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## INFLUENCE OF ATOMIZATION QUALITY ON THE DESTRUCTION OF HAZARDOUS WASTE COMPOUNDS

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The correlation between atomization quality and the destruction efficiency of hazardous organic compounds was studied in a turbulent spray flame. The atomization quality was varied by both changing spray nozzle parameters and by inducing disruptive droplet combustion (secondary atomization) within the flame. The primary atomization quality was characterized by laser diagnotic size distribution measurements. The secondary atomization quality was determined from observations of disruptive atomization intensity on a train of monodisperse droplets within a high-temperature laminar reactor.

For the primary atomization work, No. 2 fuel oil was doped with four target hazardous organic compounds (acrylonitrile, chloroform, benzene, and monochlorobenzene). The destruction efficiency of these compounds was measured under correct atomizer operating parameters and under off-design conditions in which the spray quality was degraded. The degraded spray quality conditions resulted in decreased destruction efficiency of the waste, and increased combustion intermediate emissions (carbon monoxide and total hydrocarbons). Comparison of measured droplet size distributions with performance showed that destruction efficiency was more closely correlated with the large droplet wing of the dropsize distribution than with the mean droplet size. A droplet evaporation/trajectory model showed that the appearance of the target compounds in the exhaust corresponded with the fraction of the droplets that passed through the primary reaction zone unevaporated.

The distruptive droplet combustion results showed that hazardous waste compounds are capable of inducing secondary atomization. Testing of benzal chloride (which did not cause disruptive combustion in No. 2 fuel oil) and isopropanol (which caused violent disruption) in the turbulent flame reactor showed that the occurrence of disruptive combustion correlated with increased target compound destruction efficiency and reduced combustion intermediate emissions. Thus, the results suggest that the presence of certain compounds or additives in waste streams may assist in obtaining improved performance when primary atomization is poor, as it is for slurry or sludge waste streams.

## Introduction

Incineration offers a final and complete means of disposing organic hazardous wastes. However, incineration will be accepted only if efficient waste destruction is assured. In the absence of fundamental information, overly conservative designs may be adopted, and the process may become economically unattractive.

The study of the fundamental processes that limit the efficiency of the most common class of incinerator, liquid injection,<sup>1</sup> has been underway a relatively short amount of time. Understanding of the incineration process is complicated by the complex chemistry involved in the destruction of hazardous waste compounds, and by the strong coupling between transport rate processes and chemical rates that are evident in practical systems. In addition, the escape of waste can be expected to be dominated by unusual pathways or time-temperature histories (i.e., an unusually cold path) rather than pathways characteristic of the mean flow or time-temperature history. Thus, the pathways of interest deal with the wings of the mixing and temperature distributions rather than with the mean conditions.

Past work on the flame chemistry of hazardous waste compounds has been motivated by various factors such as the inhibition effect of halogens and soot formation from complex molecules. Recently, flame work has appeared in which hazardous waste chemistry issues are addressed.<sup>2</sup> Also, the destruction rate due to the long time, moderate temperature exposure typical of afterburners has been extensively studied.<sup>3</sup> The influence of flame-zone parameters on the waste destruction efficiency performance of a sub-scale turbulent spray flame has been studied,<sup>4</sup> and an extensive evaluation of full-scale performance in the field has been completed.<sup>5</sup> However, the identity of the mechanism or combination of mechanisms that give rise to the waste emissions detected from high-efficiency incinerators has not been established. Also, the merchanisms by which wastes escape during upset conditions and the relative response of various hazardous compounds to these environments are poorly understood.

Previously, we identified poor atomization quality as one mode by which an incinerator flame zone could fail to efficiently destroy waste.<sup>4</sup> The objective of the present work is to explore this "failure condition" and to develop an understanding of the mechanism and consequences associated with both a degradation in primary atomization and the promotion of secondary atomization by disruptive droplet combustion.

### Experimental

For primary atomization, the approach was to characterize the droplet size distribution produced by a series of sub-scale nozzles. This was done under cold-flow conditions by laser diffraction. These droplet size data were directly compared with destruction efficiency results from a small-scale reactor to evaluate the influence of droplet size on destruction efficiency. In the second portion of the study, the influence of disruptive droplet combustion or "secondary atomization" on waste destruction efficiency was investigated.

The hypothesis investigated here is that the volatile wastes present in a multicomponent waste stream can, in high concentrations, induce secondary atomization<sup>6,7</sup> and improve the overall destruction efficiency. The approach was to screen a series of wastes for secondary atomization potential and compare the destruc-



FIG. 1. Spray characterization rig.

tion efficiency in the small-scale reactor for conditions where secondary atomization was present against conditions for which it did not occur.

