Oscillatory Marangoni Flow: A Fundamental Study by Conduction-Mode Laser Spot Welding

Through Marangoni flow, a surface-active agent can affect not only the weld pool depth, but also the pool surface deformation, pool surface oscillation, and ripple formation

BY S. KOU, C. LIMMANEEVICHITR, AND P. S. WEI

ABSTRACT

Marangoni flow, a fundamental subject extensively studied in welding, was further studied by using conduction-mode (non-keyhole) laser spot welding of 304 stainless steels. It was discovered that a surface-active agent, such as sulfur (S) in stainless steels, can affect not only the weld pool depth as explained by Heiple and Roper’s model, but also the pool-surface deformation, pool-surface oscillation, and ripple formation. With low S (42 ppm), the pool surface was concave and oscillatory, and the resultant weld was shallow with clear ripples. With high S (140 ppm), however, the pool surface was convex and nearly steady, and the resultant weld was deeper without clear ripples. A mechanism was proposed to explain these strikingly different phenomena. At low S the fast outward surface flow can make the pool surface concave, as shown by computer simulation. The raised surface near the pool edge is unstable; it can oscillate with oscillatory Marangoni flow, disturb solidification at the pool edge, and cause clear ripple formation. At high S, the fast inward surface flow can make the pool surface convex. Oscillation of the raised surface near the pool center, however, may not disturb solidification at the pool edge enough to cause clear ripple formation. The mechanism was verified by observing pool-surface oscillation and ripple formation immediately after turning off the laser. Furthermore, oscillatory Marangoni flow was demonstrated by flow visualization in simulated stationary weld pools of NaNO₃. Surface oscillation induced by oscillatory Marangoni flow was demonstrated with a NaNO₃ drop laser heated from above.

Introduction

Weld Pool Marangoni Flow

The weld pool surface is warmer under the heat source and cooler near the pool edge. The gradients of surface temperature (T) can induce gradients of surface tension (γ) along the pool surface because γ depends on T. Since the higher-γ liquid tends to pull the lower-γ liquid toward it-

S. KOU (kou@engr.wisc.edu) is professor, Department of Materials Science and Engineering, University of Wisconsin, Madison, Wisc.; C. LIMMANEEVICHITR is associate professor, Production Engineering Department, King Mongkut’s University of Technology Thonburi, Bangkok, Thailand; and P. S. WEI is professor, Department of Mechanical and Electro-Mechanical Engineering, National Sun Yat-Sen University, Kaohsiung, Taiwan.

KEYWORDS

Marangoni Flow
Laser
Weld Pool
Surface Tension
Thermocapillary
Stainless Steel
has been confirmed by numerous welding experiments (e.g., Refs. 1, 2, 5, 6). It has also been confirmed by physical modeling (Refs. 7–9) as well as computer modeling (e.g., Refs. 10–12). Limmaneevichitr and Kou (Ref. 7) developed a flow visualization technique to reveal steady Marangoni flow in a simulated stationary weld pool of NaNO₃, which is transparent and well documented in physical properties. They further revealed the reversal of Marangoni flow in the presence of a surface-active agent (Ref. 9). Kou and Sun (Ref. 10) developed the first computer model of heat transfer and fluid flow to calculate the unknown weld pool shape. The computer model confirmed the effect of the surface-active agent on the weld pool depth proposed by the model of Heiple and Roper. Using body-fitted curvilinear coordinates to more accurately handle boundary conditions at the pool surface, Tsai and Kou (Refs. 13, 14) demonstrated the deformation of the weld pool surface caused by Marangoni flow and density changes. They showed that fast outward surface flow (in the absence of a surface-active agent) can make the pool surface concave, which has been confirmed by more recent computer simulations (e.g., Ref. 15). They also showed that assuming a flat rigid pool surface that is not deformable can cause significant errors in calculating the weld pool depth. Tsai and Kou (Ref. 14) demonstrated the deformation of the weld pool surface induced by volume expansion caused by melting and superheating.

Hong et al. (Refs. 16, 17) developed a comprehensive thermofluid model of gas-tungsten arc spot welds with a free surface geometry and a properly posed turbulence model. They showed that predicted mean flow velocities and final weld pool dimensions in a relatively low-thermal conductivity material such as 304 stainless steel were significantly affected by the sulfur content and turbulence. He et al. (Ref. 18) and Paul et al. (Ref. 19) demonstrated in welding with a focused laser beam that the free surface can be deformed by the recoil of the weld pool.

