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Alternatives for Joining Stainless Steel to Reduce Cr(VI) Emissions and Occupational Exposures

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Welding stainless steel generates hazardous air pollutants (HAPs). Two technologies were developed to reduce these HAPs. The first, tetramethylsilane is added to the welding shielding gas, and welding proceeds as normal. Reactive oxygen species are consumed, and the resulting metal fume particles are encapsulated. In the second technique, new chromium-free welding consumables are introduced nearly eliminating Cr(VI) in the welding fume.

Keywords: welding; safety

1. Introduction

Welding operations at the Department of Defense (DOD) maintenance facilities generate fume containing metallic hazardous air pollutants (HAPs), such as cobalt, chromium (trivalent and hexavalent Cr(VI)), manganese (Mn), nickel (Ni), lead (Pb), copper (Cu), and others. The intense energy of the welding process results in the formation of fumes containing a high number concentration of submicron particles as well as a number of gaseous species including ozone (Jenkins et al. 2005, Hewett 1995). A variety of parameters, including filler metals composition, shielding gas composition, and metal transfer mode, all influence the fume characteristics (Zimmer et al. 2002). Mild steel welding fumes generally consist of iron and manganese oxides (Minni et al. 1984, Jenkins and Eagar 2005). Stainless steel welding fumes also contain manganese and iron; additionally, they contain chromium and nickel oxides (Castner & Null 1998, Heung et al. 2007). Chromium found in the fumes is often in the hazardous hexavalent state (Heung et al. 2007).

A number of metallic species present in welding fumes are potentially detrimental to worker health and ambient air quality. Mild and stainless steel welding emit manganese fumes and can cause a number of adverse health effects. Many epidemiologic studies have shown that manganese exposure can lead to a variety of neurologic problems, including a Parkinson's-like disorder known as manganism (Yuan et al. 2006, Antonini et al. 2006, Bowler et al. 2006, Halatek et al. 2005, Bowler et al. 2007). Exposure to hexavalent chromium (Cr(VI)) causes decreased lung function, asthma, and cancer (Bagchi et al. 2002, Pascal & Tessier 2004). Studies of welding emissions in California found that welding is the primary source of airborne Cr(VI) in the state (Chang et al. 2004).

Recent Occupational Safety and Health Administration (OSHA) regulations lowered the permissible exposure limit (PEL) for Cr(VI) from 52 to 5 $\mu g/m^3$ as an 8 hour time-weighted average (TWA) (OSHA 2006). The US Environmental Protection Agency's Residual Risk program has proposed an expansion of the regulation to include shipbuilding operations (EPA 1990). After the promulgation of the OSHA regulations, many facilities installed industrial ventilation systems, thus consolidating the fume emissions into a point source.

The objective of this paper is to introduce two developing technologies that reduce hazardous air pollutants (HAPs) generated during stainless steel (rich in Cr content) welding. The University of California-Davis scientists initiated the first technology, and University of Florida (UF) researchers further refined it. In this technology, tetramethylsilane (TMS) is added to the gas metal arc welding (GMAW) shielding gas, and welding proceeds as normal. The process consumes reactive oxygen species and encapsulates the resulting metal fume particles, preventing the metal's contact with lung and stomach tissue. In laboratory testing, more than 90% reduction of Cr(VI) was accomplished when 4% TMS was present. The Ohio State University (OSU) researchers developed

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a second technology. The chromium-free (Cr-free) consumables are introduced for shield metal arc welding (SMAW) and (GMAW). Preliminary metal quality work is complete. These consumables designed for stainless steel do not contain chromium; instead, they are made of nickel, copper, ruthenium, and other metals.

Both university teams completed a year of optimization and the project team (Navy engineers, UF and OSU researchers) is ready to conduct on-site demonstrations.

2. New technologies

2.1. Technology 1: Tetramethylsilane (TMS) addition technology

2.1.1. Technology description. Vapor phase sorbents are an emerging technology for the control of metal emissions from combustion sources. Numerous studies have tested the use of silica precursors for control of metal emissions in generic combustion systems. Silica, formed from oxidation of precursor chemicals, has proven effective as a sorbent for lead produced during combustion (Biswas & Zachariah 1997, Owens & Biswas 1996a, 1996b, Lee et al. 2005). Silica is capable of forming an amorphous web that effectively captures ultrafine metal aerosols and increases their effective particle size (Biswas & Zacharaih 1997, Owens & Biswas 1996a, 1996b, Lee et al. 2005, Dennis et al. 2002). The use of silica precursors to increase aggregate particle size can be used to improve the performance of filtration systems.

