C. Z. Xia*, Q. H. Liang, J. S. Zou and X. P. Xu

Microstructural Investigation of W-Cu Composite and 1Cr18Ni9 Stainless Steel Brazed Joint with Ag-Cu Filler Metal

Abstract: Tungsten-copper (W-Cu) composite and 1Cr18Ni9 stainless steel were brazed with Ag-28wt%Cu brazing filler metal in a vacuum furnace, via controlling brazing temperature 855–865 °C and holding time 30 min. Microstructure, microhardness, four-point bend strength, element distribution and phase constituents near interface of the brazed joint, were investigated by a series of standard technique. Results indicated that Ag-Cu eutectic phase near W-Cu composite side and the bulk phase near 1Cr18Ni9 steel side were formed in the brazing seam region. Microhardness of the eutectic phase was low, while that of the bulk copper-rich phase was high. Four-point bend strength was as high as 576 MPa, and the fracture occurred at interface near 1Cr18Ni9 steel side. On the brazed interface, Cu and a little W element were transited from W-Cu composite into the brazing seam region, as same as Fe from 1Cr18Ni9 steel. Ag contained in the brazing filler metal was diffused into the W-Cu composite. Element concentration of Ag was high at interface of both two base metals, which was helpful to promote the metallurgical reaction. Phase constituents of the brazed joint were Cu(Ag, Fe), Ag(Cu) and a little of $Ag_{0.7}Fe_{0.3}$ and $FeCu_4$, formed in the metallurgy of Fe from 1Cr18Ni9 steel and the Ag-Cu brazing filler metal.

Keywords: tungsten-copper composite, vacuum brazing, bend strength, element distribution, microstructure

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1 Introduction

Tungsten-copper (W-Cu) composite has a series merit of good thermal conductivity, thermal shock resistance, dimensional stability (low coefficient of linear expansion) and high temperature strength. It is a very promising material in high temperature and good thermal stability condition, which has been widely used in field of cooling and heat sink material of high-power microwave devices, heavy-duty electrical contacts and plasma facing components [1–4]. However, the poor oxidation resistance at high temperature of the W-Cu composite constrained its application range in components serviced in high temperature condition. Joining W-Cu composite and stainless steel to form compound structure, can take advantage of the high thermal conductivity, thermal shock resistance of W-Cu composite and high temperature oxidation resistance and corrosion resistant benefits of stainless steel, to improve integration performance of the compound structure.

Three difficulties have been identified in welding of W-Cu composite and stainless steel. First, linear expansion coefficient, melting point and thermal conductivity of these two materials were quite different. Second, in process of preparation and service, compound structure of W-Cu composite and stainless steel underwent circulating heat loads [5]. Stress concentration was easily formed in area of the joining interface, increasing fracture tendency and reducing mechanical properties. Third, it was sensitive to gas impurities and micro holes was easy to form in the interface of W-Cu composite joint, inducing brittleness phase formed and influencing gas tightness and carrying capacity.

Currently, considerable interest has been generated in joining and compounding of W-Cu composite. Related to joining of W-Cu composite, technological methods contained brazing [6–9], friction welding [10], liquid phase sintering [11, 12], hot isostatic pressing and diffusion bonding [13, 14], were adopted by researchers. Hiraoka et al. [6, 7] has performed the brazing of W-19vol%Cu composite using Bag-8 as the brazing metal, and researched the fracture characteristics of the brazed W-Cu composite joint. A relatived high bending yield strength of the brazed joint was obtained.

In this study, W-Cu composite and stainless steel were brazed in a vacuum of 10^{-3} Pa using Ag-Cu filler metal and an integrated W-Cu/stainless steel joint with good combination was formed. Microstructure, bending strength, microhardness distribution, elements transition and phase constitution in the brazing seam region were analyzed by means of metallography, mechanical testing machine, scanning electron microscope and X-ray diffraction. The results provide a favorable basis for further studies on joining W-Cu composite with steel and for application of the compound structure.

2 Experimental

The commercial materials used in this study were W-Cu composite and 1Cr18Ni9 stainless steel. The tungsten-copper composite was W-45wt%Cu and its microstructure was shown in Fig. 1. Chemical composition of 1Cr18Ni9 steel was shown in Table 1. Adopted filler metal was Ag-28wt%Cu, foils with a thickness of 250 μm.