### Table 1 — Compositions of 304 Stainless Steels (wt-%)

<table>
<thead>
<tr>
<th></th>
<th>%C</th>
<th>%S</th>
<th>%P</th>
<th>%Mn</th>
<th>%Si</th>
<th>%Ni</th>
<th>%Cr</th>
<th>%Mo</th>
<th>%Co</th>
<th>%Cu</th>
<th>%N</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low Sulfur</td>
<td>0.020</td>
<td>0.0042</td>
<td>0.017</td>
<td>1.80</td>
<td>0.44</td>
<td>8.12</td>
<td>18.38</td>
<td>0.05</td>
<td>0.05</td>
<td>0.09</td>
<td>0.061</td>
<td>—</td>
</tr>
<tr>
<td>High Sulfur</td>
<td>0.072</td>
<td>0.0140</td>
<td>0.038</td>
<td>1.76</td>
<td>0.48</td>
<td>8.12</td>
<td>18.20</td>
<td>0.38</td>
<td>0.10</td>
<td>0.37</td>
<td>0.051</td>
<td>—</td>
</tr>
</tbody>
</table>
pressure caused by evaporation of material from the pool surface. The only study on oscillatory Marangoni flow in the weld pool known to the authors is the computer simulation of Morvan and Bournot (Ref. 20) for conduction-mode laser welding (that is, without a keyhole), in which the pool was assumed two-dimensional (i.e., sheet-like), normal to the travel direction, and with a rigid pool surface (i.e., a straight line). An oscillation frequency of 10 to 20 Hz for aluminum or steel was suggested. It was stated “in the real cases where the free surface can be deformed, the oscillations in the melted pool will be suppressed.”

Weld Pool Oscillation

Weld pool oscillation can be used for process monitoring and control during welding. For instance, the oscillation frequency decreases significantly if the weld pool changes from partial to complete joint penetration during welding. When acted upon momentarily by an external force, such as a pressure pulse caused by a current pulse in pulsed gas tungsten arc welding, the pool tends to vibrate at natural frequencies that are related to the dimensions, penetration, and physical properties of the pool (Ref. 21). The natural frequency and amplitude of the oscillation can be measured by monitoring the arc, for instance, its voltage variation (Refs. 22, 23). Models have been developed to predict the frequencies of oscillation for various modes of oscillation (Refs. 21–27). It has been shown that the frequency of surface oscillation is proportional to $D^{-3/2}$, where $D$ is the pool surface diameter (Refs. 22, 23). Thus, the frequency can increase sharply with decreasing pool diameter.

Ripple Formation

Ripple formation has been an interesting topic of research in welding. In the classic work of Kotecki et al. (Ref. 28), ripples on complete joint penetration spot welds made by gas tungsten arc welding of thin (e.g., 1.27 mm or 0.050 in.) metal sheets were studied. Turning off the arc suddenly released the arc pressure that was “stretching the pool surfaces, setting the pool into oscillation like a drumskin.”

A one-to-one correspondence between ripples and pool oscillations was identified, and it was concluded that pool oscillations during freezing produced the weld surface ripples. The ripple spacing decreased from near the weld edge to the center. Surface oscillation of partial-penetration pools in a thicker workpiece was also confirmed. The arc pressure caused the pool surface to depress near the center and rise near the pool edge. The pool surface was set into oscillation when the arc pressure was suddenly released upon turning off the arc. Rappaz et al. (Ref. 29) studied ripples on partial-penetration spot welds of Fe-15Ni-15Cr single crystals made by gas tungsten arc welding. The weld pool, typically hemispherical in shape with a diameter of about 6 mm, solidified with ripples on the surface upon turning off the arc. The ripple spacing and amplitude both decreased from near the weld edge to the center. It was shown that the ripple spacing was equal to the solidification (growth) rate times the period of pool oscillation, that is, the growth rate divided by the frequency of pool oscillation.

The present study deals with the effect of the surface-active agent on the pool surface deformation, pool-surface oscillations, and pool oscillation frequency. The present study deals with the effect of the surface-active agent on the pool surface deformation, pool-surface oscillations, and pool oscillation frequency.
tion, and ripple formation induced by Marangoni flow in the weld pool. It may be considered as a complement to the model of Heiple and Roper (Ref. 1), which explains the effect of the surface-active agent on the weld pool depth based on Marangoni flow. It focuses on stationary weld pools, aiming to understand the basic case of stationary weld pools first before dealing with the more complicated case of moving weld pools (Refs. 30–32).

Experimental Procedure

Welding

Weld pool phenomena were studied by using conduction-mode laser spot welding of 304 stainless steels. In order to study Marangoni flow in the weld pool properly, interference from other driving forces (Ref. 2) for fluid flow must be excluded. In the case of gas tungsten arc welding, for instance, they can include arc factors such as the Lorentz force, the arc forces (the arc pressure and the shear force), and anode spot wandering. Unlike the keyholing mode, in the conduction mode, the workpiece surface is positioned below or above the focal point of the laser beam. The beam can thus be defocused to a size comparable to a gas-tungsten welding arc, e.g., 6.4 mm (¼ in.) in diameter. This will allow the results to be compared with those of arc welding in future studies to infer what arc effects are occurring. Conduction-mode laser- and electron-beam welding have been a useful tool for studying the effect of Marangoni flow on the weld pool depth and ripple formation associated with moving pools (Refs. 5, 30–32). In order to gain the fundamental understanding of Marangoni flow and its effect, however, the weld pools were stationary in the present study. The results from stationary weld pools can serve as the foundation for understanding the more complicated phenomena associated with moving weld pools, which will be dealt with in follow-up reports.