During combustion, the precursor molecule is broken apart as a result of the intense energy imparted by the welding arc. This process leads to the formation of silica (SiO_2) along with carbon dioxide and water, as shown in Fig. 1. Oxygen that is able to penetrate the shielding gas is consumed during the pyrolysis process, thereby reducing the concentration of reactive oxygen species (O, O_3) that lead to the formation of Cr(VI). The reaction shown below is the stoichiometric pyrolysis process that leads to the formation of SiO_2.

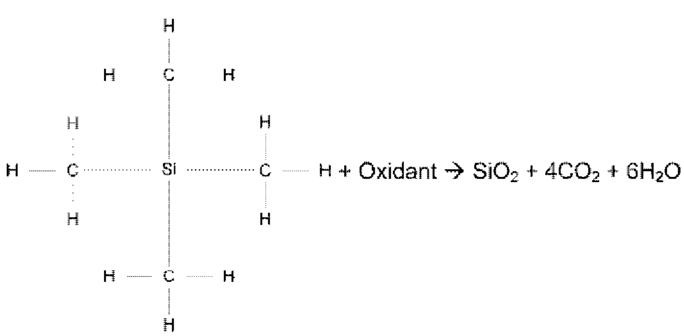


Fig. 1 Stoichiometric pyrolysis of TMS

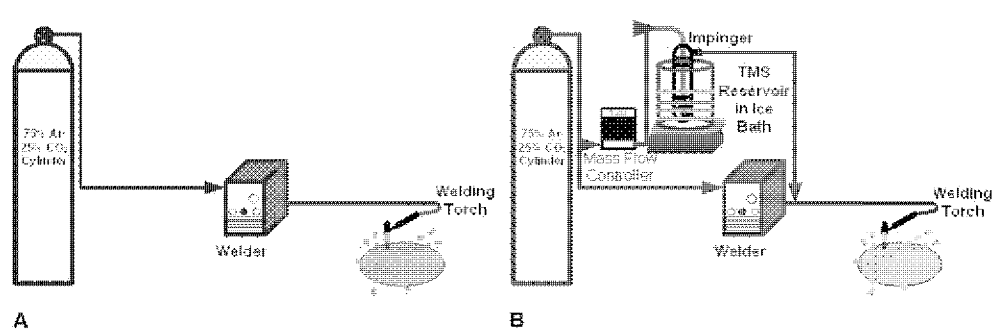
The precursor is delivered as a vapor to the weld site. This requires a carrier gas to transport the precursor vapor from a liquid reservoir to the active weld site. The Ar/CO_2 mixture already employed as shielding gas in GMAW processes can easily be used to fulfill this need. The welding system with precursor addition is compared with traditional GMAW welding in Fig. 2.

The concept of using silica precursors as welding shielding gas additives was originally developed by Dr. Daniel P. Chang, University of California, Davis. Dr. Chang conducted preliminary experiments in 2004 to test whether TMS could be used as an effective shielding gas additive for Cr(VI) control. In the limited testing he performed, Dr. Chang found that adding a small amount of TMS to GMAW shielding gas reduced Cr(VI) formation by about 50%. In early 2008, a study at UF found that adding a small amount of tetraethylorthosilicate (TEOS) to gas tungsten arc welding (GTAW) shielding gas reduced Cr(VI) by about 45% (Topham et al. 2010).

2.1.2. Technology development. The results shown are the product of a UF study of tetraethylorthosilicate (TEOS) as a gas tungsten arc welding (GTAW) shielding gas additive. TEOS is a silica precursor compound similar to TMS. The TEOS molecule contains some oxygen, which may slightly hinder its effectiveness as a reducing agent. Additionally, the vapor pressure of TMS is much higher than TEOS, which allows it to be used without requiring heating.

Changes in Cr(VI) formation were analyzed by comparing Cr(VI)/Cr during TEOS sampling and during baseline sampling. The average Cr(VI)/Cr ratio decreased by 45% from the average baseline Cr(VI)/Cr ratio (from 0.170 to 0.094). The nitrate concentrations at the baseline and TEOS sampling conditions were also analyzed. Nitrate is the end product of oxidation of atmospheric nitrogen, which requires oxygen species to form. Therefore, it can be used as an indicator of the formation of reactive oxygen species during the welding process (Dennis et al. 2002).

