

Generation and Control of Air Pollutants from Orimulsion® Combustion

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Abstract

Orimulsion® is an emulsified fuel, composed of approximately 70% Venezuelan bitumen, 30% water, and trace amounts of surfactant, and is being marketed primarily as a base-load fuel for utility boilers. Orimulsion® is now being used in power plants in five countries, and was proposed as a fuel for a plant in the U.S. In 1997, the U.S. Congress requested that the Environmental Protection Agency conduct a study to provide additional technical information regarding Orimulsion® and its potential environmental impacts. The study is being conducted by an EPA team led by the Office of Research and Development's National Risk Management Research Laboratory (NRMRL), and includes a broad review of previous work reported in the literature, visits to sites now using Orimulsion®, and a series of combustion tests conducted at NRMRL's facilities in Research Triangle Park, NC. The combustion tests measured mass emissions of carbon monoxide, oxides of nitrogen and sulfur, particulate matter, trace metals, and organic compounds generated by the combustion of two Orimulsion® formulations (one no longer produced) and a heavy fuel oil. These results were compared to emissions measured at full-scale plants and to emissions from previous tests conducted on similar equipment and fuels at NRMRL. Potential air-pollution-related issues associated with Orimulsion® combustion include elevated levels of sulfur, nickel, and vanadium, and generation of submicron particles and sulfur trioxide. These issues are similar to those associated with heavy fuel oil combustion, and can be addressed by use of appropriately designed and operated pollution control equipment.

Background

Orimulsion® is an emulsified fuel produced in Venezuela from approximately 70% bitumen (a naturally occurring heavy hydrocarbon material), 30% water, and a small amount of surfactant. Orimulsion tends to be higher in sulfur (S), nickel (Ni), and vanadium (V) content than many other fossil fuels (see Table 1), which can lead to environmental problems if the pollutants generated during combustion of the fuel are not adequately controlled. The bitumen is extracted from an area near the Orinoco River, and is mixed with water to produce a fuel that flows and burns in a manner similar to a heavy fuel oil. The name "Orimulsion" is derived from a combination of "Orinoco" and "emulsion." In 1998, the fuel's producer, Bitumenes Orinoco (Bitor), changed the formulation of Orimulsion to use a different surfactant package and to remove the magnesium compound originally added for corrosion control. The new formulation, Orimulsion 400, uses 0.13% tridecylalcohol ethoxylate and 0.03% monoethanolamine as surfactants. The original formulation was referred to simply as "Orimulsion" prior to the introduction of Orimulsion 400, but is now referred to as "Orimulsion 100" to distinguish it from the newer formulation. Bitor no longer produces Orimulsion 100.

In 1997, the U.S. Congress directed the Environmental Protection Agency (EPA) to "initiate a research activity to provide better scientific data on the qualities and characteristics of

* Orimulsion is a registered trademark of Bitumenes Orinoco, S.A.

Table 1. Elemental analyses of the three fuels tested in EPA's pilot-scale tests.

	<i>No. 6 Fuel Oil</i>	<i>Orimulsion 100</i>	<i>Orimulsion 400</i>
Carbon, %	86.45	64.20	58.12
Hydrogen, %	10.23	8.13	7.14
Nitrogen, %	0.26	0.25	0.17
Sulfur, %	2.07	3.05	2.23
Water, %	0.7	23.32	28.92
Ash, %	0.08	0.17	0.07
Oxygen, % (by difference)	0.90	0.88	3.35
Antimony, µg/g	0.78	0.57	0.35
Arsenic, µg/g	2.6	2.9	2.2
Beryllium, µg/g	<0.005	<0.005	<0.005
Cadmium, µg/g	<0.005	<0.005	<0.005
Chromium, µg/g	0.58	0.235	0.20
Copper, µg/g	0.76	<0.005	<0.005
Iron, µg/g	51	12	22
Lead, µg/g	1.8	1.9	1.4
Magnesium, µg/g	7.6	342	1
Mercury, µg/g	<0.005	<0.005	<0.005
Nickel, µg/g	47	69	59
Selenium, µg/g	0.13	2.9	0.04
Vanadium, µg/g	221	324	262
Zinc, µg/g	8.9	0.90	0.37
Energy content, Btu/lb (MJ/kg)	18,121 (42.1)	13,919 (32.4)	12,596 (29.3)

this product and the potential environmental impact of its introduction."¹ In response to this directive, EPA's Office of Research and Development conducted an investigation into the environmental impacts associated with the use of Orimulsion. NRMRL, in collaboration with other offices within EPA, developed an Orimulsion Technology Assessment Plan (OTAP) to guide the Agency's Orimulsion research.² The OTAP was developed as a three-phase program, beginning with a literature review and a series of pilot-scale tests in Phase I and continuing to a series of full-scale tests in Phases II and III, if Phase I results indicated a need for further work. Environmental assessments and toxicological tests were planned for all three phases. This paper presents results and conclusions derived from Phase I.