To study the effect of the sulfur (S) content on weld pool phenomena, two heats of 304 stainless steels were welded, one with a lower S level of 42 ppm and the other a higher S level of 140 ppm. For convenience of discussion, the former will be called low S and the latter high S. The compositions of the stainless steels are shown in Table 1. Both stainless steels were 300 mm (12 in.) long, 50 mm (2 in.) wide, and 6.4 mm (¼ in.) thick.

A HAAS HL3006 YAG laser machine was used for welding. The maximum power capacity was 3000 W. The wavelength was 1064 nm. The distance between the laser head and the workpiece was about 140 mm. The laser beam was tilted about 7 deg off the vertical line in order to keep the beam from being reflected back to the focusing lens. The laser power ranged from 1800 to 3000 W and the welding time from 1 to 8 s.

To help determine if the pool surface was concave or convex, the light from a halogen lamp was directed through a fiber-optic bundle onto the pool surface as illustrated in Fig. 2. The distance between the tip of the fiber-optic bundle and the weld pool was about 60 mm. The pool surface acted as a mirror. The image of the light (white) appears larger on a concave pool surface (Fig. 2A) and smaller on a convex one (Fig. 2B). A moving image suggests an oscillatory pool surface. Welding was videotaped with a CCD camera at 30 frames per second, with a close-up lens for observing the weld pool surface.

The resultant welds were photographed to show the ripples on the surfaces. They were then cut, polished, and etched to reveal the vertical cross sections of welds.
The etching solution consisted of 50 mL HCl, 10 g CuSO₄, and 50 mL H₂O.

**Oscillatory Marangoni Flow**

Flow visualization was conducted in a simulated stationary weld pool of sodium nitrate (NaNO₃). NaNO₃ was selected for the following reasons. First, it has a transparent melt, a surface tension that decreases significantly with increasing temperature, a low melting point, and well-documented physical properties, as shown in Table 2 (Refs. 7, 33, 34). Second, the Marangoni number Ma for the simulated weld pool of NaNO₃ can be close to those for steel and aluminum weld pools (Ref. 7). According to the similarity law of hydrodynamics, similarity in Marangoni flow between two fluid systems can be expected if the Marangoni numbers are close to each other (Ref. 34). Third, NaNO₃ has a transmission range of 0.35 to 3 µm and is, therefore, opaque to CO₂ laser (10.6 µm wavelength) just like a metal weld pool is opaque to the heat source. However, it is transparent to the He-Ne laser (0.633 µm wavelength), which as will be described subsequently, was used for flow visualization. The purity of the NaNO₃ used was above 99%.

The apparatus for flow visualization is illustrated in Fig. 3. The procedure for flow visualization in simulated weld pool of NaNO₃ has been described by Limmaneевичирт et al. (Refs. 7–9) and will be mentioned only briefly here. The NaNO₃ pool for flow visualization was held in a cylindrical glass container of 5 mm inner radius, which was a test tube cut short. The pool was essentially hemispherical in shape except its top surface was concave instead of flat. The pool was heated from above by a CO₂ laser beam positioned at the center of the pool surface to induce Marangoni flow. The beam diameter at the pool surface was 1.5 mm, and the power ranged from 2 to 14 W. The pool was surrounded by a bath of NaNO₃ melt held in a rectangular glass container, which acted as a transparent heater for the light sheet to pass through.

A laser light-cut technique was used for flow visualization. A laser light sheet was produced with the help of a 20-mW He-Ne laser and optical lenses. The light sheet was oriented such that it intersected the NaNO₃ pool to form a flat (horizontal) plane at 0.5 mm below the bottom of the pool surface. It was found that the pattern of flow oscillation was easier to recognize from above the pool than from in front of the pool. So, instead of a vertical light sheet cutting through the pool axis as in the previous studies (Refs. 7–9), a flat light sheet was used. Since the pool surface is concave, the surface flow along the pool surface cannot be revealed by a light-cut technique. So, the pattern of the return flow in a flat plane at 0.5 mm below the bottom of the pool surface was revealed by flow visualization.

Aluminum particles 20 µm in diameter were added to the NaNO₃ pool to serve as a tracer to reveal the flow pattern. The particles were illuminated by the He-Ne laser light sheet and hence revealed the flow pattern in the pool. The density of aluminum (2.7 g cm⁻³) is greater than that of the NaNO₃ melt (1.9 g cm⁻³). From Stokes's law (Ref. 35), however, the settling velocity is much slower than Marangoni flow in view of the small particle diameter.

To detect temperature oscillation in the NaNO₃ pool caused by flow oscillation, a
The thermocouple was lowered into the pool after flow visualization was finished. The thermocouple was K-type (Omega HJMQSS-010-E-6) with a stainless steel sheath. The overall diameter of the thermocouple was 0.25 mm (0.01 in.). The thermocouple tip was at a radial position of 2.5 mm from the pool axis and at a vertical position of 0.5 mm below the bottom of the pool surface, where flow visualization was conducted. A data acquisition system (Omega WB-Dynares 16 Ultra) was used to record temperature data at the sampling rate of 10 Hz. The data were analyzed by Fast Fourier Transformation (Statistica Version 5.1’97).

