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EVALUATION OF MOLTEN SCRUBBING FOR FINE PARTICULATE CONTROL



**Industrial Environmental Research Laboratory
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EVALUATION
OF MOLTEN SCRUBBING
FOR FINE PARTICULATE CONTROL

by

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FOREWORD

This document contains a review and evaluation of molten scrubbing as a novel concept for the control of fine particulate.

The study was performed for the Environmental Protection Agency, Research Triangle Park, North Carolina. Dr. D. C. Drehmel was the EPA Task Officer. The Aerotherm Program Manager was Mr. Fred Moreno. Acting as Technical Advisor for the task was Dr. C. B. Moyer. The study was performed during the period December 1975 through February 1976.

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LIST OF SYMBOLS

C'	Cunningham correction factor (dimensionless)
C_D	drag coefficient (dimensionless)
d	diameter (cm)
d_{p_a}	aerodynamic diameter ($\text{cm } \sqrt{g/\text{cm}^3}$)
f	empirical proportionality constant (dimensionless)
K_p	inertial impaction parameter (dimensionless)
ΔP	pressure drop ($\text{cm H}_2\text{O}$)
P_t	penetration (dimensionless)
Q	flowrate (cm^3/sec)
r	radius (cm)
Re	Reynolds number (dimensionless)
t	time (sec)
u	velocity (cm/sec)
y_0	distance from collector axis to limiting streamline (cm)
Z	axial distance (cm)
η	target efficiency (dimensionless)
μ	viscosity (poise)
ρ	density (g/cm^3)
σ	surface tension (dyne/cm)

Subscripts

d	drop
g	gas
l	liquid
p	particle
r	relative
t	throat
0	reference

Superscripts

—	average
~	dimensionless

SECTION 1

SUMMARY

The purpose of this study was to analyze and evaluate molten scrubbing as a new fine (≤ 3 microns) particulate control concept and to investigate its applicability to high temperature, high pressure processes.

The concept of molten scrubbing for particulate capture is indeed a novel one. Previous experience in molten scrubbing was almost exclusively concerned with sulfur removal, and placed no emphasis on particulate collection. The literature search for the study revealed the paucity of data on this subject. Only two projects were found to have any application to this subject at all. IGT's molten metal scrubbing process, however, is proprietary and their test rig is currently shut down. The work of Battelle Northwest appears to be the only applicable study with available data. Their process involves the scrubbing of both hydrogen sulfide and particulate with a molten liquid of alkali metal salts for application to hot fuel gas from a coal gasifier.

Existing theories provide a reasonably detailed description of the particle collection mechanisms involved in atomized liquid scrubbers. These theories should apply equally well at the high temperatures found in molten scrubbers, so the effects of the high temperatures on particle collection can be predicted. The collection efficiency for a given particle size decreases as the temperature of the gas increases, according to the best available design equations. These predictions, however, have not been confirmed experimentally for molten scrubbers.

Thus far, the experimental work at Battelle has revealed the following:

- Overall collection efficiency is not as great as expected (this may be attributed to poor design and/or operational problems).
- The high corrosiveness of the scrubbing medium presents a difficult materials handling problem.
- Carryover of molten liquid droplets in the scrubbed gas was much greater than the allowable specs on gas turbine inlets.

- Molten salt reactions with other gaseous pollutants can potentially degrade the scrubbing liquid.
- Decomposition of the molten salts occurs at temperatures around 900°C; thus, limit controls for very high temperatures are necessary.
- Insoluble particulate build-up necessitates the use of in-line filters in any regenerable scrubbing process.
- Energy requirements for molten scrubbers are estimated at 7 to 10 kW/Mcfm, which are typical for scrubbing processes at more conventional temperatures.

Even though it has not been adequately demonstrated as yet, the application of molten scrubbing to high temperature/high pressure advanced energy processes for fine particulate control appears possible. However, before effective molten scrubbing systems for particulate removal are available, the developmental problems described above need to be solved.

SECTION 2

INTRODUCTION

One important proposed use of low Btu fuel gas derived from coal gasification processes is the generation of electric power by combustion and expansion of the gas through a gas turbine. Process temperatures associated with coal gasification are typically near 3000°F, which dictates that the product gas be cooled to the 1800°F gas turbine inlet temperature. However, planned improvements in turbine blade technology may allow inlet temperatures up to 3000°F. Combining the high temperature gas turbine with a repowered boiler may result in an overall cycle efficiency of greater than 40 percent.

