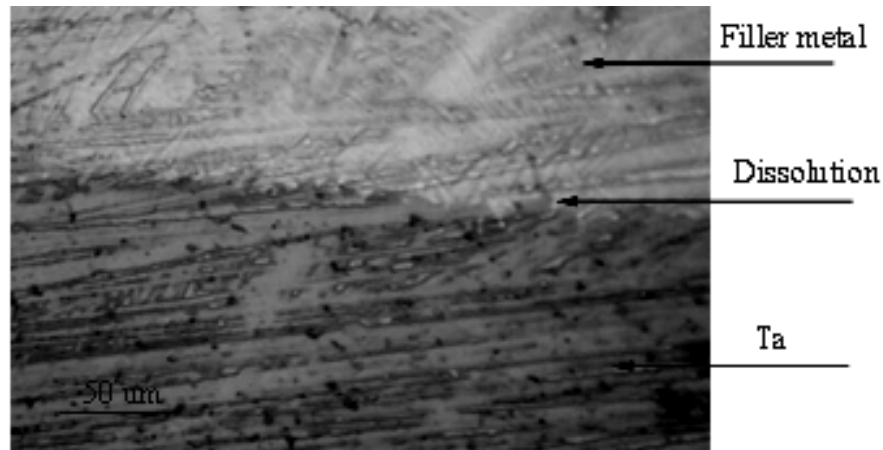


Figure (4). Interface of (Ta –Ta) assembly brazed by Cu<sup>Ti</sup> filler metal.

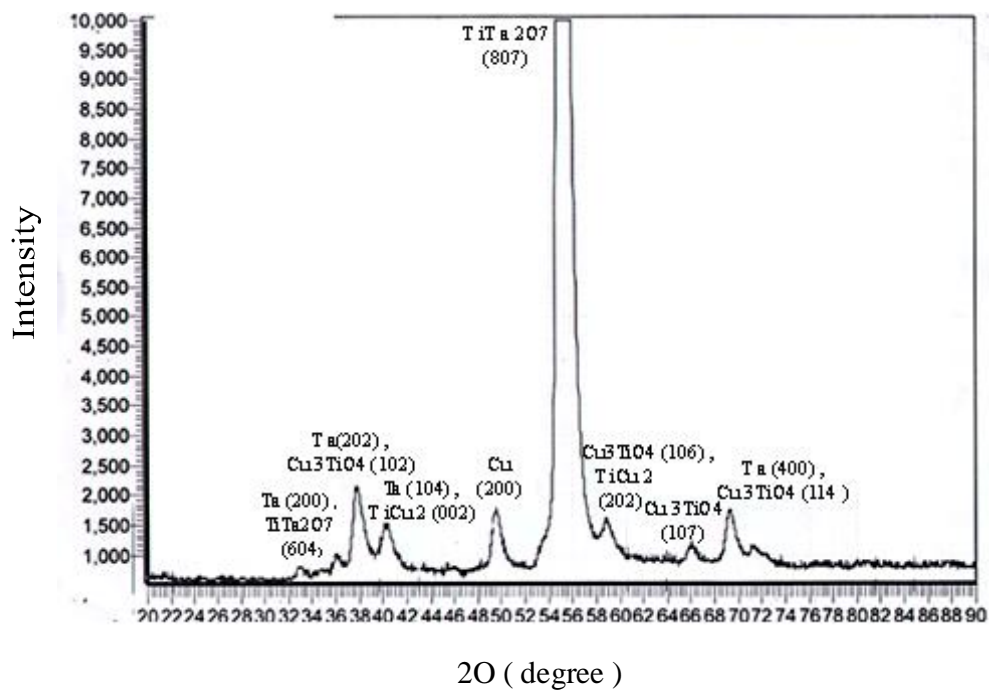


Figure (5) . X-ray diffraction chart of (Ta – to – Ta) assembly brazed by Cu<sup>8Ti</sup> filler metal.