Surface Oscillation Induced by Oscillatory Marangoni Flow

To demonstrate that oscillatory Marangoni flow can cause surface oscillation, a drop of NaNO₃ was supported by a graphite rod at the bottom and heated by a CO₂ laser beam from above, as illustrated in Fig. 4. The beam diameter was 1.5 mm at the top surface of the drop. Marangoni flow in the drop was revealed by using a flow visualization technique similar to that described previously. The flow pattern was revealed with a vertical light sheet of the He-Ne laser that cut the drop through its axis. The flow pattern was optically distorted by the lens effect of the drop.

Results and Discussion

Welds and Ripples

A significant effect of the sulfur content of 304 stainless steel on the ripple formation on its welds was observed. More than a dozen spot welds were made, ranging from 1800 to 3000 W and 1 to 8 s. The results consistently show significantly clearer ripple formation on the low-S welds than the high-S welds. Typical results will be shown in the following paragraphs. The effect of welding conditions on ripple formation will be discussed elsewhere. A clear effect of the sulfur content on the ripple formation on the seam welds of the same 304 stainless steels was also observed. The results will be reported elsewhere.

Figure 5 shows two spot welds made at the power level of 2200 W for a welding time of 1.3 s. Clear ripples are visible on the low-S weld (Fig. 5A) but not on the high-S weld (Fig. 5C). The ripples on the low-S weld (Fig. 5A) tend to be clearer and more widely separated near the weld edge than near the weld center, that is, the ripple spacing decreases from the weld edge to the weld center. These differences are consistent with the observation of Kotecki et al. (Ref. 28) and Rappaz et al.

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Fig. 9 — Pool-surface deformation of a stationary laser weld pool of 6061 Al caused by density decrease due to melting (ρ_S < ρ_L) and superheating (β > 0) (Ref. 14). Laser power: 1800 W; steady state.

Fig. 10 — Mechanism explaining how surface-active agent affects pool-surface deformation, pool-surface oscillation, and ripple formation through Marangoni flow: A — Low surface-active agent; B — immediately after turning off laser in A; C — high surface-active agent; D — immediately after turning off laser in C.

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Rappaz et al. (Ref. 29) observed in gas tungsten arc spot welding of Fe-15Ni-15Cr that the growth rate increased, slightly more rapidly than linearly, from about 4 mm/s at the weld edge (3 mm in radius) to about 6 mm/s at 1 mm from the weld center. Xiao and den Ouden (Refs. 22, 23) showed that the frequency of pool-surface oscillation is proportional to (pool radius)–3/2. Thus, as the radius decreases from the weld edge to the weld center, the growth rate seems to increase less rapidly than the pool-oscillation frequency. Since the ripple spacing is the growth rate divided by the pool-oscillation frequency (Ref. 29), it seems reasonable to expect the ripple spacing to decrease from the weld edge to the weld center.

The transition from columnar grains at the weld edge to equiaxed grains near the center (Fig. 5A) is likely to be caused by the decrease in the G/R ratio from the pool edge to the pool center, where G is the temperature gradient in the liquid at the solidification front and R the growth rate (Ref. 2). Upon turning off the laser, the temperature near the pool center dropped sharply, causing G to decrease and R to increase during solidification from the pool edge to the center. As already mentioned, Rappaz et al. (Ref. 29) showed that the growth rate increases from the pool edge to the pool center.

As expected, a significant effect of the sulfur content on the weld shape was observed. The low-S weld (Fig. 5B) is slightly larger in diameter but significantly shallower as compared to the high-S weld (Fig. 5D). These weld shapes are consistent with Heiple’s model — Fig. 1.

Figure 6 shows two larger welds made at a higher power of 2800 W for a longer welding time of 5 s. Again, clear ripples are visible on the low-S weld (Fig. 6A) but not the high-S weld (Fig. 6C). Overall speaking, the ripples on the low-S weld tend to be clearer and more widely separated near the weld edge than near the weld center. The low-S weld (Fig. 6B) is slightly larger in diameter but much shallower as compared to the high-S weld (Fig. 6D). These results are consistent with those shown previously in Fig. 5.

The small depression at the center of the weld surface (Fig. 6D) does not mean the existence of a keyhole during welding. In fact, the weld surface after welding does not necessarily represent the pool surface during welding. As will be shown subsequently (Fig. 7F–J), the pool surface was convex everywhere and there was no keyhole at the center. The absence of a keyhole can be expected because the laser beam was defocused (to 6.4 mm in diameter). The depression formed during the last moment of weld pool solidification, when the solidification front was near the center. The formation of the depression is likely to be caused by the conservation of mass. The crown of the weld in Fig. 6D above the workpiece surface suggests the overall density of the material within the fusion boundary decreases after welding. Even so, the fact that the weld surface rises well above the workpiece surface from the beginning of solidification suggests that there might not be enough material left in the pool to sustain the high weld surface throughout solidification. As shown in Fig. 5D, on the other hand, the weld surface is essentially even with the
Fig. 13 — Flow oscillation in NaNO₃ pool revealed by flow visualization of return flow at 0.5 mm below pool surface (Fig. 3). A — Steady flow at 4-W laser power; B — oscillatory flow at 6 W; C — oscillatory flow at 14 W.