Literature Review

Available technical literature was reviewed to identify problems and issues believed to be most important with respect to air pollutant emissions and control, and to evaluate the levels of emissions experienced by full-scale systems using Orimulsion.³ The review examined 24 references describing air pollutant emissions at 9 full-scale sites and 3 pilot-scale facilities. Orimulsion is currently being used as the primary fuel at nine power plants in Canada, Denmark, Italy, Japan, and Lithuania, representing 3,866 MW of electric power generating capacity and approximately 7.5 million tons (6.8 million tonnes) of fuel consumption per year. To date, no plant in the U.S. has used the fuel for other than short-term tests.

The reports in the literature indicated that increasing combustion air levels were able to control carbon monoxide (CO) emissions. In general, the conventional techniques used to reduce nitrogen oxide (NO_x) emissions from oil combustion (staged combustion, reburning, selective catalytic reduction) were reported to be applicable to Orimulsion. CO and NO_x were found to be dependent upon boiler oxygen (O₂) levels and the combustion system design, similar to other fossil fuels. CO was reported to be slightly less when burning Orimulsion than when burning

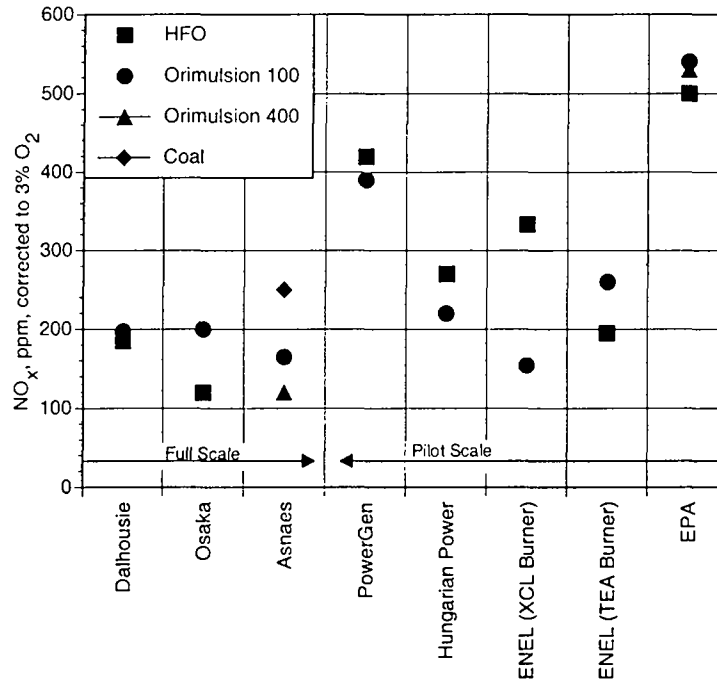


Figure 1. NO_x emissions from full- and pilot-scale facilities as reported in the literature.³

heavy fuel oil (HFO) in the same full-scale units. Figure 1 shows reported NO_x emissions from both full- and pilot-scale units burning Orimulsion and either HFO or pulverized coal. Although there is not a consistent trend for the difference between NO_x emissions from Orimulsion and HFO for the units reporting NO_x data, the reported behavior of the NO_x emissions indicates that Orimulsion behaves in a manner similar to HFO, allowing application of conventional low NO_x combustion techniques to Orimulsion.³