Upon the addition of TEOS, nitrate concentration decreased from an average baseline value of 83 to 39 ppm. This 53% decrease from baseline conditions is in line with the Cr(VI) reduction. This result demonstrates an oxidation potential of chromium within the welding fume. Previous studies used other reducing agents as shielding gas additives. For example, one study (Dennis et al. 2002) used 3% ethene and 3% nitrogen oxide and obtained



A B

Fig. 2 *A* Traditional GMAW welding system and *B* GMAW welding system modified to allow TMS addition

42% and 45% reduction in Cr(VI) formation, respectively, slightly lower than that achieved in this study.

Metal vapors formed during welding quickly condense and form primary particles a few nanometers in diameter. These nanometer primary particles at very high number concentration undergo rapid coagulation to form aggregates. The metallic aerosol is coated in a thin SiO_2 film. Silica is significantly less dense than the metal aerosols and produces a film around the metallic aerosols. The primary and aggregate metal particles formed during tungsten inert gas (TIG) welding were covered in a film of amorphous SiO_2 (Topham et al. 2010).

2.1.3. Advantages and limitations of the technology. This technology is superior to current shielding gas technology in that it addresses all of the hazardous metals produced during stainless steel welding. Nickel, manganese, and Cr^{6+} are all present within the welding fume particles. Thus, it is necessary for new technologies to address the effect of all three of these potentially harmful metal emissions. The silica coating that encapsulates the metal aerosols can be an effective means for masking the hazardous properties of all of the metals contained in the silica particles. The twofold approach of limiting oxidation potential and coating aerosols in a silica layer goes beyond previous control technologies by addressing all pollutants in the metals regardless of oxidation state. Tests using *E. coli* have been carried out, and the results show significantly reduced biotoxicity for welding fume with TMS addition (Yu et al. 2010).

The optimal operating conditions have not been determined. The amount of TMS added to achieve the performance objective while minimizing excess TMS injection is being determined in the lab. The mechanical properties of the welded metal must be analyzed to determine whether the addition of a small amount of silica to the joint will compromise the integrity of the weld.

Before this technology can be applied on a large scale there are challenges to overcome. TMS is currently not widely available as a gas. Therefore, TMS vapor is currently generated in situ from high-purity, laboratory-grade TMS liquid. If an industrial grade TMS feed becomes available, it will result in significant cost savings. Additionally, TMS in vapor form is not readily available. A source of TMS vapor in premixed gas cylinders would be an ideal solution to address this current limitation. Custom order of such a cylinder is commercially available, and its price can be reduced drastically once the TMS process is widely adopted by the industry. TMS is highly flammable, and therefore it must be handled carefully to prevent accidents in workplace environments where welding is consistently producing sparks capable of igniting the liquid. TMS is also a respiratory irritant that can cause burns in the throat and respiratory system if acutely inhaled.

2.1.4. Performance objectives. The objective of the laboratory phase of the TMS demonstration was to achieve 90% reduction in Cr(VI). This 90% reduction was measured compared with traditional GMAW welding. The performance objectives of the laboratory phase demonstration with the data requirements and the criteria for success are summarized in Table 1.

2.2. Technology 2: Cr-free consumables (coated electrode and filler wire)

2.2.1. Technology description. A consumable with nominal composition Ni-7.5Cu-1Ru has been developed as a replacement for conventional stainless steel consumables such as Types 308, 309, and 316 for welding austenitic stainless steel base metal. The new consumable appears to have comparable corrosion resistance and mechanical properties relative to the consumables it is designed to replace. The measured Cr(VI) in the fume of this electrode when welding Type 304 stainless steel is virtually nil (0.02 wt%) and represents a 100-fold reduction in Cr(VI) relative to a conventional Type 308 consumable. Use of this electrode will allow the new OSHA permissible exposure limit (PEL) for Cr (VI) to be routinely met in shop and field welding of austenitic stainless steels.

The main application of the Cr-free welding consumable is for welding stainless steel in confined spaces where providing efficient ventilation is impossible and/or is not feasible and the OSHA PEL of 5 $\mu g/m^3$ 8-hour TWA cannot be met by the standard stainless steel welding consumables.