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workpiece initially, rises gradually as solidification proceeds, and ends up without a depression.

Surface Deformation and Oscillation of Weld Pools

The evidence of the deformation and oscillation of the weld pool surface was observed in 304 stainless steel. The videotapes recorded during spot welding revealed wild surface oscillation in the case of low-S pools but little surface oscillation in the case of high-S pools. The oscillation of the low-S weld pool contradicts the statement of Morvan and Bournot (Ref. 20) “in the real cases where the free surface can be deformed, the oscillations in the melted pool will be suppressed.”

Figure 7 shows the photos of stationary weld pools extracted from the videotape recorded during welding. The diameter of the low-S pool is 8.5 mm and that of the high-S pool 7.3 mm (in Fig. 7 and, subsequently, Figs. 11 and 12), based on the welds shown in Fig. 6. The surface of the high-S weld pool (Fig. 7F–J) appears convex. The fact that the image of the halogen light (white spot) is much smaller than the pool surface further suggests that pool surface is convex. Although the light images in Fig. F–J are similar, close examination can still reveal some differences. For instance, the images in Fig. 7F and I appear slightly thinner (more elongated in the left-right direction) than that in Fig. 7G. These differences suggest that the pool surface oscillates near the center and the amplitude of oscillation is small. As for the low-S weld pool (Fig. 7A–E), the light image varies in shape clearly from frame to frame. This suggests that the pool surface is clearly oscillating. With high-speed photography, it would be possible to determine the frequency and mode of oscillation. Furthermore, the light image is much larger than that in the high-S pool, covering nearly the whole pool surface except near the pool edge, which appears much darker. This suggests that the pool surface is concave and oscillating.

The concave pool surface with low S (Fig. 7A–E) is most likely caused by Marangoni flow instead of the recoil pressure of the material evaporating from the pool surface. Otherwise, the high-S pool surface (Fig. 7F–J) would also have to be concave instead of convex. The defocused laser beam might have helped keep the beam power density low to avoid recoiling.

Computer simulation has demonstrated that Marangoni flow in the weld pool can cause the pool surface to deform in conduction-mode laser spot welding of 6061 Al alloy (Ref. 13) and steel (Ref. 15). Quantitative discussion of oscillatory Marangoni flow in 304 stainless steel and its effect on pool-surface deformation and oscillation is difficult without a computer model that can handle both flow and surface oscillation. Also, turbulence may need to be considered. As compared to the case of laminar flow, as shown by Hong et al. (Ref. 17), with turbulence the maximum velocity can be lower and the pool shallower. Since such a computer model does not yet exist, the discussion here can only be qualitative. In the following discussion, the results of computer simulation of conduction-mode laser spot welding by Tsai and Kou (Refs. 13, 14) will be used even though the workpiece material was 6061 Al alloy instead of 304 stainless steel. This is mainly because they demonstrated the individual effects of the following factors on pool-surface deformation: 1) dγ/dT < 0; 2) dγ/dT > 0; and 3) volume expansion (due to melting and superheating). The following discussion would be difficult without referring to these results.

In Fig. 8, Tsai and Kou (Ref. 13) show the surface deformation of stationary conduction-mode laser weld pools of 6061 aluminum alloy caused by Marangoni flow. The beam diameter is 8 mm at the laser power is 1800 W, and the power is on continuously to allow the weld pool to reach the steady state. Under the normal condition of dγ/dT < 0 (Fig. 8A), the outward surface flow is very fast, on the order of 1 m/s. Near the pool edge, however, it is slowed down suddenly by the edge. According to Bernoulli’s principle (Refs. 31, 35), the fluid pressure can increase as the result of a velocity decrease. Thus, near the pool edge the pressure can be expected to increase and push the pool surface upward. The extent the pool surface is pushed upward also depends on the interaction between the surface tension from the liquid, solid, and gas since these phases coexist at the pool edge. The conservation of mass requires that the pool surface be depressed near the center if it is raised near the edge. This makes the pool surface concave. More recently, Sim and Kim (Ref. 15) have also shown by computer simulation concave surfaces of stationary laser weld pools of steel (low S and conduction mode). With dγ/dT > 0 (Fig. 8B), such as in the presence of a significant amount of a surface-active agent, the fast inward surface flow converges to the center of the surface, where it is suddenly slowed down. This causes the pres-
Fig. 14 — Temperature oscillation induced by oscillatory Marangoni flow in NaNO₃ pool heated by CO₂ laser at 6 W (Fig. 13B). A — Temperature oscillation; B — frequencies of temperature oscillation determined by fast-Fourier transformation of A.

