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Intergranular penetration of liquid gold into stainless steel

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Received 16 February 2011; received in revised form 8 June 2011; accepted 18 June 2011

7 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.

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14 Q1 Keywords: Grain boundaries penetration; Liquation, 15

16 1. Introduction

During laser welding of gold and stainless steel for jew-17 elry applications, it has been observed that the liquid of the 18 molten pool, made primarily of gold, can penetrate the 19 solid stainless steel part [1]. This occurs at grain boundaries 20 21 juxtaposing the liquid pool, where the solid is under maximum tensile stresses due to thermal contraction. Typical 22 penetration distances are 10-100 µm. Taking into account 23 the interaction time with the laser beam, i.e. pulse duration, 24 a penetration velocity on the order of 0.01–0.1 m s⁻¹ can be 25 estimated. 26

Penetration of a liquid metal at grain boundaries of a 27 solid is found in the literature in various contexts. The ther-28 modynamic driving force is the surface energy decrease 29 caused by the replacement of a large misorientation grain 30 boundary of energy γ_{gb} by two solid-liquid interfaces of 31 lower energy γ_{sl} , i.e. when $2\gamma_{sl} < \gamma_{gb}$ [2-4]. However, the 32 mechanisms of intergranular liquid penetration are still 33 not fully understood. 34

Fundamental research on the underlaying mechanisms 35 and penetration velocity is mainly related to liquid metal 36 (induced) embrittlement (LME or LMIE). In this case, a 37 normally ductile metal such as Al or Cu loses its ductility 38 when it is in contact with another metal in the liquid state, 39 such as Ga or Bi [5,6]. The liquid metal penetrates the solid 40 at grain boundaries and dramatically decreases its mechan-41 ical properties. Numerous studies and reviews have been 42 published on LME and several models have been devel-43 oped. However, there is no single explanation or model 44 that is generally applicable, as the mechanisms appear to 45 vary from one system to another [7,8]. According to 46 Glickman [9], propagation of the liquid crack is controlled 47 by atomic reaction at the tip. Above a stress intensity 48 threshold K_{TH} , propagation occurs at a fixed rate V_{∞} , these 49 two parameters being highly sensitive to the nature of the 50 liquid metal. Recently, Klinger and Rabkin [10] proposed 51 a mechanism in which the concentration gradient along a 52 wetted grain boundary induces a grain boundary energy 53 gradient. This causes a net flux of atoms towards the liquid 54 crack tip, which accumulate and subsequently generate 55 stresses that are able to further propagate the liquid crack. 56 According to this mechanism, no external stress is required. 57

Another similar phenomenon observed during welding 58 is related to grain boundary liquation in the so-called 59

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heat-affected zone. Because of their higher surface energy 60 or the presence of low melting temperature intermetallics 61 caused by microsegregation in the base material, grain 62 boundaries can melt at temperatures below the solidus 63 64 [11,3]. In the presence of tensile stresses, the liquid films at grain boundaries show almost no mechanical resistance 65 66 and open, thus allowing the liquid of the weld pool to penetrate. This phenomenon is thus very similar to "healed" 67 hot tears forming during solidification, i.e. when the open-68 ing of grain boundaries by tensile stresses that normally 69 leads to hot cracks can be fed by interdendritic or inter-70 71 granular liquid [12,13].

One particular case of the presence of liquid metal at the 72 grain boundaries of steel is associated with copper. Copper 73 can be present in steel as impurities, especially when recy-74 cling car bodies which contain copper electric wires, or 75 when welding is performed with copper-containing fixtures 76 or tools around the welds. During welding or hot rolling, 77 copper can penetrate along grain boundaries in some 78 austenitic or structural steels. This leaves an intergranular 79 solid copper film that dramatically decreases both the base 80 metal strength and ductility at high temperature. The "hot-81 82 shortness" induced by the presence of copper at grain boundaries is thus called copper contamination cracking 83 (CCC). The CCC is accelerated by external or residual 84 stresses [14-16], but no detailed mechanism has been given 85 in the literature. 86