2.2.2. Technology development. The main objectives in the Cr-free consumable development was to achieve elimination of the carcinogenic Cr(VI) in the welding fume during stainless steel welding and to provide a compatible replacement of the standard stainless steel welding consumables in terms of weld corrosion

Table 1 Performance objectives of laboratory study

Performance Objective	Data Requirements	Success Criteria
Quantitative performance objectives		
Reduction of Cr(VI) generated during GMAW	Laboratory demonstration of effectiveness of TMS at reducing Cr(VI) concentration generated compared with baseline GMAW	>90% reduction in Cr(VI) generation from current GMAW process
Qualitative Performance Objectives		
Verification of SiO_2 coating on welding fume particles	TEM imagery and SMPS particle size data for traditional GMAW versus GMAW with TMS added	Visual evidence of SiO_2 coating on fume particles via TEM Increase in aggregate particle size demonstrated with SMPS
Verify that the TMS additive does not reduce the quality of the weld	AWS standards used to conduct nondestructive and destructive welding tests. Laboratory samples testing will be evaluated at NSWC Carderock Division	No negative difference in welding and material quality between traditional and TMS GMAW samples.

AWS, American Welding Society; GMAW, gas metal arc welding; NSWC, Naval Surface War Center, SMPS, scanning mobility particle sizer; TEM, transmission electron microscopy; TMS, tetramethylsilane resistance, mechanical properties, consumable welding operability, and weldability. To achieve these objectives, design criteria were imposed:

- The breakdown and repassivation potentials of the weld metal should be higher than the corrosion potential of the stainless steel substrate to prevent localized attack of the weld metal.
- If possible, the corrosion potential of the weld metal should be slightly higher than that of the solid solution substrate so that the weld metal is cathodically protected.
- The strength and ductility of the welds must meet or exceed minimum requirement for the base metals they join.
- Weldability, including susceptibility to various forms of cracking during welding, should be within the range of comparable consumables.
- The operating characteristics of the consumable should be such that it can be readily used in applications requiring manual, semiautomatic, and fully automated welding processes.

Four generations of Cr-free consumables were developed to meet the aforementioned design requirements:

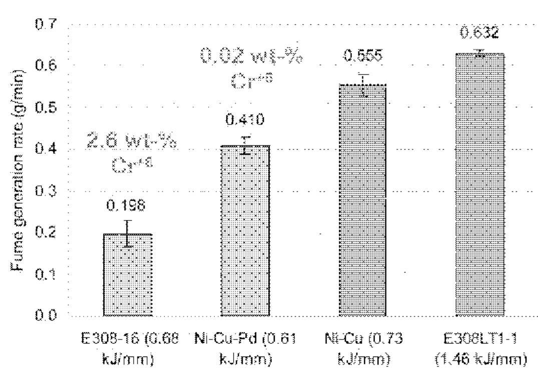
- **Generation I.** Nominal Ni-8.0Cu-0.2Pd bare wire consumable that was designed based on the results of corrosion tests on small button melts.
- **Generation II.** Special Metals Welding Products Company produced nominal Ni-7.5Cu and Ni-7.5Cu-1Pd coated electrode. The Cu and Pd were added to the coating rather than the core wire. The transfer of substantial Pd across the arc was difficult with these electrodes.
- **Generation III.** Nominal Ni-7.5Cu-1Ru-0.5Ti bare wire that was melted by Haynes International. Ru replaced Pd as a lower-cost alternative. Attempts to use this composition as a core wire for coated electrodes were unsuccessful as a result of porosity and operability problems. This wire worked very well for GTAW and GMAW applications.
- **Generation IV.** Nominal Ni-7.5Cu-4Ti-1Ru composition that was developed as a core wire for the coated electrodes in Shield metal arc welding (SMAW). The higher Ti relative to Generation III effectively eliminated the porosity and operability problems.

Thus, the final target weld metal composition that meets the design requirements for strength and corrosion resistance is nominally Ni-7.5Cu-1Ru-0.5Ti. As noted previously, this composition is achieved in the coated electrode by overalloying the core wire with Ti; the core wire has 4%Ti whereas the deposited metal has only 0.5%Ti as most of the Ti is lost in the arc.

Figure 3 provides a comparison of the fume characteristics in Ni-Cu, Ni-Cu-Pd, and E308-16 SMAW electrodes and in a flux cored E308LT1-1 electrode. The Cr-free electrodes had higher fume generation rate (FGR) than E308-16, but the content of Cr (VI) in the Ni-Cu-Pd fume was more than 2 orders of magnitude lower than in E308-16. Based on these measurements, the Cr (VI) generation rate of the E308-16 was calculated to be approximately 60 times higher than that of the Ni-Cu-Pd consumable for similar welding conditions.

