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SECONDARY EMISSIONS FROM UNDERWATER PLASMA-ARC CUTTING OF STAINLESS STEEL 
FOR NUCLEAR DECOMMISSIONING

B. WALDIE, G. PILOT*, W.K. HARRIS and H. LOYER*

Dept. Chemical and Process Engineering, Heriot-Watt University, 
Edinburgh EH14 4AS, Scotland
*C.E.A., SPIN Group, CEN Saclay, 91191 Gif-sur-Yvette, France

RESUME

Dans le cadre d’un programme interlaboratoires sur l’étude des émissions secondaires produites par la découpes par torche à arc plasma, les émissions solides et gazeuses dues à la découpes sous eau de plaques en acier inoxydable à l’aide d’une torche à arc plasma ont été analysées en détail. Des échantillons en acier inoxydable 304 d’épaisseur 20 et 40 mm ont été découpés sous des profondeurs d’eau de 0,5 et 1 m en utilisant un mélange gazeux argon/azote. Les essais se sont révélés reproductibles, les aérosols sont enrichis significativement en Mn et à un degré moindre en Cr et légèrement apparu en Ni et Co. Les concentrations et les distributions granulométriques de ces aérosols ont été mesurées. Les fines particules collectées dans l’eau sont partiellement dissoutes, ceci est due a l’acidité engendrée par l’absorption d’oxydes d’azote dans l’eau. La mesure des émissions de NO et H₂ a également été effectuée.

ABSTRACT

Detailed properties and yields of gaseous and particulate emissions from underwater plasma-arc cutting of stainless steel have been measured as part of an interlaboratory programme to assess the repeatability of the cutting process and secondary emissions. 20 mm and 40 mm thick specimens of 304 stainless steel were cut with argon/nitrogen gas at water depths of 0.5 and 1.0 m. Good repeatability was obtained. Aerosol particles evolved were significantly enriched in Mn and to a lesser extent in Cr but reduced in Ni and Co. Size distributions and concentrations of these particles were measured. Fine particles collected in the water were shown to be partially dissolved by acidity caused by absorption of nitrogen oxides in the water. Emissions of NO and H₂ were also quantified.

1. INTRODUCTION

Underwater plasma arc cutting is a valuable tool for repair and decommisioning of nuclear installations. Water surrounding the workplace and torch can absorb radiation and help collect secondary particulate emissions. Reliable prediction of the characteristics and yields of particulate and gaseous emissions is important for acceptability of the technique where active materials are involved.

The present study is part of an interlaboratory programme aimed at assessing the repeatability of cutting performance and characteristics and yields of secondary emissions. Data on characteristics and yields from individual cuts at water depths up to 9.5 metres were presented previously (1,2). In the present experiments, water depths were restricted to 0.5 and 1.0 m and repeat runs done over longer cut lengths. As before, non-active stainless steel was cut. Previous data (1,2) showed that aérosols were significantly enriched in some constituents of the stainless steel. One of the objectives here was to establish whether there was enrichment in cobalt which from an active workplace material would be particularly undesirable.

2. EXPERIMENTAL TECHNIQUES

Cutting, in the normal gravity position, was done in a 0.6 m diameter column with observation ports. The workplace was moved longitudinally and the torch laterally at a controlled speed, 60 mm/min, to achieve a cut length of 0.9 m per run. 50 l/min of 60% argon/40% nitrogen was used with a 2.5 mm diameter nozzle. Workpieces, of 304 stainless steel, were 20 mm or 40 mm thick. Current was 200 and 250 amps respectively. Cutting was done in 0.5 m and 1 m of deionised and filtered water, a fresh charge being used for each
Fine particles suspended in the water after a cut were measured by filtration of 25 and 50 l samples through 0.45 μm membrane filters. Coarser sedimented particles were collected in a tray and by careful cleaning of the vessel base after a run.

The gas sampling rig (3) was connected to the column as shown diagrammatically in Figure 1. Total effluent gas, comprising plasma gas leaving the water plus filtered air diluent, passed first through a 125 mm diameter stainless steel pipe. Sample positions for specific instruments or filters were located along the pipe. Other items incorporated included an orifice plate to measure the flowrate and a fan with a flow control valve. Various sizes of sample nozzle were available to achieve isokinetic sampling inside the pipe. Controlled flow of total effluent gas, comprising plasma effluent gas plus filtered air diluent, was 80 m³h⁻¹.

