Available online at www.sciencedirect.com
ScienceDirect
Acta Materialia 59 (2011) 6530–6537
Acta MATERIALIA
www.elsevier.com/locate/actamat

Intergranular penetration of liquid gold into stainless steel

D. Favez*, L. Deillon, J.-D. Wagnière, M. Rappaz

Laboratoire de Simulation des Matériaux, École Polytechnique Fédérale de Lausanne, Station 12, CH – 1015 Lausanne, Switzerland

Received 16 February 2011; received in revised form 8 June 2011; accepted 18 June 2011
Available online 4 August 2011

Abstract

Intergranular penetration of liquid 18 K gold into a superaustenitic stainless steel, which occurs during laser welding of these two materials, has been studied using a C-ring device which can be put under tensile stresses by a screw. It is shown that liquid gold at 1000 °C penetrates the immersed stainless steel C-ring at grain boundaries, but only when tensile stresses are applied. Based on the thickness of the peritectic phase that forms all along the liquid crack and on the transverse gold diffusion profile in steel, penetration velocities on the order of 10 μm s^{-1} are deduced.

Keywords: Grain boundaries penetration; Liquation; Laser welding; Au-Fe alloy

1. Introduction

During laser welding of gold and stainless steel for jewelry applications, it has been observed that the liquid of the molten pool, made primarily of gold, can penetrate the solid stainless steel part [1]. This occurs at grain boundaries juxtaposing the liquid pool, where the solid is under maximum tensile stresses due to thermal contraction. Typical penetration distances are 10–100 μm. Taking into account the interaction time with the laser beam, i.e. pulse duration, a penetration velocity on the order of 0.01–0.1 m s^{-1} can be estimated.

Penetration of a liquid metal at grain boundaries of a solid is found in the literature in various contexts. The thermodynamic driving force is the surface energy decrease caused by the replacement of a large misorientation grain boundary of energy γ_{gb} by two solid–liquid interfaces of lower energy γ_{sl}, i.e. when $2\gamma_{sl} < \gamma_{gb}$ [2–4]. However, the mechanisms of intergranular liquid penetration are still not fully understood.

Fundamental research on the underlaying mechanisms and penetration velocity is mainly related to liquid metal (induced) embrittlement (LME or LMIE). In this case, a normally ductile metal such as Al or Cu looses its ductility when it is in contact with another metal in the liquid state, such as Ga or Bi [5,6]. The liquid metal penetrates the solid at grain boundaries and dramatically decreases its mechanical properties. Numerous studies and reviews have been published on LME and several models have been developed. However, there is no single explanation or model that is generally applicable, as the mechanisms appear to vary from one system to another [7,8]. According to Glickman [9], propagation of the liquid crack is controlled by atomic reaction at the tip. Above a stress intensity threshold K_{TH}, propagation occurs at a fixed rate V_∞, these two parameters being highly sensitive to the nature of the liquid metal. Recently, Klinger and Rabkin [10] proposed a mechanism in which the concentration gradient along a wetted grain boundary induces a grain boundary energy gradient. This causes a net flux of atoms towards the liquid crack tip, which accumulate and subsequently generate stresses that are able to further propagate the liquid crack. According to this mechanism, no external stress is required.

Another similar phenomenon observed during welding is related to grain boundary liquation in the so-called

---

* Corresponding author.
E-mail addresses: denis.favez@a3.epfl.ch (D. Favez), lea.deillon@epfl.ch (L. Deillon), jean-daniel.wagniere@epfl.ch (J.-D. Wagnière), michel.rappaz@epfl.ch (M. Rappaz).

1359-6454/$36.00 © 2011 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.actamat.2011.06.030

heat-affected zone. Because of their higher surface energy or the presence of low melting temperature intermetallics caused by microsegregation in the base material, grain boundaries can melt at temperatures below the solidus [11,3]. In the presence of tensile stresses, the liquid films at grain boundaries show almost no mechanical resistance and open, thus allowing the liquid of the weld pool to penetrate. This phenomenon is thus very similar to "healed" hot tears forming during solidification, i.e. when the opening of grain boundaries by tensile stresses that normally leads to hot cracks can be fed by interdendritic or intergranular liquid [12,13].