Figure 4 shows an example of the magnitude of Cr(VI) reduction in the Cr-free consumable compared with E308-16 electrodes. The example assumes that no ventilation is used and that the fume is dispersed uniformly throughout the enclosed space. Using the Cr(VI) PEL value of 5 $\mu g/m^3$, a welder exposed to fume of E308-16 would be within the PEL as long they were in a room of approximately $12.5 \times 12.5 \times 3$ meters. By switching to the chromium-free consumable and making a similar weld, the allowable size of the room is decreased to $2.3 \times 2.3 \times 3$ meters. Clearly this will allow welding-related personnel to be within exposure limits during fabrication and production situations within enclosed spaces by using a chromium-free consumable. The mechanical properties of the Cr-free consumable exceeded the minimum values strength, percent elongation, and reduction in area for Type 304L stainless steel base metal and those specified by AWS A5.4 for Type E308L, E309L, E316, E316H, and E316L weld metal (Table 2 and Fig. 5).

2.2.3. Advantages and limitations of the technology. The new Cr-free welding consumable produces welds with mechanical properties that fulfill the requirements for Type 304 stainless steel and exceed the specification of AWS A5.4 for typical Type E308L, E309L, E316, E316H, and E316L electrodes for welding of stainless steel. This new consumable has welding operability, weldability, and fume generation rate that are similar to the stainless steel electrodes.



Average Welding Fume Composition**

Element	Ni-Cu (Wt-%)	E308-16 (Wt-%)
F	17.5	-
Na	38.8	0.8
Mg	0.2	0.0
Si	3.0	8.7
Cl	0.3	1.0
K	2.3	45.8
Ca	5.0	3.1
Ti	5.1	5.1
Cr	0.5	9.7
Mn	0.6	7.0
Fe	1.1	9.0
Ni	15.6	-
Cu	9.9	-

**Note that Oxygen was also present in fume from both electrodes.

Fig. 3 Comparison of the fume generation rates, fume Cr(VI) content, and bulk fume composition of Cr-free and standard stainless steel SMAW electrodes

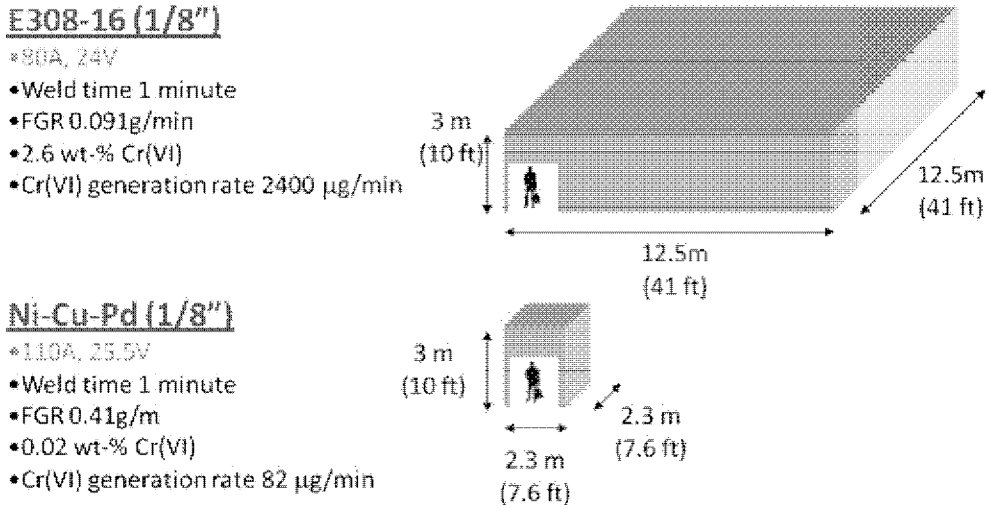


Fig. 4 Comparison of Cr(VI) generation characteristics of E308-16 and Generation II of the Cr-free welding consumable for 1 minute welding in an enclosed space