The spray characterization rig, illustrated in Fig. 1, consists of a plexiglass cylinder in which the nozzle is mounted on centerline downfired. Air is co-flowed axially around the nozzle to simulate the combustion air field and to prevent recirculation of droplets into the optical path. Two ports at opposite sides of the chamber provide access for the Malvern 2600 HSD particle size analyzer. This instrument infers dropsize distributions by measuring the angular intensity distribution of light scattered from a collimated laser beam by the spray. A model independent algorithum reduces the data and is capable of resolving multimodal distributions.

The slip-flow reactor is a simple device used to study the physical response of droplets to a high-temperature environment. In this study the reactor, shown in Fig. 2, was used to screen mixtures of No. 2 fuel oil and hazardous target compounds for secondary atomization intensity. The reactor consists of a  $5 \times 28$  cm flat-flame burner downfired into a chimney of similar dimensions. The fuel-lean flat flame is supported on a water cooled sintered stainless



FIG. 2. Slip-Flow reactor.

steel plate. The chimney is fitted with four  $15 \times 28$  cm Vycor windows for optical access. As shown in the figure, the fuel droplets are injected ballistically normal to the hot gas flow. The droplet stream is generated by the Berglund-Liu<sup>8</sup> vibrating orifice technique. Against the laminar background gas, disruptive droplet combustion can be easily detected visually. The extent of disruption was qualitatively rated in terms of an intensity scale by visual observation.

The turbulent flame reactor used to obtain flame-zone target compound destruction efficiencies is described elsewhere.<sup>4</sup> Its salient features include a turbulent, swirl stabilized spray flame formed by a hollow-cone pressure jet nozzle (Delevan WDA-series, 60° cone angle) and water-cooled stainless-steel walls to quench post-flame reactions. Samples are withdrawn after a series of mixing baffles at the reactor exhaust. The reactor is capable of generating data under conditions of variable atomization quality, stoichiometry, air velocity, air swirl, thermal loading, and with a quenching surface placed in the flame. It is thus designed to simulate at subscale the features of a full-scale liquid injection incinerator flame.

The destruction efficiency of the waste compounds was measured in the exhaust of the turbulent flame reactor by use of a Nutech Volatile Organic Sampling Train. In brief, gas samples are drawn through chilled cartridges within which the volatile organic compounds are absorbed onto Tenax-GC. After sampling, the compounds are released by thermal desorption and analyzed on a gas chromatograph equipped with a flame ionization detector. Details of the procedures are provided elsewhere.<sup>9</sup>

### **Results and Discussion**

## Primary Atomization

The purpose of the tests reported here was to quantitatively characterize the droplet size distributions from the test nozzles. The nozzles were operated at both the design points and under off-design conditions. These results were compared with the target compound destruction efficiency obtained using these nozzles in the turbulent flame reactor.

Figure 3 illustrates the droplet size distribution obtained at 0.922 gm/sec fuel flow. The data set labeled "On-Design" was obtained for a 0.922 gm/sec capacity nozzle (nominally 1 gallon/hr) and thus represents the size distribution resulting from correct operation. The "Off-Design" data set is for the identical flow rate, but an oversized nozzle (1.38 gm/sec or 1.5 gallons/hr). The use of oversized pressure jet nozzles results in low fluid pressure (1360 vs. 600 kPa) and low atomization energy.

Each of these atomizer conditions was used in the turbulent flame reactor to determine the influence of atomization quality on target compound destruction efficiency. The No. 2 fuel oil was doped to 3.0 weight percent with an equimolar mixture of target compounds: acrylonitrile, chloroform, benzene, and monochlorobenzene. The reactor was operated with



FIG. 3. Weight percent of droplets associated with each size range from laser diffraction measurements.



FIG. 4. Penetration of individual waste compounds as a function of theoretical air from the turbulent flame reactor. Symbols: acrylonitrile-squares, chloroform-circles, benzene-triangles, and monochlorobenzene-diamonds.<sup>9</sup>

an inlet air velocity of 7.1 m/sec (at stoichiometric) and a swirl number of 0.8. Both the on-design and off-design conditions were run with theoretical air as the independent variable.

Figure 4 shows the fraction of each of the target compounds that escaped destruction as a function of percent theoretical air (this is termed the "penetration" in the following discussion). The on-design nozzle results show behavior similar to that documented previously.<sup>4</sup> This includes (1) a range of low penetration between 100 and 200 percent theoretical air, (2) an increase in penetration at low theoretical air due to fuel-rich pockets breaking through the flame, and (3) an increase in emission at high theoretical air that signifies that some portions of the feed are sufficiently fuel-lean to avoid ignition, or that reacting packets are quenched by dilution prior to complete reaction.