The W-Cu composite and 1Cr18Ni9 steel samples were machined into dimensions of 20 mm × 20 mm × 5 mm and Ag-Cu filler metal into 20 mm × 5 mm foils. Then the oxidation film and greasy dirt on surface of the substrates and the filler metal were eliminated by $400^{\#}$ sandpaper and cleaned in ultrasonic using alcohol before being vacuum brazed. And the surface to be brazed was flat with a relative level of roughness, avoiding too much filler metal lost.

Finally, the test samples were assembled by sequence of W-Cu composite, Ag-Cu filler metal and 1Cr18Ni9 steel in a special fixture (as shown in Fig. 2). In the vacuum brazing process, a stationary pressure of 40 KPa was imposed on the surface of 1Cr18Ni9 steel, for making the filler metal infiltrate and spread better in the gap of the base metals. Process parameters during the vacuum brazing were: brazing temperature T = 855 to 865 °C, holding time t = 30 min, vacuum level superior to 6×10^{-3} Pa.

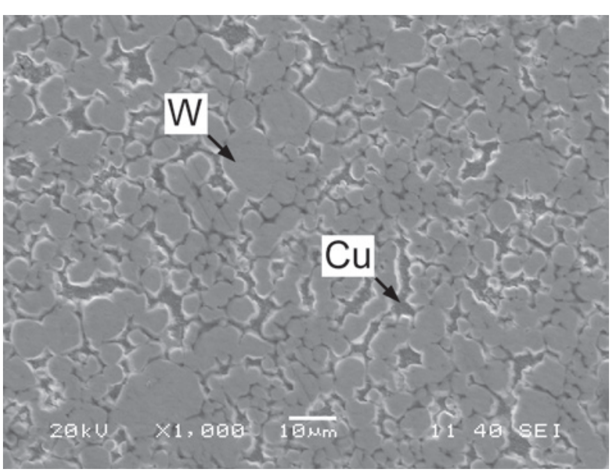

Fig. 1: Microstructure of W-Cu composite

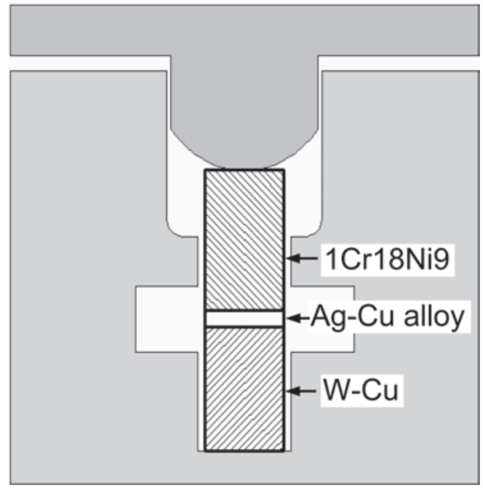

Fig. 2: Sketch of clamp used in the brazing process

Samples were cut from the brazed joint using electro discharge machining. Then they were ground via a series different type sand paper, polished and finally etched with a mixed solution of HCl, HF and HNO_3(80:13:7). Microstructural feature and microhardness distribution near the brazing interface of W-Cu/1Cr18Ni9 joint were analyzed via metallographic microscope, JSM-6480 scanning electron microscope (SEM) and microsclerometer. Element composition and phase construction on the interface were measured by means of energy dispersive spectrum (EDS) and X-ray diffraction (XRD-6000), respectively.

3 Results and discussion

3.1 Microstructure near interface of the brazed joint

W-Cu composite composed of tungsten and copper was a monomer uniformly mixed tissue of two phases, neither formed any solid solution nor metallurgical reaction

Table 1: Chemical compositions of 1Cr18Ni9 stainless steel (wt%)

C	Cr	Ni	Si	Mn	S	P	Ti	Co	Fe
0.018	17.50	9.27	0.59	0.97	0.0043	0.0278	0.13	0.14	Bal.