87 Experimental characterization of these phenomena is usually related to a specific application. In the case of 88 LME, the loss of mechanical properties is of major con-89 cern. Therefore, tensile or compression tests at room tem-90 perature are conducted after penetration [17,18], while 91 in situ experiments using transmission electron microscopy 92 or X-ray microtomography give insights into the underlay-93 ing mechanisms [5,19]. Several tests are commonly used to 94 characterize the liquation cracking susceptibility during 95 welding. One of them, the nil-ductility test, focuses on 96 97 the temperature at which the ductility of a specimen falls to zero [20]. 98

The aim of the present study was to better understand 99 the origin of liquid gold penetration into stainless steel dur-100 ing laser welding. In transverse cross-section of the welds, 101 liquid penetration appears mainly in regions at about 45° 102 103 between the weld centerline and the top surface of the steel side. Since this location also corresponds to maximum ten-104 sile stresses during welding, it was conjectured that pene-105 tration of liquid gold into solid steel requires a tensile 106 stress state of the solid material. In order to test this 107 hypothesis at temperatures at which liquid gold is in con-108 tact with solid steel (i.e. temperature in the range 900-109 1350 °C) a new specific setup had to be developed, as stan-110 dard LME tests are not applicable. 111

112 2. Experiment

With this objective in mind, a so-called "C-ring test" 113 was selected (Fig. 1). It is a standard stress-corrosion test 114



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

in which a specimen with a "C" shape can be stressed by 115 a screw parallel to the opening of the "C" (see the figure). 116 At the symmetry plane of the C-ring, the hoop stress com-117 ponent $\sigma_{\theta\theta}$ is tensile at the outer surface while it is compres-118 sive at the inner surface [21]. Furthermore, the tensile 119 component varies from zero at the location of the screw 120 to a maximum at the symmetry plane of the C-ring, this 121 maximum being adjusted by the compression (or radial 122 stress) component at the screw location. Besides stress-cor-123 rosion cracking, such a test has also been used to study the 124 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.

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

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157 (SEM) and optical microscopy (OM). To reveal the grain boundaries of stainless steel in OM, the specimens were 158 then electro-etched in 10% oxalic acid. For one specimen, 159 a cylinder of 600, um diameter was machined near the sym-160 metry plane of the C-ring using electrical discharge 161 machining. It was then analyzed by X-ray microtomogra-162 163 phy at the European Synchrotron Radiation Facility (ESRF, Grenoble, France). 164

165 **3. Results**

A typical liquid gold crack occurring at the bottom of 166 the C-ring is shown in Fig. 2a. It always starts at a region 167 of maximal tensile stress, i.e. at the outer bottom surface of 168 the C-ring, and then propagates through its whole width 169 and thickness, as shown by X-ray microtomography (see 170 Fig. 2c). Indeed, as the tensile stress at the outer surface 171 of the C-ring is released by liquid penetration, the stress 172 173 state at the crack tip remains tensile as long as the radial stress at the level of the screw remains compressive. The 174 penetration path is essentially radial, but follows grain 175 176 boundaries that are closest to this direction. At the free 177 bottom surface of the specimen (top of Fig. 2a) as well as all along the liquid crack, an intermediate phase forms 178 (see details afterwards). It should be pointed out that liquid 179 crack initiation is not immediate and varies from one sam-180 ple to another. This is probably linked with the time 181 required to dissolve the oxide skin at the surface of the 182 specimen, despite the Ridoline pretreatment. 183

When a stressed sample is heated up to 1000 °C but 184 without immersion in gold, no crack occurs. More interest-185 ingly, when an unstressed sample is immersed in the gold 186 bath, there is also no penetration. However, strong solutal 187 remelting [25] is observed at the surface of the steel in this 188 case, with the simultaneous formation of the intermediate 189 phase at the steel-gold interface, as shown in Fig. 2b. 190 The unmelted steel is at the bottom (black region), while 191 192 the quenched gold liquid (light region) is on top of the intermediate phase (intermediate grey level). Gold liquid 193 penetration still occurs along grain boundaries (light gray 194 lines appearing in between the intermediate grey level 195 regions) but, unlike in the stressed specimen, this remains 196 197 at the local scale (typically 10 µm) of the gold-steel interface. A few intermediate phase particles are visible in the 198 quenched liquid, probably due to their movement when 199 200 quenching the specimen.

