HAL Id: jpa-00224664

https://hal.archives-ouvertes.fr/jpa-00224664

Submitted on 1 Jan 1985

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
HREM AND DIFFRACTION STUDIES OF AN Al₂O₃/Nb INTERFACE

M. Florjancic, W. Mader, M. Rühle and M. Turwitb

Max-Planck-Institut für Metallforschung, Institut für Werkstoffwissenschaften, Seestrasse 92, D-7000 Stuttgart 1, F.R.G.

Abstract: Single crystals of Nb and Al₂O₃ were welded so that close-packed planes of both materials were parallel. Thin specimens (suitable for TEM) containing the interface were prepared out of the bulk bicrystal. The thickness of the foil was ~5 to 10 nm. HREM studies allowed the direct imaging of the interface. No pronounced intermediate layer (e.g., oxides, spinel) could be determined between the two constituents near the interface. Small deviations from the exact orientation of the interface are accommodated by facets. The diffraction pattern revealed pronounced streaks close to the diffraction spots of Nb. The shapes and positions of the streaks may allow the determination of the width of the distorted region.

1. Introduction

Ceramics are potential candidates for high technology engineering components. Frequently, the ceramics must be connected to a metallic component which requires a bonding of the metal to the ceramic partner. Therefore, it is not only of scientific but also of technological interest to understand the bonding mechanisms at metal/ceramic interfaces. It is known that the bonding governs the mechanical properties of the joints.

To understand the bonding mechanisms different investigations are required. Firstly, it must be determined if a chemical reaction layer between the metallic and the ceramic component was formed during the bonding. Pepper [1] found that such an intermediate layer is formed at certain interfaces and influences the mechanical properties of the joint substantially. Secondly, the structure of the interface must be determined, if possible, to the atomic level. The results of the latter investigations can be used for the development of an atomic model and of the bonding mechanisms at the interface and, furthermore, for an understanding of the properties of the metal/ceramic interface.

In this paper recent observations are reported concerning the structure of a niobium/sapphire (Al₂O₃) interface. The observations are performed by high resolution electron microscopy (HREM).

2. Experimental Details

Single crystals of niobium (99.9% purity) and of alumina (99.9% purity) were diffusion-bonded for two hours in a dynamical vacuum of 1.3 × 10⁻⁴ Pa under a compressive load of 10 MPa. Following orientation relationship between the Nb and Al₂O₃ single crystals was adjusted with an experimental accuracy of ± 1°:

\[(0001)_{\text{Al}_2\text{O}_3} \parallel (110)_{\text{Nb}} \text{ and } [2110]_{\text{Al}_2\text{O}_3} \parallel [001]_{\text{Nb}}.\]

The preparation of a thin specimen suitable for high resolution TEM is extremely difficult and requires the following steps. A rod (3 mm diameter, rod axis \([001]_{\text{Nb}} \parallel [2110]_{\text{Al}_2\text{O}_3}\) must be drilled out of the bonded block and the interface
must lie in the center of the rod with the plane of the interface parallel to the rod axis. Disks (thickness 0.2 mm) must be cut off the rod by using a diamond blade saw. Subsequently, the disks have to be polished to a thickness of ~100 μm. The central area of the specimen is then thinned mechanically to a thickness of ~10 μm by dimpling. Finally, the specimens must be ion-thinned with low energy argon ions (3 keV). During ion-thinning the specimen is kept at liquid N₂ temperature to reduce contamination at the specimen surface and also to reduce radiation damage. A 1 nm to 2 nm thick evaporated carbon layer avoids electrical charging of the specimen during the observations in the electron microscope.

The specimen (which included the edge-on Nb/Al₂O₃ interface) was investigated using a JEOL 200CX TEM (high resolution polepiece). Direct lattice images (HREM images) could only be performed of regions where the foil thickness was smaller than ~10 nm.

3. Experimental Results

The specimen was tilted in the microscope so that the interface and a certain crystallographic direction ([001]ₙb and [2110]ₐl₂o₃) were parallel to the incoming electron beam. The alignment of the specimen had to be done extremely careful since the HREM micrographs depend sensitively on small misalignments. A selected area diffraction pattern of the bicrystal is shown in Fig. 1. The diffraction spots of Nb and Al₂O₃ can be identified and, in addition, faint streaks. The latter are marked by arrows on the micrograph. The streaks are formed by a region close to the interface. It is expected that from the position and the length of the streaks information on the distortions near the interface can be evaluated. The close-packed planes (110)ₙb and (0006)ₐl₂o₃ are tilted against each other by ~3° (Fig. 1). The misalignment is observed in different regions of the interface.