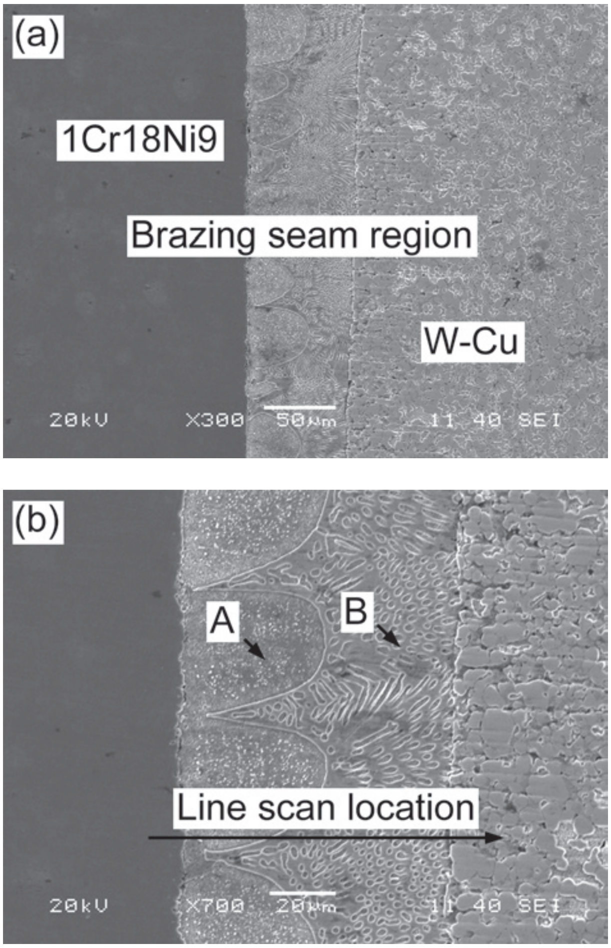

Fig. 3: Microstructure of the brazing joint of W-Cu composite and 1Cr18Ni9 stainless steel

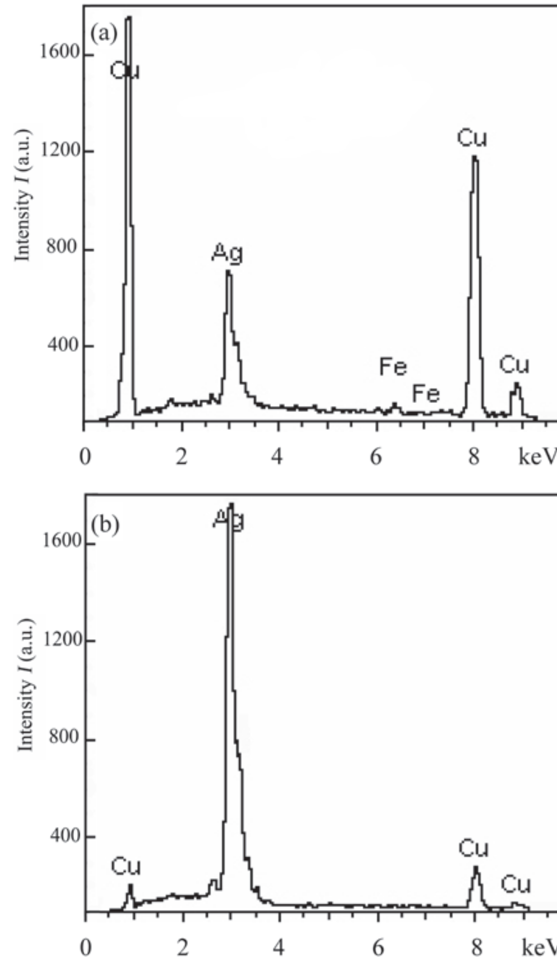

Fig. 4: EDS results of different area in brazing seam region: (a) A point; (b) B point

occurred to form intermetallic compounds (IMC). Microstructure near interface of brazed joint of W-Cu composite and 1Cr18Ni9 steel was shown in Fig. 3. The microstructure was characterized different to conventional metal.

While W-Cu composite and 1Cr18Ni9 steel being brazed with Ag-28Cu filler metal, the brazing seam region and the substrates formed a good metallurgical bonding with a width about 0.07~0.1 mm. A slight diffusion layer existed between the brazing seam region and the substrates, no obvious micro cracks or hole defects observed.

