Available online at www.sciencedirect.com

Journal of Nuclear Materials 321 (2003) 129–134

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Strength proof evaluation of diffusion-jointed W/Ta interfaces by small punch test

Jing-Feng Li [a,*], Masayoshi Kawai [b], Kenji Kikuchi [c], Tadashi Igarashi [d], Hiroaki Kurishita [e], Ryuzo Watanabe [f], Akira Kawasaki [f]

[a] *State Key Laboratory of New Ceramic and Fine Processing, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, PR China*

[b] *High Energy Accelerator Research Organization, Tsukuba 305-0801, Japan*

[c] *Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken 319-1195, Japan*

[d] *Department of Metallurgy and Materials Science, Osaka Prefecture University, Osaka 599-8531, Japan*

[e] *Institute for Materials Research, Tohoku University, Oarai-machi, Ibaraki-ken 311-1313, Japan*

[f] *Department of Materials Processing, Tohoku University, Sendai 980-8579, Japan*

Received 15 January 2003; accepted 7 April 2003

Abstract

For the development of tantalum-clad tungsten targets for spallation neutron sources, the bonding strength of tantalum–tungsten interface was investigated by means of an easy-to-use and miniaturized small punch (SP) test, in which a punching load is vertically applied to the center of a jointed disk. Cracks initiated and propagated in the tungsten side for all the samples hot-isostatically pressed (HIPed) at temperatures from 1673 to 2073 K, whereas no crack and debonding were observed in the interface, indicating that the jointed interface is strongly bonded. The recrystallization of tungsten occurs and results in its strength reduction, consequently the crack-initiating load decreases with HIPing temperature. The finite element analysis of the measured SP testing results shows that the maximum bonding strength can exceed 1000 MPa. The present study shows that SP test is suitable for the strength evaluation of jointed tantalum–tungsten interfaces.

1. Introduction

High density spallation neutron sources will be increasingly used in many fields, including physics, chemistry, materials science, medicine, and earth science. The construction of high intensity spallation neutron source (SNS) is under way in Japan [1] and in USA. One of the key technologies of that project is the development of the target materials that generate high-density neutrons when irradiated by high-energy proton beams. Heavy metals are promising alternative candidate target materials because they produce high neutron intensities, and tungsten has an additional advantage since its post-irradiation half life of decay is much shorter compared to its neighboring elements [2,3]. However, tungsten shows poor corrosion resistance against water coolant due to the formation of ragged tungsten hydroxide [4,5], and intense susceptibility to radiation embrittlement [6,7]. One of the solutions to these problems is to clad tungsten with a corrosion-resistant material such as tantalum, titanium, stainless steel. Among them, tantalum may be the best cladding material because tantalum has a good combination of chemical and mechanical properties, and tungsten and tantalum are compatible due to their complete solid solubility [2].

Kawai et al. [2] have recently fabricated tantalum-clad tungsten targets for their spallation neutron source by using a hot-isostatic pressing (HIPing) process. Fig. 1

---

[*] Corresponding author. Tel.: +86-10 62784845; fax: +86-10 62771160.

E-mail address: jingfeng@mail.tsinghua.edu.cn (J.-F. Li).

0022-3115/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/S0022-3115(03)00237-X

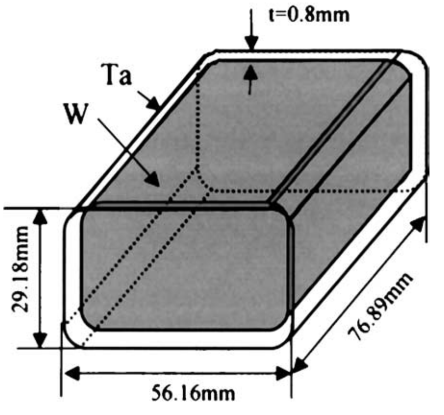

Fig. 1. Configuration and dimensions of Ta-clad W target.