Fig. 3a shows an enlarged view of the intermediate 201 phase region (light gray region), surrounded by the 202 quenched gold liquid (light region), with the dark grey aus-203 tenite phase on the left. This intermediate phase has all the 204 205 characteristics of a peritectic phase [13]. The arrow with the 12 markers indicates points where energy-dispersive X-ray 206 207 analysis (EDX) measurements were performed, starting in 208 the quenched liquid gold phase and ending in austenite. The measured solute profiles of the six main elements 209 (Au, Ag and Cu for the 18-carat gold, Fe, Ni and Cr for 210 stainless steel) involved in the C-ring test are shown in 211



Fig. 2. (a) OM image of liquid gold penetration at stainless steel grain boundaries under an applied stress. An intermediate reaction occurs at the *s*-*l* interface, leading to the formation of an intermediate phase. (b) Backscattered electron (BSE)-SEM image of a gold-steel interface of an unstressed specimen showing the formation of the intermediate phase. The steel, intermediate phase and gold appear dark, intermediate gray and light gray, respectively. (c) Three-dimensional image of the gold liquation crack surface, as reconstructed from X-ray microtomography observations.

Fig. 3b. Focusing our attention on the main constituents, 212 i.e. Au and Fe, point 1 is close to the nominal composition 213 of 18-carat gold (70 instead of 75 wt.% Au). The Au com-214 position is nearly constant in the quenched gold liquid 215 phase (points 2 and 3) and then suddenly decreases to 216 about 10 wt.% in the intermediate phase (point 4). It expe-217 riences a slight decrease in this phase, down to 8.5 wt.% at 218 point 9. In the austenite phase (points 10-12), the gold 219

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Fig. 3. (a) Enlarged BSE-SEM view of the intermediate phase (intermediate gray level), in between the quenched liquid gold (light region) and the dark austenite. The arrow with 12 markers indicates where EDX measurements were performed. The measured profiles at those points for Au (\triangleleft), Cu (\bullet), Ag (\triangleright), Fe (\times), Ni (*) and Cr (+) are shown in (b).

composition exhibits a small diffusion profile. The Fe com-220 position shows a reverse profile, starting at about 5 wt.% in 221 the quenched gold liquid and ending at 50 wt.% in the aus-222 tenite. For the minor elements, nickel and chromium show 223 the same tendency as iron, while silver, and to some extent 224 copper, do not diffuse much into the intermediate or aus-225 226 tenite phases.

Using BSE, the gray level in the austenite phase in 227 Fig. 3a directly reflects the amount of gold [26]. Thus, the 228 brightness level profile measured at various locations 229



Fig. 4. Example of concentration profile used to estimate the time allowed for Au-diffusion (sample 7).



Fig. 5. Width of the peritectic phase at various positions along the penetration path (sample 7).

within the austenite phase, perpendicular to the interface, 230 can be used to deduce the Au solute layer. Such a measure-231 ment is shown in Fig. 4, with a complementary error func-232 tion fitted to the measured points. Such a profile allows the deduction at a given location of the time of contact between austenite and liquid gold. Repeated at various lengths along the gold-filled crack, such Au profile measurements in austenite provides a first method of estimating the speed at which liquid gold penetrates the austenite grain boundaries (see next section).