The brazing seam region of joint of W-Cu composite and 1Cr18Ni9 steel joint mainly contained eutectic phase near W-Cu composite side and bulk phase near 1Cr18Ni9 steel side. EDS analysis of A and B point with different morphology in the brazing seam region was carried out and the results was shown in Fig. 4. According to the binary phase diagram of Ag-Cu [15], phase of A point was a solid solution of Ag dissolved in Cu phase, with a chemical composition of Cu 75.85%, Ag 22.79%, Fe 1.36% (wt%, shown in Fig. 4a). A little iron element transited into the brazing seam region and formed Cu(Ag, Fe) solid solution. The phase of B point was a solid solution of Cu element dissolved in the eutectic Ag-Cu phase, with a chemical composition of Cu 17.74%, Ag 82.26% (wt%, shown in Fig. 4b).

3.2 Bend testing and microhardness

3.2.1 Four-point bending strength

Bending yield strength of the obtained joint was evaluated using a four-point bending test method at room temperature. The bending test samples were machined by electro discharge machining with dimensions of 5 mm × 5 mm × 40 mm and tested on an electronic mechanical testing machine (CMT5205) using a special fixture with a crosshead speed of 0.5 mm/min. Sketch of four-point bending

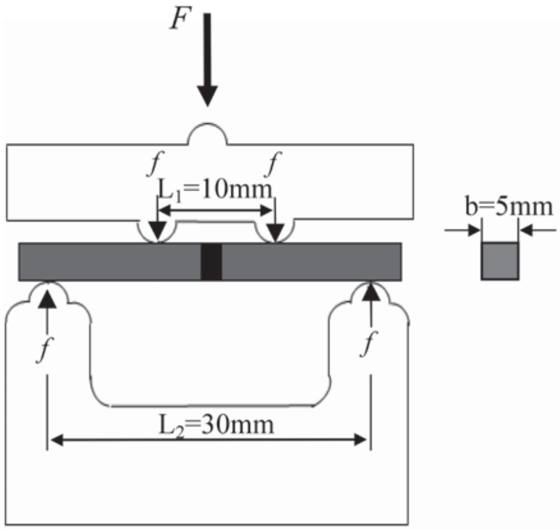

Fig. 5: Sketch of four-point bending test

test was shown in Fig. 5. From a load-displacement curve, maximum (σ) strength was calculated via the following equation.

$$\sigma = \frac{3Pa}{bh^2}$$

In this equation, P is the load at the maximum point. $2a$ is the span of supporting pins. b (mm) and h (mm) are the specimen width and thickness, respectively. After calculated, the four-point bending strength of the obtained joint was 576 MPa with an average maximum load 2.4 kN. Furthermore, the bending fracture occurred at the brazing seam region near the 1Cr18Ni9 stainless steel side, illustrated that a high brazing strength was obtained for the W-Cu composite and 1Cr18Ni9 stainless steel joint.

3.2.2 Microhardness

For further analyzing influence of microstructure of brazing interface on joint performance, microhardness distribution was tested, as shown in Fig. 6, by MH-5 microsclerometer with the parameters: 50 gf loading and a load time of 10 s.

Because of the special monomer mixed tissue, microhardness of W-Cu composite performed a wide level 130 to 300 HM, while microhardness of 1Cr18Ni9 steel was 200 to 220 HM. And microhardness (65 to 135 HM) of brazing seam region was lower than that of the substrates. Meanwhile, the width of brazing seam was small, formed a special structure of "hard clip soft". Microhardness of eutectic phase in the brazing seam near W-Cu composite side