Table 2 Mechanical properties of Ni-Cu, Ni-Cu-Pd and Ni-Cu-Ru weld metals

Weld Metal	Base Metal	Failure Location	0.2% Proof Stress, MPa	Tensile Strength, MPa	Elongation, %	Reduction in Area, %
Ni-Cu	304L	Weld metal	307	597	33.2	43.0
Ni-Cu-Pd	304L	Weld metal	263	531	31.7	52.9
Ni-Cu-Ru	304L	Weld metal	279	540	52.0	54.0
Minimum Values			0.2% Proof Stress, MPa	Tensile Strength, MPa	Elongation, %	Reduction in Area, %
304L			170	480	40	50
AWS A5.4-92 (AWS 1992): E308L, E309L, E316, E316H			–	520	35	–
AWS A5.4-92 (AWS 1992): E316L			–	490	35	–

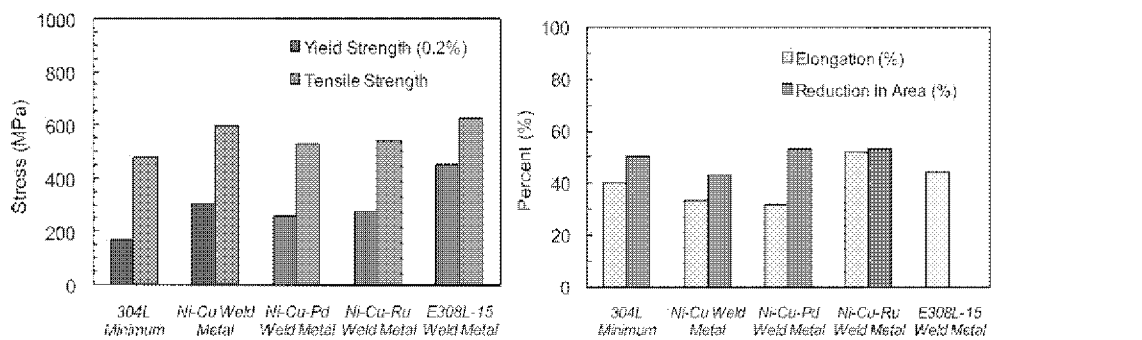


Fig. 5 Mechanical properties of the Ni-Cu, Ni-Cu-Pd, and Ni-Cu-Ru welds

The main advantage of the new Cr-free welding consumable over the Type E308 welding electrodes is that it nearly completely eliminates the carcinogenic Cr(VI) in the welding fume generated during welding of austenitic stainless steel. Use of this electrode will allow the new OSHA permissible exposure limit (PEL) for Cr(VI) to be routinely met in shop and field welding applications. No other available stainless steel consumables for welding the 300-series stainless steels meet the OSHA PEL.

A disadvantage of the new Cr-free welding consumable is its high price. The cost analysis of Generation II of this consumable that was alloyed with 1 wt% Pd had predicted an increase in the welding cost at Navy shipyard applications between 75% and 200% (ANSI/AWS A5.14/A5.14M-97). The price of palladium at $4,500/lb is the basis for this cost analysis. The palladium was substituted with ruthenium in Generation IV of this consumable, which is used in the current project.

Table 3 Performance objectives and acceptance criteria

Performance Objective	Standard, Code, or Specification	Acceptance Criterion
Electrode wire composition	ANSI/AWS A5.11-2010	
ERNiCuRu composition		Target compositions of ERNiCuRu wire wt%: C(0.1); Mn(0.5); P(0.02); S(0.02); Si(0.5); Cu(5-10); Ni(Remainder); Ru(1.5); Al(1.0); Ti(1.0); Other Elements (0.5) [Note: Single values are maximum]
Electrode dimensional characteristics	ANSI/AWS A5.4-2006 ANSI/AWS A5.11-2010	Dimensional tolerances
ERNiCuRu wire diameter		$D = 0.045 \pm 0.002$ in.
ENiCuRu rod diameter and length		$D = 0.125 \pm 0.003$ in.
ENiCuRu coating eccentricity		$L = 14 \pm 1/4$ in.
Welding operability	Qualitative comparison to E308L-15 and E308L-16	Comparable to E308L-15 and E308L-16
Arc stability, slag detachment, welders satisfaction		
Weld deposit composition	ANSI/AWS A5.11-2010	
ENiCuRu weld deposit composition		Target compositions of ENiCuRu weld deposit, wt%: C(0.1); Mn(0.5); P(0.02); S(0.02); Si(0.5); Cu(5-10); Ni(Remainder); Ru(1.5); Al(1.0); Ti(1.0); Other Elements (0.5) [Note: Single values are maximum]
ENiCuRu weld metal mechanical properties		
Ultimate tensile strength	ANSI/AWS A5.14-2009 ASTM A 666-03	Min 70 ksi
Elongation	ANSI/AWS A5.14-2009 ASTM A 666-03	Min 35%
Bend test	ANSI/AWS A5.4-2006	Max three fissures; max. length 3/32 in.
Emission of Cr(VI) in welding fume		
ENiCuRu: welding fume generation rate	ANSI/AWS F1.2-2006	Not more than 50% higher than E308L and E308LT1-1
ENiCuRu: content of Cr(VI) in the welding fume	ANSI/AWS F1.2-2006	Not exceeding 0.25 wt%
ENiCuRu: Extremely low Cr(VI) emission	OSHA 1910.1026	5 $\mu g/m^3$ 8 hour TWA
Weldability characteristics	ANSI/AWS A5.4-2006	
ENiCuRu weld radiography soundness		No cracks, incomplete fusion, and incomplete penetration No slag inclusions and rounded indication in excess of permitted