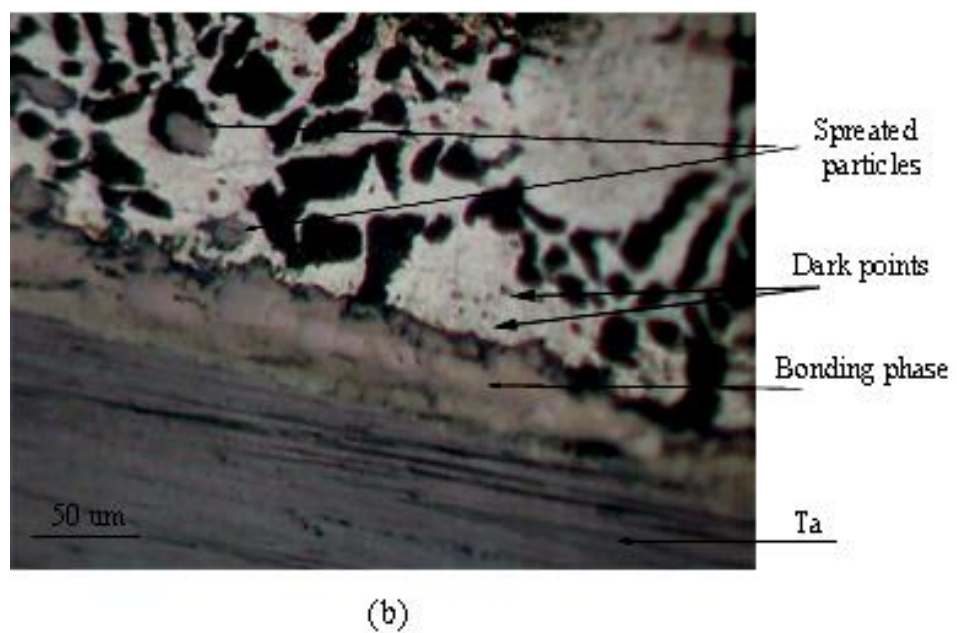
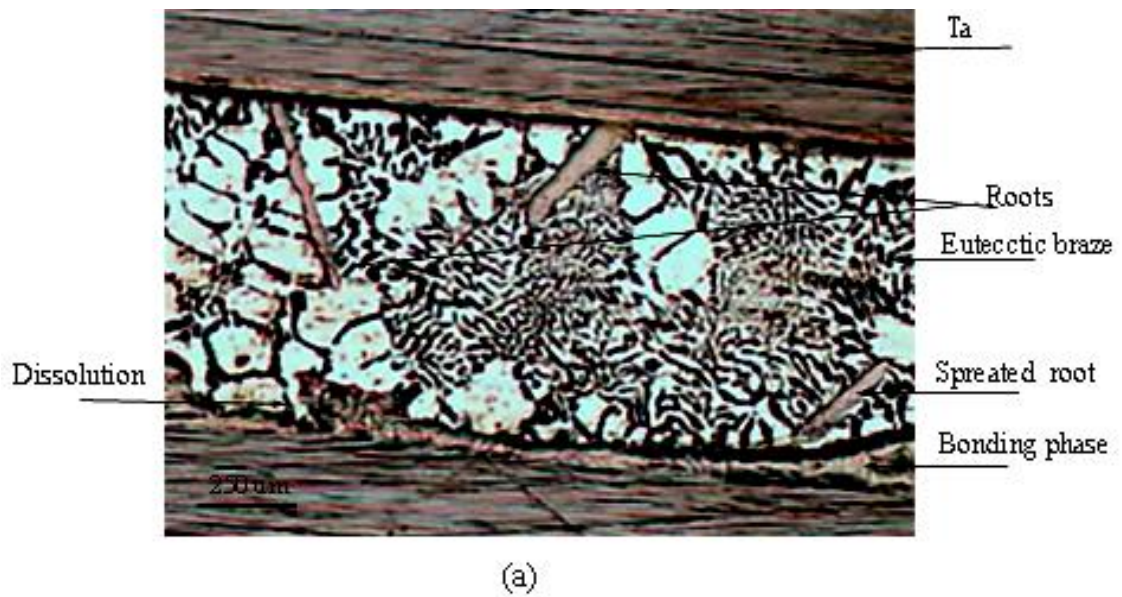


Figure ( 6 ). (a and b ) Represents optical micrographs for ( Ta – to – Ta ) assembly brazed by eutectic AgCuTi filler metal .

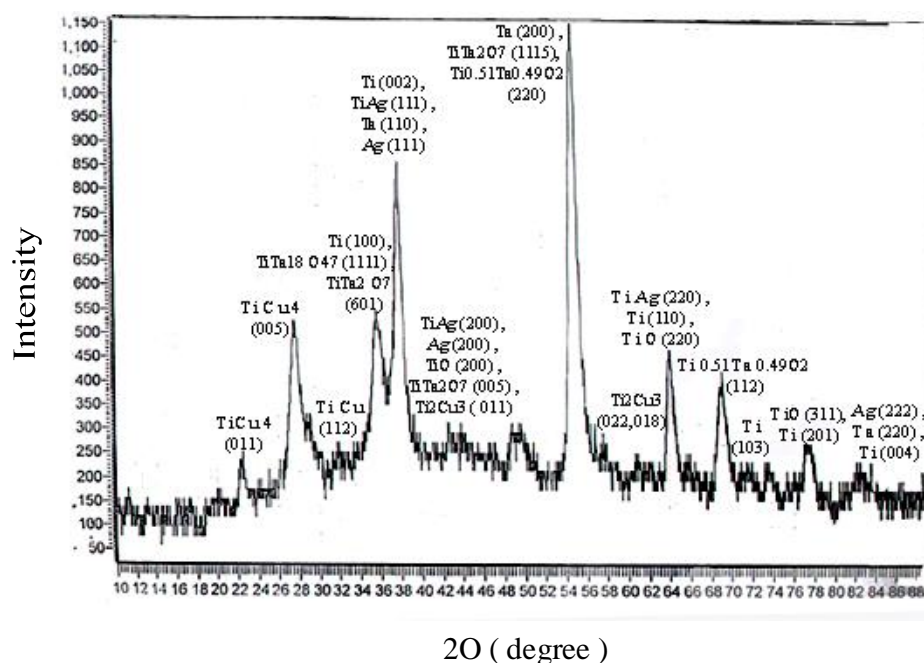


Figure ( 7 ) . X- ray diffraction chart for (Ta – to – Ta ) assembly brazed by eutectic AgCuTi filler metal .

## 4. Conclusions

1. The Tantalum to tantalum brazed assembly by Ag4Ti exhibits good joining with aggression of tantalum and dark point were observed in brazed region .
2. The Tantalum to tantalum brazed assembly by Cu8Ti exhibits additional improvement in joining with low cost as compounded with high active filler metal alloy .
3. The bonding phases in most assemblies brazed by CuTi , AgTi and AgCuTi filler metal are  $TiTa_2O_7$  and  $Ti_{0.51}Ta_{0.49}O_2$  with good interface bonding layer with TiO for using autectic filler metal alloy .

## 5. References

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