A second method is provided by the thickness of the 240 intermediate phase (light grey region in Fig. 3a). As shown 241 in Fig. 5, this thickness varies along the length of the goldfilled crack: from about 7 to 8 µm close to the outer C-ringliquid interface (where tensile stresses apply) it is only about 2 µm at the mid-thickness of the C-ring (i.e. at about 1 mm from the outer C-ring-liquid interface). In this figure, the square of the intermediate phase thickness w^2 is shown on the left scale as a function of the penetration depth y, while the correspondence to w in microns is displayed on the right scale. This $w^2 - y$ diagram is used to show the usual dependence of $w(\sqrt{t_c})$, where the diffusion time t_c is 251 given by y/v, where v is the penetration velocity of the 252 gold-rich liquid. 253

4. Discussion

In order to understand the liquid-gold-assisted liquation 255 of austenite grain boundaries, it is necessary to first con-256 sider the thermodynamics, and thus the phase diagram, 257 of the system. As there is no phase diagram available for 258 this six-element system, we will discuss the results with 259 the help of the binary Au-Fe phase diagram of the two 260 main constituents (see Fig. 6a [27,28]), but with some con-261 sideration of the effects of the other solute elements. First, 262 since the 904L stainless steel is austenitic, we discard the 263 high- and low-temperature ferritic single-phase regions. 264 Second, as the liquidus of 18-carat gold is about 900 °C, 265 i.e. about 164 °C below the melting point of pure gold, 266 the peritectic invariant observed at 1173 °C in the binary 267

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Fig. 6. (a) Phase diagram of the binary Au-Fe diagram [28,27]. The dashed line is the apparent temperature of the melt translated by 164°C to account for the composition of the 18-carat gold (see text). (b) Diffusion couple at the gold-steel interface at time t. Interfaces are assumed to be at equilibrium. With time, the concentration profile in γ -Fe evolves according to Eq. (1) and the width of the peritectic phase w increases as stated by Eq. (4).

Au-Fe system is also assumed to occur 164 °C lower, i.e. at 268 1009 °C, in the case of the 18-carat gold-steel system. 269 Alternatively, a dashed line in Fig. 6a has been drawn 270 164 °C higher than the bath temperature of the C-ring test, 271 i.e. at 1164 °C. Although its position is debatable, it must 272 be below the invariant temperature (in this case only 9 °C 273 274 below) in order to explain the formation of the intermedi-275 ate (peritectic) phase between the austenite and the quenched liquid. On the other hand, as the composition 276 277 of gold within the peritectic phase is fairly uniform (see Fig. 3b), it must also be close to the peritectic temperature. 278 Based on the experimental results and with the help of 279 280 the Au-Fe binary phase diagram, a mechanism for the initiation and propagation of grain boundary liquation can be 281 282 proposed. When liquid gold comes into contact with stainless steel, solutal remelting occurs together with the forma-283 tion of a peritectic phase. Therefore, liquid gold must 284 285 dissolve iron, while gold diffuses into solid austenite and 286 the peritectic phase nucleates and forms in between. This explains the grey-level contrast of the various phases seen 287 in the BSE pictures of Figs. 2b and 3a. Remelting can pro-288 gress faster at grain boundaries, since the diffusion of gold 289

290 is faster than in the bulk, and the remelting of grain bound-291 aries occurs earlier because of their high energy. This explains the formation of a liquid gold film, especially at 292 293 high energy grain boundaries, in both stressed and unstressed specimens. 294

The dramatically enhanced penetration of gold at grain 295 296 boundaries of stressed specimens is still not fully under-297 stood: it could be due to enhanced diffusion, as suggested by Klinger [10], but also to liquid suction in the crack dur-298 ing crack opening. Indeed, as the liquid gold enters into 299 contact with a new steel grain boundary, the peritectic 300 301 phase forms while the liquid becomes enriched or even saturated with iron (nickel and chromium being omitted for 302 the sake of clarity). When no tensile stress is applied, this 303 304 is a purely diffusive process that is controlled by the rate

doi:10.1016/j.actamat.2011.06.030

at which new "fresh" unsaturated liquid gold reaches the 305 grain boundary. However, tensile stress, which is intensi-306 fied at the tip of the liquid-filled crack, combined with dis-307 solution of iron into the liquid at the crack tip, makes it 308 move forward while the two sides of the crack open more. 309 This sucks new liquid by convection, dramatically enhanc-310 ing the apparent diffusion coefficient of gold along the 311 grain boundaries. The path follows grain boundaries, 312 because the replacement of a large-misorientation grain 313 boundary in austenite by two solid-liquid interfaces with 314 a partially iron-saturated gold liquid is energetically favor-315 able. However, if the grain boundary energy determines the 316 local path of the crack, its overall propagation is mainly 317 driven by the stress state in order to release the maximum 318 strain energy. 319