Fig. 1: Selected area diffraction pattern of the region close to the Nb/Al₂O₃ interface. Streaks (marked by arrows) can be observed besides the reflections of both crystals.
Fig. 2: Lattice image of the crystalline region close to an Nb/Al₂O₃ interface. Different crystallographic planes of Al₂O₃ and Nb are identified. Lattice distortion occurs only in regions very close to the interface. Steps (see arrow!) can be observed. Foil normal of Nb (lower part of micrograph): [001]; foil normal of Al₂O₃ (upper part of micrograph): [2110].

Fig. 2 represents a lattice image (HREM image) of the crystalline regions near the interface. Besides the central beam, the following reflections are used for imaging: four (110) reflections of Nb, the (01T), (01T2), (0114), (0114), (0006) and (0006) reflections of Al₂O₃.

The lattice image (Fig. 2) reveals that the interface lies parallel to the close-packed plane of Al₂O₃. Atomic steps and facets can also be identified. A qualitative inspection of Fig. 2 shows that distortions of the lattices (usually the Nb lattice) are only observed very close to the interface. At the most, four lattice planes of the crystals adjacent to the interface are strained.

No intermediate or reaction layer (NbO₂) could be observed in any of the specimens studied so far. This result is in contrast to the observations by Morozumi et al. [3] who observed an NbO₂ layer (thickness up to ~10 µm) between Al₂O₃ and Nb at interfaces which were diffusion bonded under similar conditions as noted above.

Faceting is observed if the average orientation of the interface deviates from the close-packed planes of Al₂O₃ and Nb (Fig. 3). One facet is always parallel to the close-packed planes of Al₂O₃ while the orientation of the others is usually inclined with respect (i) to the close-packed plane and (ii) to the foil normal. The different facets can clearly be identified on Fig. 3b.

A quantitative interpretation of the HREM micrographs is not possible by simple inspection. The atomic structure near the interface can only be determined if HREM micrographs, taken under different focussing conditions of the objective lens, are compared to computer-simulated images. The simulated images must be calculated for different atomic models of the interface and for the instrumental conditions of the electron microscope (cf. Spence [4]).
1. Facets are observed if the orientation of the interface (w.r.t. certain planes in Nb and Al₂O₃) deviates from the low energy plane. Fig. 3a: complete facetted region; Fig. 3b: high magnification of a section of Fig. 3a.

4. Discussion

The HRTEM work done so far does not allow a quantitative evaluation of the atomic structure near the interface. However, some of the observations described above can give information on the bonding mechanisms.

Both alumina [5, 6] and Nb [7] undergo plastic deformation by creep during bonding at 1973 K. In addition, small amounts of Al₂O₃ should be dissolved in Nb at 1973 K due to the thermodynamical considerations of Schulze et al. [8]. Both the plastic deformation and the dissolution of small amounts of alumina may allow the adjustment of the relative orientation of the Al₂O₃ with respect to the Nb single crystal and also of the interface with respect to both adjacent crystals. It can be assumed that the observed structure represents the energetically favourable configuration.

The close-packed planes of Nb and Al₂O₃ are not exactly parallel (see Fig. 3). There exists a systematic deviation of ~ 3° which was also observed by Feldman [9] between epitaxially grown Nb foils and the Al₂O₃ substrate. The reason for this misorientation is not yet understood. The streaks observed on selected area diffraction pattern (Fig. 1) indicate that a thin layer close to the interface is distorted. However, some of the streaks may be formed by double diffraction.
Further experiments and model calculations are required for a quantitative explanation.

The interface region described by Morizumi et al. [3] was not observed although the experimental conditions for diffusion bonding were very similar in our and Morizumi’s et al. experiments. Further experimental studies may explain the difference.

A detailed explanation of the structure of metal/ceramic interfaces requires first a quantitative evaluation of HREM micrographs. It is expected that this quantitative evaluation should yield the coordinates of the atoms (ions) close to the interface. In a second step, the observation must be compared to models of metal/ceramic interfaces. It is expected that the pure geometrical models (e.g. the coincidence model) now available may not be sufficient for explaining the structure and energy of metal/ceramic interfaces.

5. Acknowledgement

The authors acknowledge the experimental assistance of Mrs. R. Paulini and Ms. D. Waidelich during the difficult preparation of the specimens suitable for HREM.

6. References