A mechanism is proposed to explain the effect of the surface-active agent on the pool-surface deformation, pool-surface oscillation, and ripple formation through Marangoni flow, as shown in Fig. 10. As already explained previously (Fig. 8A), in the absence of a significant amount of a surface-active agent, the fast surface flow is outward toward the pool edge, the pool surface can be raised near the edge, and the pool surface can become concave (Fig. 10A). A pool surface raised significantly above the workpiece surface without any support is unstable and sensitive to disturbances. It can oscillate with the surface flow if the flow is so strong as to become oscillatory, that is, oscillatory Marangoni flow. Upon turning off the laser, the pool-surface temperature drops immediately. However, the temperature drop is greater at the center of the pool surface than at the edge. Thus, the temperature gradients along the pool surface also drop, and the outward surface flow is weakened. Consequently, the pool surface may still be concave and the pool surface may still be raised near the pool edge, but not as much as before turning off the laser (Fig. 10B). The amplitude of surface oscillation may also drop. Surface oscillation may continue briefly after the laser is turned off. Since solidification occurs at the pool edge, usually fast in spot welding, it may be disturbed by the surface oscillation near the pool edge. Consequently, clear ripples may form. The solidification rate, for instance, is about 7 mm/s based on Figs. 6A, 11F, and 11G. As mentioned previously, Rappaz et al. (Ref. 29) observed in gas-tungsten arc spot welding of Fe-15Ni-15Cr a solidification rate of 4 mm/s near the pool edge and 6 mm/s near the pool center.

In the presence of a significant amount of a surface-active agent, the fast surface flow is inward toward the pool center, and the pool surface can become convex (Fig. 10C). As mentioned previously, the pool surface can be convex near the center and depressed near the edge (Fig. 8B) but has to be convex everywhere considering volume expansion due to melting and superheating (Fig. 9). The raised surface near the pool center is unstable and can oscillate with oscillatory Marangoni flow. However, pool-surface oscillation may be weak near the pool edge even if it is significant near the pool center. When the laser is turned off, the already weak surface oscillation near the pool edge may diminish immediately. Thus, solidification at the pool edge may not be disturbed much and ripple formation may not be clear — Fig. 10D.

This mechanism was verified by observing the pool surface of 304 stainless steel immediately after turning off the laser. The low-S weld pool in Fig. 11 shows clear surface oscillation and ripple forma-
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Surface Oscillation Induced by Oscillatory Marangoni Flow

Oscillatory Marangoni flow is demonstrated in a simulated weld pool of NaNO₃ by flow visualization using the apparatus shown previously in Fig. 3. As shown in Table 2 for NaNO₃, dγ/dT = −0.056 dyne/(cm °C). Since dγ/dT < 0, the surface flow is outward toward the pool edge. The return flow below the pool surface is inward toward the pool axis, as indicated by the arrowheads in the schematic flow pattern in Fig. 3.

The return flow revealed by the He-Ne laser light sheet in a flat (horizontal) plane 0.5 mm below the bottom of the pool surface is shown in Fig. 13. At the laser power of 4 W, the inward return flow is steady and it converges to a central point — Fig. 13A. At 6 W, however, it no longer converges to a point but to a short boundary formed between two opposing streams of flow — Fig. 13B. As indicated by the two opposite arrowheads, the opposing streams and their boundary oscillate. The flow pattern repeats itself in about 1.4 s. So, the onset of flow oscillation occurs between 4 W and 6 W, and the oscillation frequency is about 0.7 Hz (= 1/1.4 s). With a further power increase to 14 W, the flow becomes faster, as reflected by the thinner flow lines — Fig. 13C. However, the mode and frequency of oscillation seem to remain essentially unchanged.

The results of temperature measurements indicate clear evidence of temperature oscillation induced by oscillatory Marangoni flow, as shown in Fig. 14. As mentioned previously, the thermocouple was lowered into the pool to the plane of the light sheet, that is, 0.5 mm below the bottom of the pool surface, after flow visualization was finished, in order not to interfere with flow visualization. The pool temperature oscillates periodically — Fig. 14A. Fast Fourier transformation of the temperature data shows that the primary oscillation frequency is about 0.8 Hz (Fig. 14B), which is close to the frequency of flow oscillation 0.7 Hz — Fig. 13B.

Surface Oscillation Induced by Oscillatory Marangoni Flow

The evidence of surface oscillation induced by oscillatory Marangoni flow was demonstrated with the help of a NaNO₃ drop heated from above by a CO₂ laser beam — Fig. 4. Surface oscillation, if any, was difficult to detect in the hemispherical drop at the pool surface upon turning off the laser. The light images of the pool surface in Fig. 11 B–J indicate pool-surface oscillation. The light image does not remain unchanged. Instead, it varies in shape and location from image to image. Thus, immediately after the laser is turned off, pool-surface oscillation can still continue briefly, for instance, for about 0.3 s (from Fig. 11B through J). The coexistence of multiple light images in a single photo of the pool surface (e.g., Fig. 11E) suggests that the pool surface oscillates at a frequency much higher than the frequency of imaging during videotaping, that is, 30 Hz. The arrowhead in Fig. 11F indicates a ripple forming at the pool edge, that is, the solidification front. This is consistent with the presence of clear ripples on the surface of the resultant weld — Fig. 6A. The ripples that form as the surface oscillation diminishes are not as clear (Fig. H–J). This is consistent with the fact that ripples become less clear from the edge to the center of the resultant weld — Fig. 6A.