Gas analysis was by a range of commercial instruments utilizing thermal conductivity (H₂), u.v. absorption (O₃) and chemi-luminescence (NO, NO₂). Aerosol particle size distributions were measured by a combination (4) of inertial impactor and diffusional spectrometer (S.D.I. 2001). Elemental metal contents of fine suspended particles and aerosol particles were determined by X-ray fluorescence or I.C.P.

3. RESULTS AND DISCUSSION

Cut quality was very consistent in the repeat runs as indicated in Figure 2. Mean total yields and distributions of collected solid emissions for the three sets of conditions (Table 1) show, as before (1, 2), that sedimented dross is the major constituent, over 90% mass. Suspended particles and aerosol particles, which account for much smaller proportions, can however pose greater collection problems. For all except attached slag, the scatter on repeat runs at the same condition is within 10% of the mean values in Table 1. Sedimented dross particles are generally smaller from the 20 mm steel (Figure 3), due presumably to the shorter liquid metal flow path before detachment.

The 25% reduction in aerosol yield obtained by increasing the water depth from 0.5 to 1 m agrees with the trend found previously (1) at other cutting conditions. Size distributions of aerosol particles were similar for the three sets of conditions with mass mean sizes in the range 0.21 to 0.24 microns. The form of the distribution for 20 mm at 0.5 m is shown in Figure 4. The above mean values are based on the distribution calculated from chemical analysis of Fe on the collecting elements in the particle size analyzer. Measurement by weighing of total masses gives an apparent bimodal distribution (Figure 4) but that is considered less reliable due to the very small total masses on some of the collecting elements.

Elemental analysis of aerosol particles shows some interesting features (Table 2). There is significant enrichment in manganese and to a lesser extent in chromium, as compared to the parent stainless steel. Manganese is enhanced by a factor of about 12 times or more and chromium by about 2. This enhancement in manganese confirms earlier results (1) where factors over 10 were reported. There was slight evidence of chromium enrichment previously (2) but the present results are more definite and repeatable. Cobalt concentrations are actually lower in the aerosol than in the parent steel. The experimental enrichment factors in Table 2 are in the same relative order as the vapour pressures of the pure elements at a given assumed melt temperature, 2200°K. Thus manganese has the highest vapour pressure and cobalt the lowest of the four elements considered. There is no uniform factor giving a simple quantitative relationship between relative enhancement and relative vapour pressure but this is hardly surprising considering the complexity of the physical and chemical factors involved. In all cases however, the enrichment factor varies with relative volatility to a power less than one, as might be expected if other factors such as diffusion were also involved. No attempt is made to compare copper concentrations as some copper could come from the torch nozzle. Present results differ from those of Hagashi et al. (5) who reported an aerosol composition similar to that of the stainless steel but did not determine the manganese content.

Another feature of the aerosol particle composition is the high ratio of total mass to total elemental metal content, an average of about 1.9 (Table 2). The ratio for the fine water suspended particles is also high, about 1.7 average (Table 3). For Fe, the major elemental constituent, formation of Fe₂O₃ or Fe₃O₄ would give ratios of 1.43 or 1.38 respectively. Nitriding is also unlikely to give such high ratios. Hydrated magnetite, Fe₂O₃.4H₂O has a ratio of 1.81. The exact composition of the aerosol and fine water suspended particles is therefore not resolved but they appear not to be simple oxides and/or nitrides.
Regarding gases produced in the process, \( H_2 \) and NO are the principal effluent components of concern. All concentrations quoted apply to the stream after dilution with 80 m\(^3\)h\(^{-1}\) air. For 20 mm thick steel, the \( H_2 \) concentration in total effluent gas (Figure 5) is about 4,200 p.p.m. and reasonably steady and reproducible. For 40 mm steel, concentrations are somewhat higher and fluctuate during a cut (Figure 6). The concentrations and fluctuating nature were similar in 0.5 m and 1.0 m water. In an earlier study of gas evolution (6) reaction between molten steel and water was suggested as a major mechanism. It is possible that the fluctuations in hydrogen are associated with variations in the length of streamers of molten steel attached to the back of the workplace when cutting 40 mm material. Predominance of such a metal/water reaction also helps explain the relative absence of oxygen which might be expected if dissociation or more especially electrolysis were major contributors (6). When account is taken of the different electrical power levels, 26.4 and 35 kW respectively, the hydrogen evaluation yields per kW are essentially the same 0.223 and 0.226 l.min\(^{-1}\).kW\(^{-1}\) for the two thicknesses of steel at 0.5 m depth. Electrical power was found previously (6) to be the best correlating parameter for hydrogen evolution at constant speed. Yields per kW are about half of those at a lower cutting speed (2). There is no variation with water depth over the range studied.