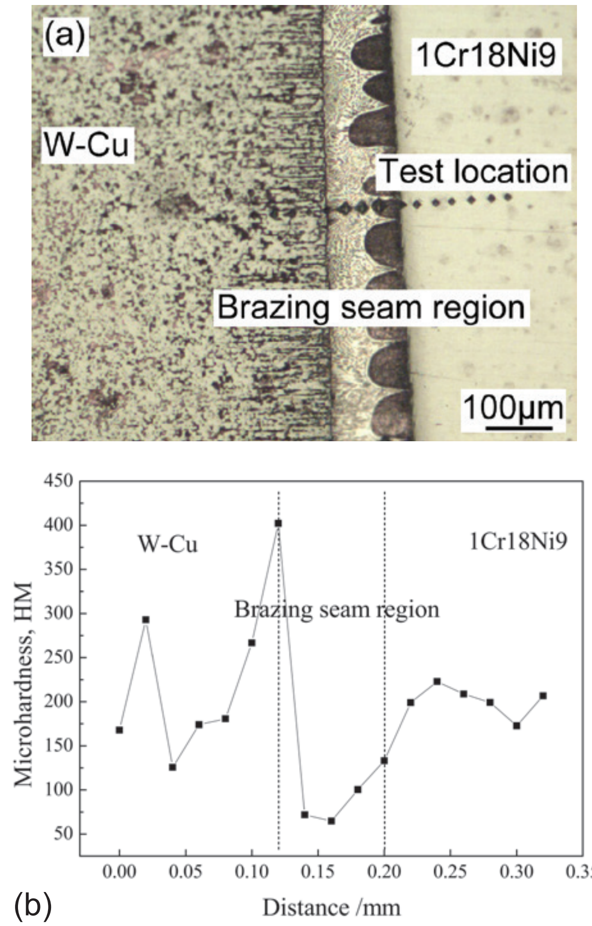

Fig. 6: Microhardness distribution near interface of the brazed joint: (a) Measured location; (b) Microhardness profile

was low (65 to 72 HM), while that of the bulk copper-rich phase near 1Cr18Ni9 steel side was relatively high (100 to 135 HM). Tendency of microhardness distribution in the brazing seam was invovled to different elements distribution. The eutectic phase was rich in silver element and the bulk phase was rich in copper element.

Low microhardness in the brazing seam showed that no obvious brittle phase formed, so can assure that the metal in brazing seam region has good plasticity and toughness. Meanwhile the bending strength of the obtained joint was high, so a compound joint with good toughness and strength performance could be obtained.

3.3 Elements distribution near the brazing interface

Elements distribution plays an important role in determining microstructure feature and performance of the brazed joint. Elements distribution across interface of the obtained joint of W-Cu composite and 1Cr18Ni9 steel was