Sulfur dioxide (SO₂) concentrations from Orimulsion (upstream of any control) were consistent with SO₂ concentrations from other fuels with similar sulfur contents. The literature reported that conventional flue gas desulfurization systems could remove up to 95% of SO₂ generated by the combustion of Orimulsion, which would result in controlled emissions of approximately 125 ppm. Particulate matter (PM) concentrations from Orimulsion were reported to be similar to those from HFO, at between 160 and 350 mg/Nm³ (uncontrolled). PM size distributions were reported to be slightly smaller than those from HFO, with 98-100% of particle mass in sizes smaller than 10 μm in aerodynamic diameter, and 80-97% of particle mass smaller than 1 μm in aerodynamic diameter. Full-scale results demonstrated that electrostatic precipitators (ESPs) can be used to control PM emissions to a level similar to those of other fossil fuels.³

In general, Orimulsion was reported to behave similarly to a HFO. Under proper handling, Orimulsion was reported to ignite easily, generate stable flames, and be compatible with existing ignition and flame detection systems. There were no indications that conventional pollution control systems could not be successfully applied to control emissions from Orimulsion combustion, although differences such as the water content of Orimulsion needed to be accounted for in system design and operation.³

The literature does suggest, however, several areas of possible concern with regard to the combustion of Orimulsion. Particle size distributions, the levels of metals in the fuel, and the high sulfur content (and potential for high sulfur trioxide [SO₃]) are all areas that need to be well characterized before the environmental impacts associated with Orimulsion use can be better understood. Several of these issues were addressed in the pilot-scale tests conducted by EPA as part of the Orimulsion Technology Assessment Program.

EPA Pilot-Scale Test Program

The OTAP included a series of pilot-scale tests to provide the opportunity to directly compare pollutant concentrations generated by the combustion of Orimulsion 100, Orimulsion 400, and a HFO in a single closely controlled combustor. This approach would remove as many uncertainties as possible associated with changes in boiler design and operation. A series of tests were conducted at NRMRL's combustion research facilities in Research Triangle Park, NC, using a pilot-scale combustor designed to simulate the behavior of a large water-wall boiler.

NRMRL's Package Boiler Simulator (PBS) is a research combustor rated at 3×10^6 Btu/hr (879 kW), and is designed to burn either liquid or gaseous fuels. The PBS burner has an air-atomizing nozzle and was operated at heat input rates below full load to accommodate the higher volume of Orimulsion required to maintain a consistent heat input rate for all fuels. The PBS has a 10 in. (25 cm) inside diameter (ID) refractory-lined burner section connected to a water-cooled transition section of the same ID. The transition section provides for staged air or fuel injection through several ports, and connects to a Dowtherm-cooled boiler section, which has a 24 in. (61 cm) ID and is 110 in. (279 cm) long. The combustion gases pass out of the boiler section to the vertical stack, where ports are located for taking extractive samples.

Flue gases from the PBS are ducted to the facility's air pollution control system (APCS), which consists of a secondary combustion chamber, a fabric filter, and a wet acid gas scrubber. The APCS allows the PBS to operate under poor combustion conditions that intentionally generate higher-than-normal pollutant emissions during research studies without emitting excessive pollutants to the environment.

The fuel supply system can influence the stability of emulsified fuels such as Orimulsion. The system should minimize shear rates through pumps, piping, and fittings as much as possible, and should be able to maintain the appropriate temperature range during operation. The original fuel supply system used by the PBS was designed for HFO and was used during operation with the No. 6 fuel oil. For operation with Orimulsion 100 and Orimulsion 400, the fuel supply system was modified to use a Moyno[®] pump that generated substantially less shear compared to the original gear pump, and to eliminate the pressure relief valves and the continuous circulation loop used in the original supply system.

Magnesium hydroxide [$\text{Mg}(\text{OH})_2$] was injected into the boiler during testing of Orimulsion 400. This was done to simulate operations at full-scale units that also injected $\text{Mg}(\text{OH})_2$ as a means to minimize deposits on boiler tubes. The additive was injected into the flame at a rate of between 0.35 and 0.54 g/min during operation at 1×10^6 Btu/hr (293 kW) to achieve a molar ratio of between 2.1 and 3.8 mol Mg to 1 mol V in the fuel.

The PBS uses continuous emission monitors (CEMs) for measurement of combustion gas composition. Concentrations of CO, carbon dioxide (CO_2), NO_x , O_2 , SO_2 , and total hydrocarbons (THCs) are measured by CEMs and continuously recorded using a computerized data acquisition system.