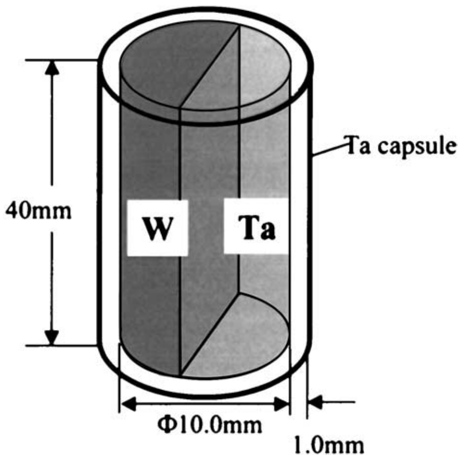

Fig. 2. Configuration and dimensions of bonded Ta–W sample.

shows the configuration and dimensions of the target block. The inside W block (29.18 mm × 56.16 mm × 76.89 mm) was capsulated into a tantalum can with a thickness of 0.8 mm by electron beam welding in vacuum. The HIPing process is conducted to bond the cladding tantalum ‘skin’ onto the tungsten block at an optimized temperature. The bonding integrity of the interface between the tantalum overlayer and the tungsten block is of the first concern for the spallation target because bonding defects, if any, would result in the peeling-off of the corrosion resistant surface layer, thus shortening the lifetime of targets. Increasing HIPing temperature is effective in endowing the interface with high bonding strength. But the recrystallization and grain coarsening of tungsten and tantalum may be enhanced simultaneously during HIPing at high temperature, which would reduce the fracture toughness and hence the mechanical reliability of the target. It is, therefore, necessary to optimize HIP cladding temperature. The motivation of the present study was to provide some intuitive information to help optimize the HIPing temperature through the measurement of the bonding strength by using the small punch (SP) testing method; which is originally developed for the mechanical evaluation of nuclear materials using miniature-sized disk-shaped testing pieces [8]. Some interesting and informative results were obtained by punching the bonded tantalum and tungsten interface in the biaxial bending mode.

2. Experiments and simulation

The SP samples were prepared by the following procedures. First, tungsten and tantalum billets with a semicircle cross section (radius = 5 mm) were sampled from the same kind of materials as the target. The rectangular longitudinal cross section was polished to the same finish as the tantalum can and tungsten block for the targets. The polished sections of the both billets were put together and inserted into a tantalum tube (inner diameter = 10 mm, thickness = 1 mm), as shown in Fig. 2. The tube was vacuum-sealed by electron beam welding, and HIP-treated under the same conditions as in the HIP-cladding: under pressure of 200 MPa and enveloped by zirconium foils and tantalum plates as an impurity gas getter. Four HIPing temperatures, 1673, 1773, 1973 and 2073 K were selected, and the holding time was set at 3 h for all the HIP-treatments for the fabrication of the target. Finally, the HIP-treated samples were sliced into thin disks, two bonded halves of which are tungsten and tantalum, respectively. After polishing the disk surface, the specimens were subjected to SP testing, in which the load was applied to the specimen center, i.e., at the center of the joint interface, as shown in Fig. 3. For the SP tests, the disk was supported by a die with a center hole of 4.5 mm in diameter,

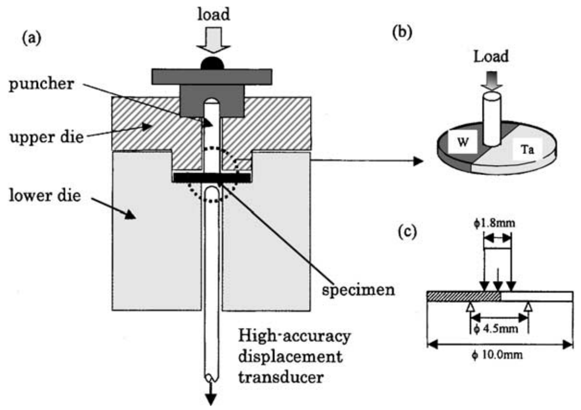

Fig. 3. (a): schematic of small punch (SP) test; (b): the sample setup for evaluating bonded W–Ta disk; (c) dimensions for calculation model.

and was punched by a plat-head rod of 1.8 mm in diameter; and the details of the SP method can be found elsewhere [9,10].