Comparison of the on-design and off-design data shows that the penetration below 100 percent theoretical air is not significantly different. Penetration above 200 percent theoretical air is similar, although values are increased by a factor of 2-3 over the on-design case. However, in the nominal operating range of 100 to 200 percent theoretical air the penetration is substantially increased for the off-design condition. Thus, a condition in which the flame alone was quantitatively destroying the waste for on-design atomization was reduced to a condition for off-design in which a small, but significant fraction of the waste bypassed flame zone destruction.

Two hypothetical mechanisms can be identified by which poor atomization quality can influence penetration. In the first, droplets which are too large to evaporate in the available time penetrate to the reactor wall. The liquid evaporates on the wall and exits the reactor along the cold wall boundary layer. In the second mode, the droplets penetrate through the flame-zone without fully evaporating until well into the post-flame region. Here, mixing or temperature is not sufficient to ensure complete destruction. (Note that the failure of the flame zone to efficiently destroy the waste does not preclude destruction in the post-flame zone. However, it is a necessary condition that the flame zone be inefficient for the entire device to be inefficient.)

In both cases the inefficiency arises due to unevaporated droplets escaping the flame. Thus, the key to interpreting the data is to use the measured droplet size distributions to define the fraction of the feed that escapes the flame unreacted.

The tool used to define the fraction of the feed that escapes the flame unreacted is the evaporation time plot contained in Fig. 5. This plot is based on the droplet evaporation model developed earlier<sup>4</sup> based on Spalding's work.<sup>10</sup> The droplet diameter for which flame penetration is predicted exceeds 150 microns. Since the Sauter-mean diameter of the on- and off-design sprays were respectively 72 and 109 microns, a treatment based strictly on mean diameter shows that all droplets evaporate within the flame.

An alternative approach is to calculate the fraction of each of the size classes that would be expected to exit the flame without evaporation. These calculations, the result of which are presented in Fig. 5, show that all of the emissions are associated with the largest droplet sizes. For the on-design condition the fraction of original mass estimated to escape the flame unevaporated is 0.4 percent, and for the off-design condition, the estimate is 3.9 percent. It must be recognized that some uncertainty in resolution is introduced into the data by the broad size classes used at the large diameter



FIG. 5. Evaporation time and calculated percentage of original mass penetrating flame unevaporated as a function of original droplet size.

limit of the laser diffraction sizing technique. However, direct comparison of the two cases shows approximately an order of magnitude increase in the amount of material escaping the flame zone. This illustrates the key role that characterizing the "wings" of the distribution functions, and, in particular, the wings of the droplet size distribution have on understanding waste emission mechanisms from incinerators.

These findings suggest a methodology for evaluating nozzle performance in relation to a particular incinerator geometry. This consists of an evaluation of the nozzle spray angle, droplet size, and droplet velocity. This information is matched with the incinerator geometry to yield the approximate maximum tolerable droplet size to avoid the wall or primary reaction zone.

### Secondary Atomization

Adequate primary atomization may be difficult for some waste streams that are unusually viscous or contain solids (i.e., sludges and slurries). Also, portions of the nozzle may wear or become plugged during use, resulting in a reduction of atomization quality.

Secondary atomization by disruptive combustion has been investigated as a means of improving combustion efficiency through the in-flame reduction of droplet diameter via fragmentation. Reviews of the experimental data<sup>6,7</sup> and theory<sup>11</sup> are available in the literature. Since most wastes are multicomponent mixtures of varying volatility, secondary atomization may occur naturally and may be a means of overcoming the limitation imposed by primary atomization.

The two questions addressed on secondary atomization in the present work were (1) whether secondary atomization can be induced by the presence of hazardous compounds in No. 2 fuel oil, and (2) whether this secondary atomization has the capability of improving target compound destruction efficiency. Five compounds were selected for doping into No. 2 fuel oil for secondary atomization screening. The compounds were selected to represent a broad range of volatility with respect to the fuel oil (boiling range: 483–533 K). These included dichloromethane, acrylonitrile, benzene, isopropanol, and benzal chloride. The boiling points are shown in Table I.