A Thermo Systems, Inc., scanning mobility particle sizer (SMPS) was used to measure particle size distributions for particles with diameters in the range of 0.01 to 1.0 μm . SMPS samples were extracted from the PBS stack isokinetically and diluted with filtered nitrogen (N_2) to a ratio of approximately 5 parts N_2 to 1 part stack gas. Additional details of the system design and operation are described elsewhere.^{4,5}

Particle size distributions were also measured using an in-stack cascade impactor. An Andersen[®] impactor was used in a modified California Air Resources Board (CARB) Method 501.⁶ These tests modified the CARB method slightly to allow for use in the research combustor. The CARB method places the impactor precutter in the stack, whereas the impactor was placed outside the stack for the PBS tests. Only three runs were used during research testing rather than the seven runs specified by the CARB method. The use of both the SMPS and the impactor allowed the particle size distribution to be determined over a larger range of sizes than would be

possible when using only one of the two instruments individually. The use of the impactor also provided captive size-segregated samples for later analysis.

Particle concentrations were determined using EPA Method 5,⁷ and EPA Method 29 was used to determine metal concentrations in the flue gases.⁸ The particle concentration option was used during the Method 29 operation, but the mercury option was not used, meaning that the Method 5 procedure used the same train as Method 29, allowing a single sampling train to be used to determine both PM mass and metal concentrations. The Method 29 samples were analyzed for arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), Mg, Ni, antimony (Sb), V, and zinc (Zn).

EPA Method 0030 was used to sample the concentration of volatile organic compounds (VOCs) in the flue gases of all three fuels.⁹ EPA Method 0010 (sometimes referred to as a Modified Method 5) was used to sample semivolatile organic compounds for all three test conditions.¹⁰ Triplicate samples were taken of the semivolatile organic compounds and VOCs.

Results

Three fuels, Orimulsion 100, Orimulsion 400, and a No. 6 fuel oil, were tested at a constant heat input rate of 1×10^6 Btu/hr (293 kW). The O₂ levels for the tests were designed to be at a level that resulted in 100 ppm of CO or less. However, fluctuations in the exhaust draft resulted in significant fluctuations in CO levels, and the measured average O₂ levels during the tests were slightly higher than desired, ranging between 2.9 and 3.5%. Four test runs were made for each fuel, with fuel flow rates remaining relatively constant across the runs. Average concentrations of CO, NO, SO₂, and PM are presented in Figure 2 for each of the three fuels tested.

Average CO emissions were between approximately 15 and 40 ppm (corrected to 3% O₂) for all runs. The average CO emissions for No. 6 fuel oil were slightly lower than for either Orimulsion, but CO emissions were measured at below 20 ppm for at least one test run for both Orimulsion formulations. As noted above, CO emissions are strongly dependent upon O₂ level, and much of the variation in CO may be due to changes in O₂ levels during the test runs. Average O₂ levels for the three conditions were 2.8% for Orimulsion 100, 3.5% for Orimulsion 400, and 3.4% for No. 6 fuel oil. The Orimulsion tests also showed higher variability than did the No. 6 fuel oil tests. Much of this variation was believed to be due to more and larger changes in O₂ level during the Orimulsion test runs than were seen during the No. 6 fuel oil runs. CO increased significantly for all three fuels as O₂ levels dropped below a certain level.

Nitrogen oxide (NO) emissions averaged near 500 ppm (corrected to 3% O₂) for each of the three fuels. Here, the NO values were much steadier across test runs for Orimulsion 100 and Orimulsion 400 than for No. 6 fuel oil. Given the range of uncertainty in the average values, no significant difference in NO emission levels was found between the three fuels. There was a slight drop in NO with decreasing stack O₂, similar to other hydrocarbon fuels.

Average SO₂ emissions as measured using CEMs were found to be essentially the same for each of the three fuels, at 1000 ppm (corrected to 3% O₂). Although the average SO₂ measurement for the No. 6 fuel oil was slightly lower than either of the Orimulsion formulations, the measured variability in the average value for the fuel oil makes it impossible to state that there is any significant difference between SO₂ emissions from the No. 6 fuel oil used in these tests and either of the two Orimulsion formulations.

Using the mini acid condensation system (MACS) sampling train,¹¹ SO₂ concentrations were measured at 1220 ppm for the No. 6 fuel oil, 1640 ppm for Orimulsion 100, and 2010 ppm for Orimulsion 400. Based on the analyses of the fuels' sulfur contents, if 100% of the sulfur were to be emitted as SO₂, one would expect SO₂ concentrations to be roughly 1000 ppm for the No. 6 fuel oil, 2400 ppm for Orimulsion 100, and 1800 ppm for Orimulsion 400.