Based on SEM-EDX image analysis, two fairly simple 320 estimations of the liquid gold penetration velocity can be 321 conducted for the C-ring test. Looking at Fig. 6b, which 322 is a sketch of the transverse profile shown in Fig. 3, the 323 peritectic phase is exactly like a diffusion couple: it is 324 located between the liquid phase and the γ -austenite phase. 325 Note that the peritectic phase, labeled γ -Au in Fig. 6(b), is 326 also face-centered cubic. Interesting solid-state transforma-327 tions in the Au-Fe system (e.g. continuous and discontinu-328 precipitation) occurring below the ous peritectic 329 temperature have been analyzed in Refs. [1,26]. Neglecting 330 the time for nucleation of the peritectic phase (since it 331 propagates along the crack boundaries), the thickness of 332 the peritectic γ -Au phase (Fig. 5) and the width of the 333 Au diffusion profile in the austenite phase (Fig. 4) can both 334 be used to estimate the time allowed for Au-diffusion, i.e. 335 the time separating the end of the experiment minus the 336 time at which the liquid gold reached the position of the 337 sample where the transverse profile is analyzed. Consider-338 ing the time for diffusion at several known positions along 339 the crack, the propagation velocity of the crack can be 340 deduced.

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342 The assumptions made for these two types of calculations are as follows: 343

- The two interfaces γ -Fe/ γ -Au and γ -Au/liquid are at 344 345 equilibrium and their kinetics is only driven by solute diffusion. 346
- 347 • The composition $C_{\text{(Au)}}$ in the liquid is uniform (i.e. complete mixing), considering the fast diffusion in the liquid 348 and the width of the crack (i.e. large Fourier number). 349
 - The formation of the peritectic phase is instantaneous once liquid gold reaches a position.
 - The compositions at the two interfaces, $C_{\underline{\ell}Au}$, $C_{\underline{A}u\ell}$, C_{AuFe} and C_{FeAu} are given by the EDX measurements (see Fig. 3b and Table 1). They are constant over time.
 - The Au-profile in austenite is not affected by the movement of the γ -Au/ γ Fe interface.
- The initial conditions at a given location are: no gold in 357 the austenite phase, thickness of the peritectic phase 358 arbitrarily small and liquid composition given by $C_{\ell Au}$. 359 Even though the diffusion of gold into austenite and 360 the formation of the γ -Au peritectic phase gradually 361 362 pumps gold from the liquid and could possibly lead to the solidification of the crack region, it is assumed that 363 the flow of fresh liquid gold induced by the opening of 364 the crack is sufficient for the liquid phase to always 365 remain. 366
- Preferential diffusion along grain boundaries of the γ -Fe 367 and γ -Au phases and perpendicular to the crack is not 368 considered - or, more specifically, since the diffusion 369 coefficient at grain boundaries is unknown, it is included 370 in an averaged bulk diffusion coefficient, which is not 371 accurately known. 372

Under these hypotheses, the first method, based on the 374 diffusion profile of Au in the austenite phase, is based on 375 the standard diffusion equation into a semi-infinite 376 medium: 377 378

$$C(y, t_c) = C_{\text{FeAu}} \text{erfc}\left(\frac{y}{2\sqrt{D_{\text{Fe}}t_c}}\right)$$
(1)

where y is the transverse distance in the γ -Fe phase mea-381 sured from the γ -Fe/ γ -Au interface, t_c is the time allowed 382 for diffusion, in this case the time of contact between liquid 383 gold and austenite, erfc(u) is the complementary error func-384 tion and $D_{\rm Fe}$ the interdiffusion coefficient of Au in the γ -Fe 385 phase. From the concentration profile of gold in austenite 386 (see Fig. 4), Eq. (1) allows the time t_c to be deduced if 387 one knows $D_{\rm Fe}$. Repeating this measurement at various dis-388 389 tances along the gold-filled crack gives the speed of liquid penetration. 390