For homogeneous and brittle materials, stress and fracture strength can be easily calculated by using an equation [8,9] derived on the basis of elastic mechanics. However, the present case is fairly complicated since the two materials, which are bonded together to support the load, have different materials properties, such as Young's modulus and Poisson's ratio. Therefore, in the present study, finite element method (FEM) was used to make out the stresses generated during the SP loading. For the FEM analysis using ANSYS system, a z-axis symmetrical disk model, as shown in Fig. 3, was used for the calculation limited to elastic deformation. It is assumed that the load is uniformly applied to the center of the disk, and the two halves of the mesh have the materials properties of tungsten and tantalum, respectively.

All the SP tests were conducted in Ar atmosphere under 0.1 MPa, by using jigs made of SiC ceramics. The crosshead speed for the SP tests was 0.05 mm/min, which corresponds to a nominal strain rate of $\sim 1.25\times10^{-4}$ s^{-1}. The deformation was measured using a high-accuracy transducer connected to a detecting bar, which was attached onto the center of disk specimens. The circle surfaces of the SP specimens were well polished, and then the fracture surface and paths were observed by scanning electron microscopy (SEM) after testing. SEM also was used to observe the microstructural changes of the tungsten and tantalum materials subjected to the HIPing treatments at various temperatures. For the SEM observation, these two materials were polished and chemically etched using a solution containing HF and HCl and H_2O mixed at an equal volume fraction.

3. Results and discussion

Figure 4 shows the typical SP load–displacement curves of the jointed specimens HIPed at various temperatures. For all the specimens, a first load drop appeared at different load levels, and then the load and displacement increased again. To clarify the cause of this phenomenon, the testing was interrupted when the load drop was observed. The SEM observation found that small cracks formed in the SP specimens even though the whole specimen looked undamaged. Therefore, it was revealed that the load drop corresponded to the crack initiation. As shown in Fig. 5, cracks formed and propagated within tungsten side in all the specimens, regardless of different HIPing temperatures. This observation suggests that the W–Ta interfaces formed under the present HIPing conditions are well jointed. At least, no debonding occurred before the crack initiation. However, the stress value (not the load drop) corresponding to the crack initiation is still unknown because an additional stress concentration should be generated in the jointed disk due to the difference of elastic properties for tungsten and tantalum; their Young's moduli are 411 and 186 GPa, respectively. Therefore, FEM was conducted to clarify the relationship between the stresses and the applied SP load. To the first approximation, the thermal residual stresses, which is caused by the cooling during fabrication process, was not incorporated into the FEM calculation because of its difficulty.

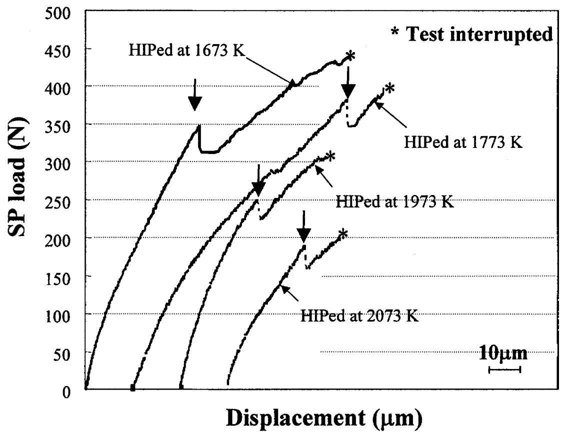

Fig. 4. Representative SP load–displacement curves of the specimens HIPed at various temperatures. Note that the tests were interrupted before the eventual fracture.

Figure 6 shows the distribution of the stresses that are parallel and normal to the jointed interface when the whole sample is subjected to a load of 196 N. Because tungsten has a larger Young's modulus than tantalum, as shown in Fig. 6(a), large stresses were generated in tungsten side, particularly concentrated near the center of the disk, with a peak stress reaching 836 MPa. On the other hand, high tensile stresses were also normally applied to the jointed interface, as shown in Fig. 6(b), and the peak value reaches 607 MPa for the same SP load (196 N). It is worthy to note that the first crack initiated in the specimens HIPed at 2073 K approximately under a load of 196 N. This result means that the tungsten cracked because its fracture strength is close to 836 MPa, whereas the bonding strength is higher at least than 607 MPa. In other words, the proof strength of the interface bonded at 2073 K is this value (607 MPa), being about 72% of the crack-initiating stress at W-side.