PM emissions do show some differences between the three fuels. The Orimulsion 400 and No. 6 fuel oil had PM concentrations that were approximately 25% lower (at 150 mg/Nm³) than those from Orimulsion 100 at approximately 200 mg/Nm³. The analysis of Orimulsion 100

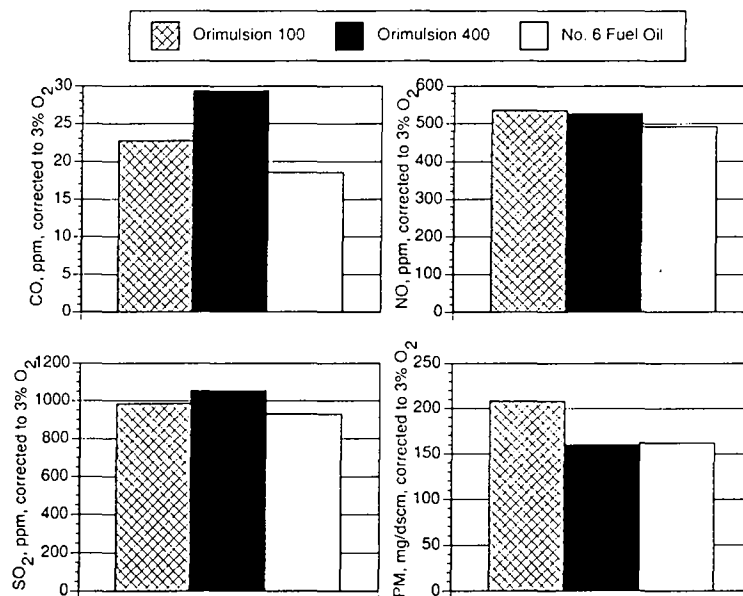


Figure 2. CO, NO, SO₂, and PM concentrations measured during EPA's pilot-scale tests.

showed both higher ash levels and higher amounts of Mg than were present for either of the other two fuels. These differences are likely to have accounted for the difference in PM concentrations between the three fuels. Loss on ignition (LOI) values were determined for PM samples from each of the fuels. The samples were collected on the large dilution sampler filter¹² downstream of a cyclone designed to remove particles larger than 2.5 μm in diameter. Of the three fuels, only the No. 6 fuel oil had any measurable amount of mass in the cyclone catch. The cyclone catch and samples of the large filters for each fuel were subject to LOI analyses. The filters all indicated no measurable LOI (above that measured for a blank filter), and the No. 6 fuel oil cyclone catch had an LOI value of 59%. The high LOI measurement is not unexpected, as the larger particles in the No. 6 fuel oil sample are likely to be largely unburned carbon.

Particle size distributions show a notable difference between Orimulsion and the No. 6 fuel oil. Approximately 80% of the total particle mass captured was smaller than 1 μm in diameter for both Orimulsion 100 and Orimulsion 400, compared to 50% of the particle mass for the No. 6 fuel oil. Approximately 90% of the particle mass was smaller than 2.5 μm in diameter for both Orimulsion formulations, compared with approximately 75% for the No. 6 fuel oil. All three fuels have a bimodal particle size distribution to at least a slight degree, with Orimulsion 400 and the No. 6 fuel oil showing a larger coarse (particles > 6 μm in diameter) mode than the Orimulsion 100. The coarse mode is likely to be due to incomplete combustion of the bitumen droplets in the case of Orimulsion and of the fuel spray droplets in the case of the No. 6 fuel oil.

Results from the SMPS (see Figure 3) provide more detail regarding the particle size distributions for particles smaller than 1 μm in diameter. Even in this size range, there are differences in the size distributions. The Orimulsion 400 and No. 6 fuel oil are quite similar, with modes between 0.06 and 0.08 μm, while the Orimulsion 100 has a smaller mode at just larger than 0.1 μm. The SMPS measurements for the No. 6 fuel oil show a slight indication of the mode near 1 μm, as dV/d(log Dp) begins to curve upward for particles larger than about 0.3 μm.