The second model is based on the diffusion couple that 391 392 forms during the peritectic reaction, more precisely on

Table 1 Interfacial compositions of Au (in wt.%). $C_{\ell Au}$ C_{FeAu} $C_{Au\ell}$ C_{AuFe} 70 8.5 2.5 11

the thickness $w(t_c)$ of the y-Au phase. Assuming that the 393 interfacial compositions are fixed (see Table 1), the two 394 interfacial solute balances while diffusion of gold is occur-395 ring are given by: 396

$$\frac{dy_1}{dt} = \frac{-D_{Au}}{C_{Au\ell} - C_{\ell Au}} \left(\frac{C_{AuFe} - C_{Au\ell}}{y_2 - y_1} \right)$$

$$\frac{dy_2}{dt} = \frac{1}{C_{FeAu} - C_{AuFe}} \left[-D_{Fe} \left[\frac{dC}{dy} \right]_{y_2} + D_{Au} \left(\frac{C_{AuFe} - C_{Au\ell}}{y_2 - y_1} \right) \right]$$
(2)
$$(3) \qquad 399$$

where $y_1(t)$ and $y_2(t)$ are the positions of the two interfaces 400 on each side of the γ -Au peritectic phase (see Fig. 6b). We 401 have assumed that the Au-solute profile in the peritectic 402 phase is linear, while the gradient of Au in the austenite 403 phase at the interface y_2 can be deduced from Eq. (1). Sub-404 tracting Eq. (2) from Eq. (3) gives the evolution law of the 405 thickness $w(t) = (y_2(t) - y_1(t))$: $\frac{406}{407}$

$$\frac{dw}{dt} = \frac{a}{w(t)} + \frac{b}{\sqrt{t}} \tag{4}$$

with

$$a = D_{Au}(C_{AuFe} - C_{Au\ell}) \left(\frac{1}{C_{FeAu} - C_{AuFe}} + \frac{1}{C_{Au\ell} - C_{\ell Au}} \right)$$
$$b = \sqrt{\frac{D_{Fe}}{\pi}} \frac{C_{FeAu}}{C_{FeAu} - C_{AuFe}}$$
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The first term on the right-hand side of Eq. (4) is the driving force for diffusion across the peritectic phase and is proportional to the composition difference $(C_{Au\ell} - C_{AuFe})$. In the binary phase diagram of Fig. 6a, this Au composition difference is the length of the segment on the dashed line between the γ -Au solidus and the γ -Au solvus.

The second term in Eq. (4) slows down the growth of the peritectic phase by diffusion in the austenite. Setting the initial condition w(0) = 0, Eq. (4) admits the particular solution:

$$w(t) = \left(\sqrt{b^2 + 2a} + b\right)\sqrt{t} \tag{5}$$

A more general solution, with any arbitrary initial condition, can be found with Mathematica[®], but it is then implicit and quite complex.

While the first method is only dependent on the value of $D_{\rm Fe}$, the second one, based on the thickness of the peritectic phase, is a function of both D_{Au} and D_{Fe} . Indeed, when $a \gg b^2$ (i.e. $D_{Au} \gg 0.1 D_{Fe}$, using values from Table 1), Eq. (5) can be simplified to $w(t) \approx \sqrt{2at}$, in which w(t) is independent of $D_{\rm Fe}$.

Furthermore, an estimation of these diffusion coeffi-437 cients is not straightforward, as the self-diffusion coeffi-438 cients of Au and γ -Fe at 1000 °C (resp. 10^{-13} m² s⁻¹ and 439 $10^{-16} \text{ m}^2 \text{ s}^{-1}$) are different by three orders of magnitude, 440 making the classical Boltzmann-Matano analysis unappli-441 cable [29]. Fig. 7 shows the penetration velocity of liquid 442

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Fig. 7. Calculated penetration velocity as a function of D_{Au} and D_{Fe} for sample 7.