As compared with conventional tensile or beam bending tests, the stress state in the present SP loading is complex as shown in Fig. 6. Nevertheless, the strength data determined by SP method are not so much different from that from the conventional uniaxial tests because the failure occurs at the maximum stressed area. In fact, our previous study [11] found that the fracture strength values of ceramics measured by SP method were about

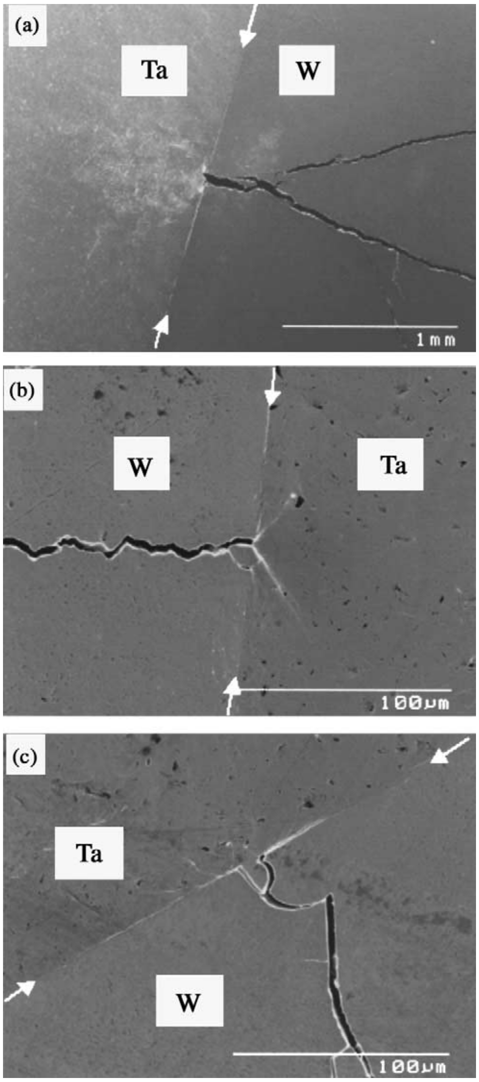

Fig. 5. SEM micrographs of the tensile surface (bottom surface) of the tested specimens. Note that cracks initiated and propagated in the W side in all the specimens. The arrows indicate the bonded interface.

10% lower than that determined by conventional four-point bending test, and explained that the ceramics fractured at lower SP load because of the influence of its biaxial feature.

Figure 7 shows the changes in crack-initiating load/stress of the W-side and the proof strength of the W/Ta bonding interfaces as a function of HIPing temperature. For the specimen HIPed at 1773 K, the crack-initiating load was twice higher than that HIPed at 2073 K, suggesting that the bonding strength may exceed 1000 MPa. Such a high bonding strength was obtained probably because complete solid solubility is possible in the tungsten and tantalum system. In fact, the interdiffusion layer was reported to be 3.5 μm for the similar interface HIPed under the same condition [2]. Even for the sample HIPed at the lowest temperature (1673 K), as shown in Fig. 8, a W grain at the jointed interface was pulled out

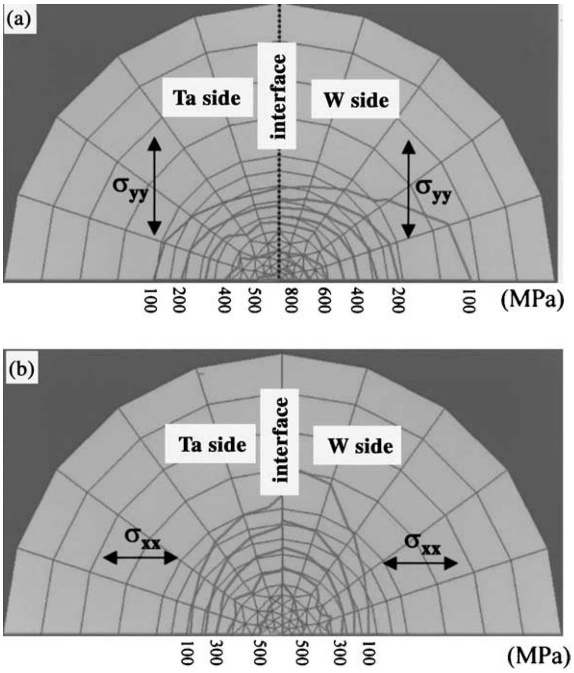

Fig. 6. Stress distribution in the tensile surface of a SP disk subjected to a load of 196 N. (a): stress parallel to the bonding interface; (b): stress normal to the bonding interface.