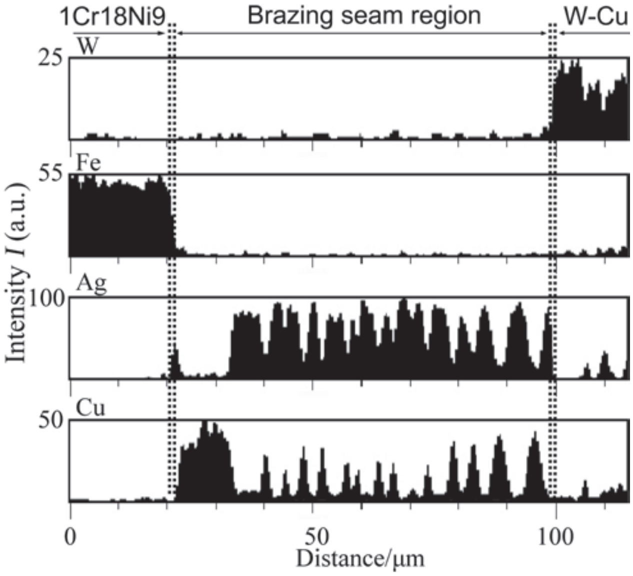

Fig. 7: Elements distribution across the interface of W-Cu/1Cr18Ni9 brazed joint

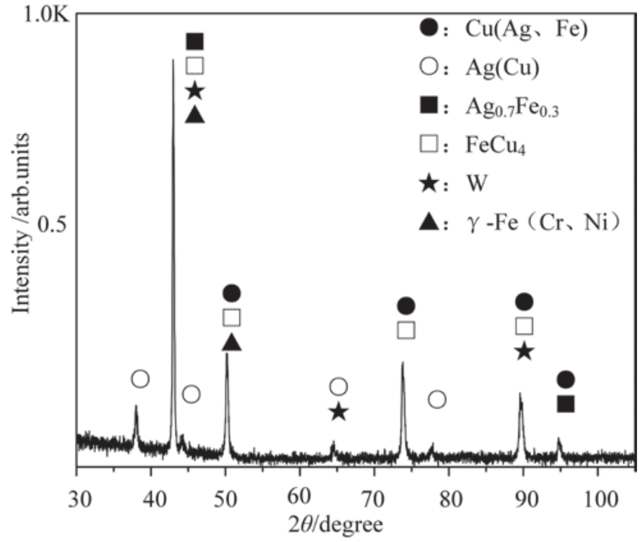

Fig. 8: X-ray diffraction patterns

measured by INCA Energy Diffraction Spectrum (as shown in Fig. 7). The test location was shown in Fig. 3b by the arrow. A tiny diffusion layer (about 6 to 10 μm) was formed between the brazing seam and the substrates. While being brazed, copper and a little tungsten elements transited from W-Cu composite into the brazing seam region. Binary phase of tungsten-silver has a similar property as well as tungsten-copper. Neither any solid solution nor intermetallic compounds (IMC) formed. Therefore tungsten transited into the brazing seam region was existed in form of simple substance. However it was observed by SEM that no bulk tungsten phase of substance existed in the brazing seam, confirmed that only a small amount of W transited. Fe element was transited into the brazing seam region from the 1Cr18Ni9 steel.

Elements from the substrates dissolved into the brazing seam; at the same time elements of the liquid brazing filler metal diffused into the base metal. In the process of brazing W-Cu and steel with Ag-Cu filler metal, a tiny of Ag element diffused into the 1Cr18Ni9 steel, while more Ag element diffused into the W-Cu composite. Obvious transition of Ag from the filler metal into W-Cu composite was related to the diffusion voids of Cu element transited from the substrates into brazing seam and eutectic metallurgical reaction between Ag and Cu (eutectic temperature 779 °C). On interface of the brazing seam region and both of substrates, high concentration level of Ag element was formed, showed that Ag content in the brazing filler metal has a good wetting on these two substrates.

3.4 X-ray diffraction analysis

To further clarify phase constitution near interface of the brazed joint, XRD analysis was carried out in the bending fracture of W-Cu alloy together with brazing seam obained by four-point bending test. The obtained result was compared with data from the Joint Committee on Power Diffraction Standards (JCPDS) to determine the existed phase, as shown in Fig. 8.

Cu-rich phase Cu(Ag, Fe) and Ag-rich phase Ag(Cu) was the main components existed in brazing seam of the joint of W-Cu composite and 1Cr18Ni9 steel. Meanwhile metallurgical reaction was occurred on the interface between 1Cr18Ni9 steel and Ag-Cu filler metal. It induced that a little IMC phases of $Ag_{0.7}Fe_{0.3}$ and $FeCu_4$ formed, dispersively distributed in the bulk phase of Cu-rich Cu(Ag, Fe) solid solution to improve the compactness of brazing seam. In the pattern of X-ray diffraction, W phase from W-Cu composite and γ-Fe(Cr, Ni) austenite from the 1Cr18Ni9 steel were also existed.

Due to the special monomer mixed tissue of two phases, uniform irregularities were existed on the surface of W-Cu composite. While being brazed, the melt filler metal infiltrated in depression area of the surface of W-Cu composite, and formed a mechanical occlusion effect. So contact area of the filler metal and substrate was raised to improve the bonding strength of brazing interface to achieve a securely connecting between W-Cu composite and 1Cr18Ni9 steel.

4 Conclusions

1. Via adopting Ag-28wt%Cu filler metal, W-Cu composite and 1Cr18Ni9 steel was successfully brazed with process parameters, brazing temperature 855 to 865 °C, holding time 30 min and vacuum level superior to 6×10^{-3} Pa. In brazing seam region, Ag-Cu eutectic phase and bulk phase near 1Cr18Ni9 steel were formed. And microhardness of the brazing seam region was relatively low and average strength of the tested specimens reached 576 MPa via the four-point bending, with fracture occurred near the W-Cu alloy side.
2. Elements distribution analysis showed that Cu and a little W element from W-Cu composite and Fe element from 1Cr18Ni9 steel transited into the brazing seam. Meanwhile Ag element diffused from the filler metal into W-Cu composite. A high concentration of Ag element existing on the brazing interface of W-Cu composite and 1Cr18Ni9 steel was observed, obviously improving the interfacial reaction and ensuring performance of the obtained joint.
3. X-ray diffraction analysis illustrated that the brazing seam mainly contained of Cu-rich phase Cu(Ag, Fe) and Ag-rich phase Ag (Cu). Metallurgical reaction was occurred between Fe element from 1Cr18Ni9 steel and the melt Ag-Cu filler metal, and a small amount of $Ag_{0.7}Fe_{0.3}$ and $FeCu_4$ were formed.

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