ANSI/AWS A5.4-92 (ANSI 1992); ANSI/AWS A5.14-97 (ANSI 1997a); ANSI/AWS A5.11-97 (ANSI 1997b); ANSI/AWS F1.2-2006 (ANSI 2006); ASTM A 666-03 (ASTM 2003); OSHA 1910.1026 (OSHA 2006)

Because of the relatively lower price of ruthenium (currently $1,700/lb), this substitution will significantly reduce the costs of welding operations with the new consumable.

2.2.4. Performance objectives. The performance objectives of the laboratory phase demonstration with the respective governing standards, codes, and specifications, and the acceptance criteria for success are summarized in Table 3.

3. Conclusions

For approximately 1 year, laboratory demonstrations were conducted to optimize the two technologies at both universities (UF and OSU). The next step is to demonstrate the two technologies at several Navy welding facilities. Welders at these facilities typically repair equipment and perform a wide variety of welding procedures.

A field demonstration project is planned to conduct on-site demonstrations to validate the two welding technologies during on-site operations in order to control HAPS, particularly hexavalent chromium, generated during typical welding operations. As a side benefit, occupational exposures to Cr(VI) and other metals are expected to be reduced. In addition to environmental occupational health issues, this demonstration includes a material quality evaluation. Destructive and nondestructive testing will determine if welded stainless steel plates processed with the new technologies will yield equivalent or better (mechanical properties, soundness of welded parts, microstructure, corrosion resistance) components when compared with those welded with the existing technology.

The authors considered several test protocols including simulating an American Welding Society laboratory gravimetric test method that uses a test chamber to collect the emissions as aerosols. While the chamber collects virtually all welding emissions, it does not reflect the "real world" operations. For instance, chambers are designed for flat automatic welding and not vertical and horizontal welding by a competent welder. The project's goal is to simulate actual welding industrial conditions as much as possible.

Welding maintenance operations are performed on a wide variety of DOD tactical platforms including ships, tanks, armored vehicles, and so forth throughout DOD. Since DOD depots and shipyards are the primary repair facility for many of the platforms, welding is ubiquitous and used at almost every installation. While most DOD weapons systems and platforms consist of mild or carbon steel, most facilities join stainless steel components during the course of their work projects. Some facilities work more frequently with stainless than others. This project focuses on ferrous base metals and does not address aluminum and other lightweight metals. Ferrous metals are currently the materials that are most widely used in all systems. The OSU technology specifically focuses on austenitic stainless steel operations.

Three demonstration sites are currently proposed. Tooele Army Depot joins stainless steel ducting for furnaces and other munitions destruction equipment that require a high degree of corrosion protection. Puget Sound Naval Ship Yard performs ship repair work and component repair galleys and other shipboard locations requiring a high degree of corrosion resistance containing stainless steel. US Coast Guard, Integrated Support Center (USCG ISC) Portsmouth, VA also conducts a fair amount of stainless steel welding.

Field testing consists of 3 separate weeks of environmental and industrial hygiene sampling and test plate preparation for later weld quality testing at Naval Surface Warfare Center Carderock Division. Work with Tooele is farther along, and a demonstration was anticipated in the late summer and fall of 2011. After data analysis and a Cost and Performance Report are prepared for the first site, a DOD review committee will perform a go/no-go determination. The authors will present field test data in the future.

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