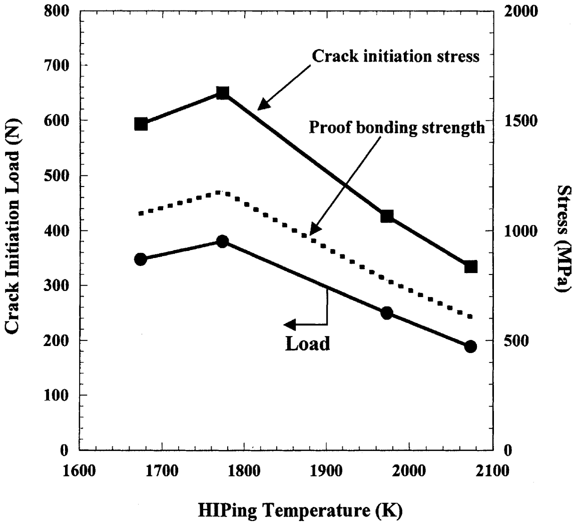

Fig. 7. Changes in crack-initiating load/stress of the W side and the proof strength of the W/Ta bonding interfaces as a function of HIPing temperature.

from W side, demonstrating the strong bonding state of the W and Ta interface.

The remaining question is why the crack-initiating load decreased with HIPing temperature when the samples were HIP-treated at temperatures higher than 1773 K. The answer was found by observing the microstructural changes caused by the HIPing process. As shown in Fig. 9, the grain size significantly increased when the HIPing temperature exceeded 1773 K, due to the recrystallization of polycrystalline W. Therefore, the crack-initiating load decreased because the W side became increasingly weaker with HIPing temperature when the HIPing temperature exceeded 1773 K. The crack-initiating load increased somewhat when the HIPing temperature was raised from 1673 to 1773 K, probably because the HIPing at moderate temperatures was beneficial to the strength improvement of the rolled polycrystalline W.

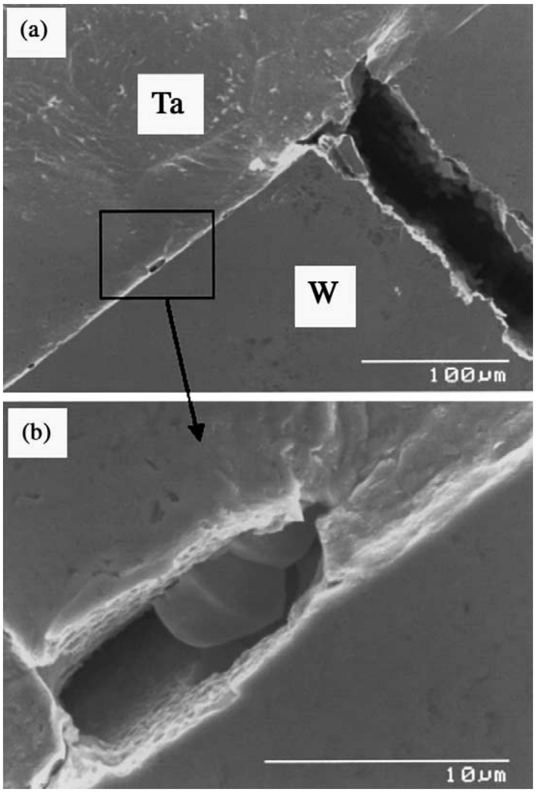

Fig. 8. Representative SEM micrographs showing a W grain at the interface was pulled out. (a): low magnification, (b): high magnification.

4. Conclusion

The present study demonstrates that SP test combined with numerical stress analysis is suitable for the evaluation of bonding strength of the HIP-jointed W–Ta interfaces.