One particular case of the presence of liquid metal at the grain boundaries of steel is associated with copper. Copper can be present in steel as impurities, especially when recycling car bodies which contain copper electric wires, or when welding is performed with copper-containing fixtures or tools around the welds. During welding or hot rolling, copper can penetrate along grain boundaries in some austenitic or structural steels. This leaves an intergranular solid copper film that dramatically decreases both the base metal strength and ductility at high temperature. The "hot-shortness" induced by the presence of copper at grain boundaries is thus called copper contamination cracking (CCC). The CCC is accelerated by external or residual stresses [14–16], but no detailed mechanism has been given in the literature.

Experimental characterization of these phenomena is usually related to a specific application. In the case of LME, the loss of mechanical properties is of major concern. Therefore, tensile or compression tests at room temperature are conducted after penetration [17,18], while in situ experiments using transmission electron microscopy or X-ray microtomography give insights into the underlaying mechanisms [5,19]. Several tests are commonly used to characterize the liquation cracking susceptibility during welding. One of them, the nil-ductility test, focuses on the temperature at which the ductility of a specimen falls to zero [20].

The aim of the present study was to better understand the origin of liquid gold penetration into stainless steel during laser welding. In transverse cross-section of the welds, liquid penetration appears mainly in regions at about 45° between the weld centerline and the top surface of the steel side. Since this location also corresponds to maximum tensile stresses during welding, it was conjectured that penetration of liquid gold into solid steel requires a tensile stress state of the solid material. In order to test this hypothesis at temperatures at which liquid gold is in contact with solid steel (i.e. temperature in the range 900–1350 °C) a new specific setup had to be developed, as standard LME tests are not applicable.

2. Experiment

With this objective in mind, a so-called "C-ring test" was selected (Fig. 1). It is a standard stress–corrosion test in which a specimen with a "C" shape can be stressed by a screw parallel to the opening of the "C" (see the figure). At the symmetry plane of the C-ring, the hoop stress component $\sigma_{\theta\theta}$ is tensile at the outer surface while it is compressive at the inner surface [21]. Furthermore, the tensile component varies from zero at the location of the screw to a maximum at the symmetry plane of the C-ring, this maximum being adjusted by the compression (or radial stress) component at the screw location. Besides stress–corrosion cracking, such a test has also been used to study the penetration at room temperature of steel or aluminum by mercury [22,23]; however, to the best of our knowledge, no application has been made at elevated temperature. One of the reasons for this is that, at high temperature, the C-ring material (and possibly the screw itself) may creep, thus partially releasing the imposed stress [24]. The initial imposed stress level was found to be not so important, as long as the screw was tightened (even by hand only) at room temperature. During heating, some additional stress could be induced by differential thermal expansion of the screw and C-ring. However, the behavior of steel at the temperature of the liquation test (1000 °C, i.e. about 2/3 of the melting point) is such that creep becomes dominant. Thus, the remaining stress in the C-ring is also a function of the strain rate, which itself depends on the penetration speed. In any case, if only partially released, the hoop stress component remains compressive/tensile inside/outside the C-ring and increases from the screw to the symmetry plane. This test can provide qualitative information on the effect of stress on liquid gold penetration.

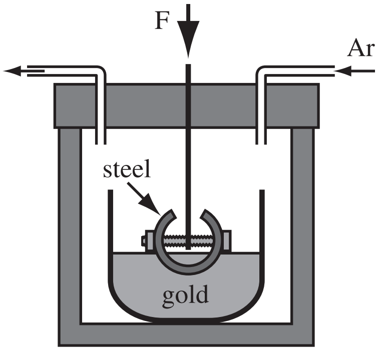

Fig. 1. Experimental C-ring setup. The inner/outer diameters of the C-ring are 20/24 mm; its thickness is 20 mm.

The simple testing procedure using the C-ring was as follows: a crucible containing gold and a steel C-ring, precleaned with Ridoline, were placed in a furnace at 1000 °C, under an argon flux. After melting and homogenization of the melt, the C-ring was immersed in liquid gold for a given time. The sample was then withdrawn and quenched in air. The bath was a classical 18 carat yellow gold (Au–12.5 wt.%Ag–12.5 wt.%Cu) and the C-ring was a superaustenitic 904L stainless steel.

For metallographic observations, the samples were sectioned and polished using SiC papers down to 1 μm, and were then observed by scanning electron microscopy