gold deduced for one of the C-ring specimens using this 443 second method, as a function of both diffusion coefficients. 444 This two-dimensional plot is only theoretical since the time 445 is limited by the total time of the experiment. Knowing that 446 the whole thickness of the C-ring is 2 mm and that the total 447 immersion time of this specimen is 480 s, a minimum veloc-448 ity of 4.17 μ m s⁻¹ is first deduced (line drawn in Fig. 7). On 449 the other hand, a maximum bound of this velocity has been 450 set by considering realistic values of the solid-state diffu-451 sion coefficients ($D = 10^{-12} \text{ m}^2$ [29]). Therefore, a realistic 452 value of the gold liquid velocity at austenite grain bound-453 aries can be obtained for $D_{Au} = 10^{-12} 10^{-13} \text{ m}^2 \text{ s}^{-1}$. A value of $D_{Au} = 3 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ was selected for this sec-454 455 ond method. 456

Using this coefficient, the liquid penetration velocity 457 deduced from the peritectic thickness at various positions 458 along the liquid-filled crack becomes a weakly dependent 459 function of $D_{\rm Fe}$. It is shown for two specimens with open 460 circles and triangles in Fig. 8. The corresponding filled 461 symbols for the same specimens have been obtained with 462 the austenite diffusion profiles method (i.e. the results for 463 sample 7 are calculated with data from Fig. 4 and Eq. (1) 464 for method 1 and data from Fig. 5 and Eq. (4) for method 465 2). Reasonable agreement between the two methods is 466 obtained for $D_{\rm Fe} = 3 \times 10^{-14} \,{\rm m}^2 \,{\rm s}^{-1}$. With such values, 467 the penetration velocity of gold is only a few microns per 468



Fig. 8. Calculated penetration velocities as a function of $D_{\rm Fe}$ for two samples $(D_{Au} = 3 \times 10^{-13} \text{ m}^2 \text{ s}^{-1} \text{ for method } 2)$.

second. This value is considerably smaller than that associated with laser welding [1,26]. Indeed, in laser welding, gold 470 cracks of typically 10–100 um have been observed in the regions of maximum tensile strains. Considering the solid-472 ification time associated with spot laser welding (a few mil-473 liseconds), a velocity in the range $0.01-0.1 \text{ m s}^{-1}$ can be 474 deduced. This large discrepancy is probably due to the much larger stresses/strains involved in this process compared to the C-ring test.

5. Conclusion

A new setup has been proposed for the study of inter-479 granular penetration of a liquid metal, in this case gold, 480 into a solid subjected to tensile stresses, in this case a C-ring 481 made of superaustenitic stainless steel. It has been shown 482 that, without tensile stresses, remelting is localized at the 483 C-ring-liquid interface. When the C-ring is compressed 484 by a screw, liquid penetration occurs along grain bound-485 aries, starting from the region under maximum tensile 486 stresses and propagating inward. Measuring the thickness 487 of the peritectic phase and of the gold diffusion profile in 488 austenite, a penetration velocity in the range of a few 489 microns per second has been deduced. This value is much 490 lower than that occurring during laser welding, probably 491 as a result of a much less severe stress state. Although 492 the C-ring offers a very interesting mean of observing and 493 understanding grain boundary liquation, more accurate 494 measurements are required: (i) to obtain reliable diffusion 495 coefficients in the phases; (ii) to eliminate any "offset" times 496 associated with the possible formation of an oxide layer at 497 the surface of the C-ring prior to its immersion; and (iii) to 498 know the precise stress state of the C-ring. The component 499 itself, as well as the compressive screw, certainly creep dur-500 ing the test, thus releasing part of the stress imposed before 501 the immersion test. High-temperature gauges could be used 502 to eliminate this problem. 503

Acknowledgements

Electron microscopy analyses were performed at the CIME, Interdisciplinary Centre for Electron Microscopy (EPFL). The authors are grateful to all staff members of the ID 19 beamline of ESRF for assistance with X-ray tomography experiments, within the framework of the project ANR-05-BLAN-0286-01 TOMOSOLIDAL supported by the Agence Nationale de la Recherche.

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Please cite this article in press as: Favez D et al. Intergranular penetration of liquid gold into stainless steel. Acta Mater (2011), doi:10.1016/j.actamat.2011.06.030

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