When a jointed W–Ta disk is subjected to the SP test, cracks initiate and propagate in the W side, whereas no crack and debonding are observed in the interface, indicating that the jointed interface is strongly bonded. The

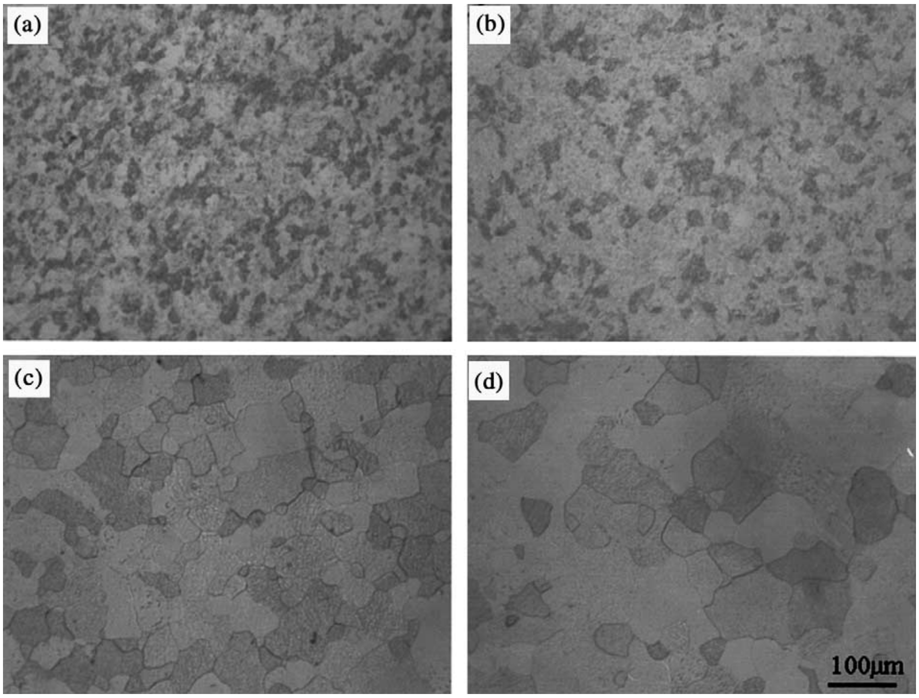

Fig. 9. SEM micrographs comparing the microstructures of W block subjected to HIP-treatments at different temperatures.

finite element analysis of the measured results shows that the maximum bonding strength can exceed 1000 MPa.

If the HIPing temperature is too high, recrystallization of W occurs and results in its strength reduction, consequently the crack-initiating load decreases with HIPing temperature. The present study shows that the optimal HIPing temperature is 1773 K for cladding W with Ta.

References

[1] Y. Oyama, S. Ikeda, JAERI-KEK Joint Project Team: Status of Spallation Neutron Source Program in High Intensity Proton Accelerator Project, JAERI-Conf 2001-002/KEK Proceedings 2000-22, ICANS-XV, Tsukuba, 6–9 November 2000, p. 19.

[2] M. Kawai, K. Kikuchi, H. Kurishita, J.-F. Li, M. Furusaka, J. Nucl. Mater. 296 (2001) 312.

[3] R. Watanabe, J.-F. Li, A. Saito, H. Kurishita, T. Igarashi, K. Okamura, K. Kikuchi, M. Furusaka, M. Kawaii: 15th International Plansee Seminar 2001, 28 May–1 June 2001, Reutte, Austria, 4, 1.

[4] K. Sugimoto, Corrosion Resistance of Tungsten and Its Alloys for Spallation Target in Aqueous Solution, KEK Proceedings, 2002-6, 2002, p. 126.

[5] Y. Ishijima, K. Kakiuchi, T. Furuya, H. Kurishita, M. Hasegawa, T. Igarashi, M. Kawai, J. Nucl. Mater. 307–311 (2002) 1369.

[6] J.W. Davis, V.R. Barabash, A. Makhankov, L. Plochl, K.T. Slattery, J. Nucl. Mater. 258–263 (1998) 308.

[7] H. Ulmaier, F. Carsughi, Nucl. Instrum. and Meth. B 101 (1995) 406.

[8] J. Kameda, Bull. Jpn. Inst. Metals 25 (1986) 520.

[9] M. Saito, H. Takahashi, H.D. Jeong, A. Kawasaki, R. Watanabe, JSME 57 (1991) 522.

[10] J.-F. Li, R. Watanabe, Mater. Trans. JIM 40 (1999) 508.

[11] J.-F. Li, A. Kawasaki, R. Watanabe, J. Jpn. Inst. Metals 56 (1992) 1450.