On the other hand, the high-S weld pool in Fig. 12 shows the absence of clear surface oscillation and ripple formation at the pool edge during solidification. The photos in Fig. 12A and B show the pool surface immediately before and after turning off the laser, respectively. The image of the halogen light (white in color) on the pool surface becomes much smaller immediately after the laser is turned off — Fig. 11B. When the temperature gradients along the pool surface decrease immediately after the laser is turned off, two things can be expected to happen. First, the amplitude of surface oscillation decreases and this makes the light image smaller. Second, the convexity of the pool surface decreases and this makes the light image larger. The smaller light image (Fig. 12B) suggests that the oscillation amplitude decreased but the convexity did not change much. This can be considered as to further suggest that the convex surface of the high-S weld pool is mainly caused by volume expansion due to melting and superheating — Fig. 9. The absence of significant variations in the shape and location of the light image from frame to frame (Fig. 12B–J) indicates the absence of significant pool-surface oscillation during solidification. The arrowhead in Fig. 12G indicates no clear ripple forming at the pool edge during solidification. This is consistent with the absence of clear ripples on the surface of the resultant weld — Fig. 6C.

Temperature oscillation induced by oscillatory Marangoni flow has been observed in liquid bridges of high Prandtl-number materials such as silicone oils and NaNO₃ melts (e.g., Ref. 33). A liquid bridge is usually established between the bottom surface of a higher-temperature rod and the top surface of a lower-temperature rod, the two rods being vertical, coaxial, and identical in diameter. Yang and Kou (Ref. 36) have demonstrated temperature oscillation induced by oscillatory Marangoni flow in a liquid bridge of molten Sn, which is a low Prandtl-number material like other metals and semiconductors.

The Marangoni number, which has been used to represent the extent of Marangoni flow, is a dimensionless number defined as follows:

\[
\frac{\alpha}{\mu L} \left( \frac{\Delta T L}{d\gamma/dT} \right)
\]

where \(d\gamma/dT\) is the temperature coefficient of surface tension, \(\Delta T\) the temperature difference between the center and edge of the pool surface, \(L\) the characteristic length, \(\mu\) the dynamic viscosity, and \(\alpha\) the thermal diffusivity. The characteristic length \(L\) can be taken as the radius of the pool surface. As the laser power is increased, \(\Delta T\) can be expected to increase, so is \(Ma\). Thus, Marangoni flow becomes stronger, that is, increasing in velocity, and oscillatory. As mentioned previously, the similarity law of hydrodynamics suggests that similarity in Marangoni flow between two fluid systems can be expected if the Marangoni numbers are close to each other (Ref. 34). As also mentioned previously, the Marangoni number \(Ma\) for the simulated weld pool of NaNO₃ can be close to those for steel and aluminum weld pools (Ref. 7).

Though it does not affect the conclusions of the present study, it is worth mentioning that there have been two different theories about the onset of oscillatory Marangoni flow in liquids. One suggests that oscillations result from deformation of the free surface (Refs. 37, 38) while the other suggests that Marangoni flow becomes oscillatory beyond a critical value of \(Ma\) (Refs. 39, 40).
NaNO₃ pool because the concave pool surface was supported by the container wall near the pool edge — Fig. 3. Pool surface oscillation, however, can be revealed clearly if the pool surface near the pool edge is raised without anything to support it.

Figure 15 is an example of Marangoni flow in the NaNO₃ drop. The induced Marangoni flow pattern is somewhat toroidal (like a donut), with a fast surface flow from near the top of the drop (warmer) to its bottom near the edge of the graphite rod (cooler), as indicated by the arrowhead. A vortex is visible near the surface on the right hand side of the drop. A similar vortex, however, cannot be seen on the left-hand side because the flow is asymmetric and unsteady, typical of oscillatory Marangoni flow. Since the flow pattern is optically distorted by the lens effect of the drop, the details of the flow pattern are not important. What is more important is that the flow pattern clearly varies with time, that is, oscillatory Marangoni flow.

As the power of the laser beam was increased gradually (for instance, to 17.5 W), the flow became faster and oscillatory, and the drop began to shake visibly as shown in Fig. 16. Due to the shape of the drop and its small size, the oscillation frequency probably exceeded the speed of the DV recording system, judging from the blurred images of the drop.

Conclusions

Oscillatory Marangoni flow and its effects on weld pool phenomena including pool-surface deformation, pool-surface oscillation, and ripple formation, were investigated by using conduction-mode laser spot welding of 304 stainless steels. Within the experimental conditions used, the conclusions based on the results observed are as follows:

1) A significant effect of the surface-active agent, such as S in 304 stainless steels, on the pool-surface deformation, pool-surface oscillation, and ripple formation has been discovered. This effect of the surface-active agent complements that on the weld pool depth reported in the classic work of Heiple and Roper (Ref. 1).