NO concentrations are repeatable and steady with time for both 20 and 40 mm thick steel. There is some evidence (Figure 7) of a slight reduction with depth, supporting the trend of increased absorption of oxides of nitrogen with depth reported (1) for depths to 9.5 m. NO concentrations are again low, about 18 p.p.m. after dilution. The water becomes acidic during cutting due to absorption of oxides of nitrogen. Here the pH decreased more than before (1,2), to 3.2 in some runs, because of the longer run times. This acidity resulted in an average of 86% of the metal constituents in the fine suspended particles being dissolved after a period of 13 weeks. Such dissolution would complicate cleaning of the water.

Ozone concentration in the total effluent after dilution reached a maximum of 55 p.p.b. and tended to decrease with time after the first 2-3 minutes of a run to zero within 10 minutes.

CONCLUSIONS

Good repeatability was obtained in cut quality and secondary emission measurements. No significant effects of water depth were found over the range 0.5 to 1 m except for a reduction in aerosol emissions by a factor of 1.3. The submicron aerosol particles were enriched or depleted in metallic elements according to the relative order of the volatilities of those elements. Thus the aerosol particles were depleted in cobalt as compared to the parent steel. Fine particles left suspended in the water were substantially dissolved after 13 weeks due to acidity from by-product oxides of nitrogen. \( H_2 \) and NO were the main gaseous effluents, the former fluctuating in cuts in 40 mm steel.

ACKNOWLEDGEMENTS

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REFERENCES

Table 1 Yield and distribution of solid emissions

<table>
<thead>
<tr>
<th>Thickness (mm)</th>
<th>Water depth (m)</th>
<th>Total collected (g/m²)</th>
<th>Attached slag (%)</th>
<th>Sedimented dross (%)</th>
<th>Suspended particles (%)</th>
<th>Aerosol particles (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>0.5</td>
<td>810</td>
<td>8.52</td>
<td>90.99</td>
<td>0.477</td>
<td>0.014</td>
</tr>
<tr>
<td>40</td>
<td>0.5</td>
<td>1957</td>
<td>3.74</td>
<td>95.97</td>
<td>0.277</td>
<td>0.008</td>
</tr>
<tr>
<td>40</td>
<td>1.0</td>
<td>1990</td>
<td>2.62</td>
<td>97.11</td>
<td>0.270</td>
<td>0.006</td>
</tr>
</tbody>
</table>

Table 2 Ratios of elemental metal contents of aerosol particles and parent stainless steel

<table>
<thead>
<tr>
<th>40 mm 304 s.s.</th>
<th>Aerosols</th>
<th>Mean Enrichment Ratio</th>
<th>rel.* V.P.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr/Fe</td>
<td>0.243</td>
<td>0.485</td>
<td>0.503</td>
</tr>
<tr>
<td>Ni/Fe</td>
<td>0.137</td>
<td>0.137</td>
<td>0.134</td>
</tr>
<tr>
<td>Mn/Fe</td>
<td>0.024</td>
<td>0.320</td>
<td>0.314</td>
</tr>
<tr>
<td>Co/Fe</td>
<td>0.0039</td>
<td>0.0023</td>
<td>0.0026</td>
</tr>
<tr>
<td>Cu/Fe</td>
<td>0.025</td>
<td>0.020</td>
<td>0.020</td>
</tr>
<tr>
<td>Total mass</td>
<td>1.99</td>
<td>1.88</td>
<td>2.00</td>
</tr>
</tbody>
</table>

* For pure element relative to that of Fe at a nominal 2,200°C.

Table 3 Composition ratios for fine water suspended particles

<table>
<thead>
<tr>
<th>20 mm/0.5 m</th>
<th>40 mm/0.5m</th>
<th>40 mm/1 m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total mass</td>
<td>1.72</td>
<td>1.73</td>
</tr>
</tbody>
</table>

| Fe+Ni+Cr+Mn+Co+Cu | 1.72 | 1.73 | 1.64 |
Figure 1 Schematic of experimental rig

Figure 2 Front and rear views of cuts
Figure 3  Size distribution of sedimented dross

Figure 4  Size distribution of aerosol particles, 20 mm/ 0.5 m water
Figure 5  \( \text{H}_2 \) concentration (p.p.m.) in total effluent, 20 mm, 0.5 m

Figure 6  \( \text{H}_2 \) concentration (p.p.m.) in total effluent, 40 mm, 0.5 m

Figure 7  NO concentration (p.p.m.) in total effluent, 40 mm, 0.5 m and 1.0 m