Metals concentrations differed slightly across the three fuels, largely in relation to the amount of metal present in the fuels. The No. 6 fuel oil had significantly higher concentrations of Cu, Fe, Sb, and Zn than either of the two Orimulsion formulations. The two Orimulsion formulations had higher concentrations of Mg due to the addition of Mg-based additives to the fuel (for Orimulsion 100) and to the flame (for Orimulsion 400), compared to the Mg concentration of the No. 6 fuel oil flue gases. Ni and V concentrations were of the same order of

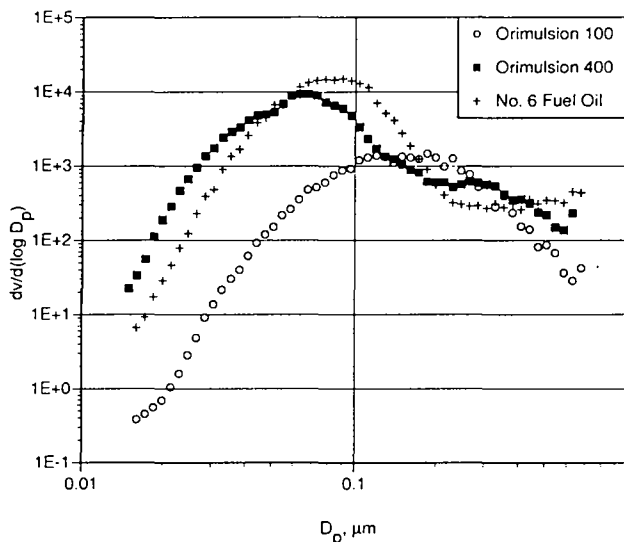


Figure 3. Particle size distributions for the three fuels tested in EPA's pilot-scale test program, as measured by SMPS.

magnitude for all three fuels.

There were no significant differences in VOC emissions between the three fuels, even if variability in measurements is not considered. The largest difference in concentrations of a particular compound between the three fuels was for benzene, with roughly 2.4 $\mu\text{g}/\text{dscm}$ difference between Orimulsion 400 (at 3 $\mu\text{g}/\text{dscm}$) and No. 6 fuel oil (at 0.6 $\mu\text{g}/\text{dscm}$). This difference may be high on a percentage basis, but in absolute terms is very small.

The concentrations of semivolatile organic compounds in the flue gases of the three fuels were relatively low. The compound with the minimum concentration detected in all three fuels was naphthalene, at a level of just over 2 $\mu\text{g}/\text{dscm}$ in Orimulsion 400 flue gases. The highest concentration of the semivolatile organic compounds was the 9.3 $\mu\text{g}/\text{dscm}$ of di-n-butyl phthalate measured in No. 6 fuel oil flue gases. Differences in semivolatile organic compound emissions between the three fuels are slight.

CO_2 emissions are of interest due to the role CO_2 is suspected to play in global climate change. Coal will release between 60 and 75 lb carbon per 10^6 Btu (26-32 g/MJ), while fuel oil's carbon release rates are near 47 lb carbon per 10^6 Btu (20 g/MJ).¹³ Orimulsion's carbon release rate is nearly the same as that for fuel oil. Thus, Orimulsion will generate less CO_2 per unit energy input (and per unit production) than will coal. This difference is being exploited in Denmark, where the conversion from coal to Orimulsion at the Asnaes Power Station is credited with reducing CO_2 emissions by 16%, which is one quarter of Denmark's total national target reduction of 20%.

Conclusions

Emissions of air pollutants from Orimulsion are not significantly different from those from other fossil fuels. From the perspective of air pollutant emissions, Orimulsion fundamentally behaves like a HFO, and the air pollution control technologies applicable to HFO are applicable to Orimulsion. The most significant difference in emissions characteristics is that PM emissions appear to be in a slightly smaller size range than those from HFO, but are significantly smaller than those produced by pulverized coal combustion. Emissions of SO_3 from Orimulsion also appear to be somewhat higher than for other fossil fuels, largely due to the high levels of S and V. Emissions of metals such as Ni and V may also be higher than for other fossil fuels due to the higher level of these elements in the fuel. These results are consistent with the physical characteristics of the fuel.

Results from both full- and pilot-scale tests indicate that emissions from the combustion of Orimulsion can be adequately controlled using commercially available air pollution control technologies. As with any application, proper design, operation, and maintenance are necessary to ensure adequate performance, but there is no indication that special modifications or new control technologies are required to adequately control emissions.

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