Each of the compounds were screened in the

Compound Boiling Point (K)	Isopropanol 355	Acrylonitrile 352	Dichloromethane 312	Benzene 478	Benzal Chloride 478
Percent in Fuel					
0.5	N	N	, N	N	Ν
2.0	S	N	N	N	Ν
5.0	v	S	S	S	Ν
10.0	v	R	S	S	Ν
20.0	v	R	R	R	N

TABLE I. Secondary Atomization Intensity

Disruption Intensity: N-None, S-Sporadic, R-Regular, V-Violent

slip reactor at 0.5, 2, 5, 10, and 20 weight percent in the No. 2 fuel oil. The results are presented in Table I in terms of an arbitrary intensity scale. These results indicate that secondary atomization is active only for compound concentrations above 2 percent except for isopropanol, which was active above 0.5 percent. Also, for secondary atomization to occur, some difference between the boiling points of the base fuel and the added compound must exist. For example, benzal chloride, which has a boiling point comparable with that of No. 2 fuel oil, showed no activity at any concentration. The results indicate that intensity is not entirely a function of boiling point differential. For example, isopropanol has a boiling point of 355 K, but it induced a substantially more active reaction than dichloromethane (312 K). Thus, other factors than boiling point differential (e.g., compound polarity) are related to intensity.

The screening tests indicated that isopropanol and benzal chloride represent the limits of secondary atomization intensity. As a result, these two compounds were selected for destruction efficiency tests in the turbulent flame reactor at 0.5, 2.0, and 10.0 weight percent in No. 2 fuel oil. The experiments were designed to determine the effect of compound concentration for (1) a compound for which no secondary atomization occurs across the entire concentration range, and (2) a compound for which no secondary atomization occurs at low concentrations, but a strong response is obtained at high concentrations. Thus, benzal chloride yields the concentration dependence in the absence of secondary atomization. Any strong additional concentration dependence for isopropanol can be attributed to an increase in secondary atomization intensity with concentration.

The test condition corresponded to the offdesign atomization condition illustrated in Fig. 3. In all other respects, the turbulent flame reactor was set for high efficiency operation (120 percent theoretical air, 0.8 swirl number). Thus, the only variables were test compound type and concentration. The destruction efficiency results are shown in Fig. 6. Waste penetration is plotted against the weight percent waste in the fuel for the two test compounds. Benzal chloride shows an approximately one order of magnitude decrease in penetration between 0.5 and 10 percent waste concentration. Since no secondary atomization takes place for this compound, the concentration effect on penetration must be due to other factors. For isopropanol, however, the effect of concentration is much more pronounced. Between 0.5 and 10 percent the penetration



FIG. 6. Penetration of benzal chloride and isopropanol as a function of concentration in the No. 2 fuel oil from the turbulent flame reactor.

decreases from greater than  $10^{-3}$  to below 5 ×  $10^{-6}$ . Significantly, this increase in efficiency occurs concurrently with an increase in secondary atomization intensity from none to violent. Thus, at least a substantial portion of the difference in behavior between benzal chloride and isopropanol can be attributed to secondary atomization.

In addition to waste destruction efficiency, the overall combustion efficiency was also influenced by the dopants. In Fig. 7 the CO and total hydrocarbons emissions (measured as  $CH_4$  by a flame ionization detector)<sup>9</sup> are plotted as a function of target compound concentration. Note that compound type and concentration were the only variables; in all other respects each of the experiments were identical. The data show three key points:

- 1. Increased isopropanol concentration *increased* combustion efficiency. This occurred concurrently with increased secondary atomization intensity.
- 2. Increased benzal chloride concentration *decreased* combustion efficiency. This occurred in the absence of secondary atomization.
- 3. At 0.5 percent, where secondary atomization was absent for both compounds, the CO and hydrocarbon emissions for the benzal chloride were lower than those for isopropanol.

The final point illustrates that other dopantdependent mechanisms are active.

This work suggests that the efficiency of liquid injection incinerators can be improved by blending small amounts of high-volatility liquids into the waste stream. The blending agent



FIG. 7. Carbon monoxide and total hydrocarbon emissions (dry, corrected to 100% theoretical air) as a function of benzal chloride and isopropanol concentration from the turbulent flame reactor.

may be a second waste stream of markedly different volatility rather than a pure organic liquid. Such blending may be most appropriate under conditions where atomizer performance is expected to be a limiting factor, as in slurry or sludge combustion.

### Conclusions

- 1. The degradation of atomization quality appears to influence target compound destruction efficiency by penetration of the largest droplets through the flame zone or to the wall.
- 2. Hazardous compounds are capable of caus-

ing disruptive droplet combustion (secondary atomization) when diluted in fuel oils.

3. The occurrence of secondary atomization was found to correlate with improved waste compound destruction efficiency in a turbulent spray flame operating in an atomizer limited mode.

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