2) A low-S (e.g., 42 ppm) pool surface can be concave and oscillating, and the resultant weld shallow and with clear ripples. A high-S (e.g., 140 ppm) pool surface, on the other hand, can be convex and much less oscillatory, and the resultant weld deeper and without clear ripples.

3) A mechanism has been proposed to explain how the surface-active agent can affect pool-surface deformation, pool-surface oscillation and ripple formation through Marangoni flow. With little surface-active agent present, as the fast outward surface flow slows down suddenly near the pool edge, the pressure, and hence, pool surface near the edge can rise and make the pool surface concave. The raised pool surface near the edge is unstable and can oscillate with oscillatory Marangoni flow. Immediately after turning off the laser, surface oscillation may still continue briefly to disturb solidification at the pool edge and cause ripple formation. In the presence of a significant amount of a surface-active agent, however, the fast inward surface flow can make the pool surface convex and the raised pool surface near the pool center can oscillate. Since solidification occurs at the pool edge instead of the pool center, it may not necessarily be disturbed significantly to cause clear ripple formation.

4) The mechanism has been verified with 304 stainless steels. A low-S laser weld pool can exhibit a concave pool surface, brief but significant pool-surface oscillation immediately after turning off the laser, and clear ripple formation during solidification. A high-S laser weld pool, on the other hand, can exhibit a convex pool surface, immediate stop of pool-surface oscillation after turning off the laser, and no clear ripple formation during solidification.

5) Oscillatory Marangoni flow has been demonstrated by flow visualization in a simulated stationary weld pool of NaNO₃. The frequency of flow oscillation matches that of the temperature oscillation it induces in the pool.

6) Pool-surface oscillation (shaking) induced by oscillatory Marangoni flow has been demonstrated with a NaNO₃ drop laser-heated from above.

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References


Ductile-Fracture Resistance in X100 Pipeline Welds Measured with CTOA

Use of the crack tip opening angle test for determining resistance to ductile fracture of high-strength welded pipeline materials was evaluated.

BY E. DREXLER, P. DARCIS, C. N. MCCOWAN, J. W. SOWARDS, D. MCCOLSKKEY, AND T. A. SIWEERT

ABSTRACT

A test for evaluation of resistance to ductile fracture [crack tip opening angle (CTOA)] was found to reveal changes in crack extension through the heat-affected zone (HAZ) and weld interface in X100 pipeline steels. This test provides a long ligament for crack extension data not available in Charpy or drop-weight tear specimens, and is much less expensive than the full-scale burst tests. The ductile-fracture resistance of girth welds, perpendicular to the growing crack, and seam welds and their HAZs, parallel with the crack, were evaluated. Analysis of the data reveals some general differences, such as changes in CTOA and crack extension rate as the crack moved through the base metal, HAZ, and girth weld material. The values for CTOA were observed to increase and the crack extension rate decreased as the crack moved through the weld and approached the weld interface. The plastic deformation appears to be strongly influenced by the properties and geometry of the narrow HAZ, the weld interface, and the tougher base material. Consequently, the CTOA of the HAZ associated with the girth weld was larger than that of the seam-weld HAZ. It was not possible to obtain CTOA data for the seam weld with the crack parallel within the weld, because the crack immediately diverted out of the weld material into the HAZ. The CTOA values from both girth welds and seam-weld HAZ were smaller than those of the base material.

Introduction

Crack tip opening angle (CTOA) is a crack ductility test that is widely accepted for assessing the likelihood of steady-state tearing behavior in the aluminum alloys found in older aircraft. Recently, it has gained acceptance in the pipeline community as a fracture parameter for pipeline design (Refs. 1, 2). The modified double cantilever beam (MDCB) specimen design advocated by Hashemi et al. (Refs. 3, 4) and Shterenlikht et al. (Ref. 5) was adopted for this work. Advantages of this specimen design include a long ligament arm (200 mm) for steady-state tearing and higher constraints, approaching those seen in pipeline material in service (Ref. 4). This design has been used successfully to generate values for the resistance to crack extension for pipeline base metals, such as X52, X80, and X100 (Refs. 6, 5, 3, respectively).

Welding of X100 base metal has many simultaneous challenges. Beyond the difficulty of obtaining a weld that is compatible with the base material under static loads, there are the complexities of understanding the influence of the weld should a running crack initiate and then propagate due to the combination of loads from service pressure, other factors such as third-party damage and any ground movement, and residual stresses introduced during the joining process (Refs. 7, 8).

Procedure

Materials

Weld material and the associated HAZ from X100 experimental pipelines were tested with MDCB specimens to obtain CTOA data. The certified composition for X100 pipeline is found in Table 1. The sections had a diameter of 1.32 m (52 in.) and were 20.6 mm (0.81 in.) thick. The pipe was received already welded. Girth welds were made manually with shielded metal arc welding (SMAW), and seam welds were produced by automatic submerged arc welding (SAW) with materials and procedures representative of future field production. More details on the se-