Successful operation of such a system is predicated on the conservation of sensible heat of the fuel gas. However, the raw gas from the gasification process contains impurities, notably sulfur compounds and particulate, which are incompatible with turbine operation and must therefore be removed. Since the fuel gas must remain at elevated temperatures, a high temperature clean-up process is required. Such technology is not immediately available, but many novel concepts have been proposed. In addition to conserving the sensible heat of the hot gas stream, many of these concepts have been proposed as fine (≤ 3 microns) particulate cleanup processes. Fine particles have recently come to be recognized as being much more significant air pollutants than larger particles.

One proposed concept, molten scrubbing, utilizes venturi or spray scrubbing with molten fluids that remain in the liquid state at the cleaning temperature. This report presents a review and evaluation of the molten scrubbing processes currently under development in the U.S.

The work presented in this report was performed by Acurex/Aerotherm for the EPA's Industrial Environmental Research Center as Task 24 on Contract 68-02-1318. The scope of work was defined by EPA as follows:

1. Conduct a limited literature search to determine if similar scrubbing devices have been developed or proposed by others. Determine if the device is truly novel and define the mechanisms which are responsible for particulate capture.
2. Determine the reliability and significance of any experimental data submitted.

3. Assess the practicality of the device for collection of fine particulate. Estimate the capital and operating costs for the device evaluated.
4. Determine which sources might be controlled by the system and estimate the probability of successful application.

The evaluation of the molten scrubbing concept is presented in the following sections of this report.

SECTION 3

LITERATURE SEARCH

The search for literature concerned with molten scrubbing occurred in three steps:

- In-house literature review
- Telephone interview with molten scrubbing researchers for information on their processes
- Computer-aided literature search of NTIS, COMPENDIX, and Physical and Chemical Abstracts resources, performed by the Lockheed DIALOG service.

The first step produced very little in the way of useful information. The telephone calls uncovered reports, papers, and the general status of the Battelle, IGT, and Atomics International processes. The final step using the computer search produced several papers of general interest, all of which are listed at the end of the reference section of this report.

In all, the literature search clearly revealed that molten scrubbing is a very new and developing process, and that there is a growing recognition of its potential as a possible high temperature fuel gas purification process.

SECTION 4

PROCESS DESCRIPTIONS

Although some pilot- and lab-scale experience with molten scrubbing has been obtained by the Pacific Northwest Laboratory of the Battelle Memorial Institute (Battelle-Northwest), the Atomics International Division of Rockwell International (AI), and the Institute of Gas Technology (IGT), in all cases the data reported have been, at best, incomplete, especially with respect to particulate scrubbing.

The Institute of Gas Technology (IGT) in Chicago recently operated a small lab-scale molten metal droplet contactor designed for sulfur and particulate removal. The metal in this case was presumably molten iron. The device is currently shut down, and since the process is proprietary, no further information is available (Reference 2).

AI investigated a pilot scale molten scrubbing process aimed at removing SO_2 from boiler stack gases (Reference 3). The AI scrubber used the ternary eutectic of $(\text{Li,K,Na})_2\text{CO}_3$ (M.P. 396°C) as the working fluid. Gas scrubbing was effected with a spray contactor operating at 455°C , and mist elimination was accomplished with a York wire mesh demister. However, particulate collection was of secondary concern in the AI process; in fact, an electrostatic precipitator was used before the scrubber to reduce the flyash load of the gas before scrubbing the SO_2 .

Battelle-Northwest has developed a molten scrubbing process designed to remove H_2S and particulate from fuel gas produced by coal gasification (Reference 1). Because the Battelle-Northwest data is the most comprehensive, this process will be singled out for more thorough discussion below. Observations and conclusions based on Battelle-Northwest's experience should be generally applicable to all molten scrubbing processes, as other proposed processes should differ in, at most, choice of working fluid or gas-liquid contacting scheme. The underlying principles of operation supporting the Battelle-Northwest process, and the problems encountered should be universal to all molten scrubbing processes.

The Battelle-Northwest pilot-scale scrubbing process is shown schematically in Figure 1. Fuel gas from a gasifier is heated to 850°C and flows through a venturi where it atomizes a stream

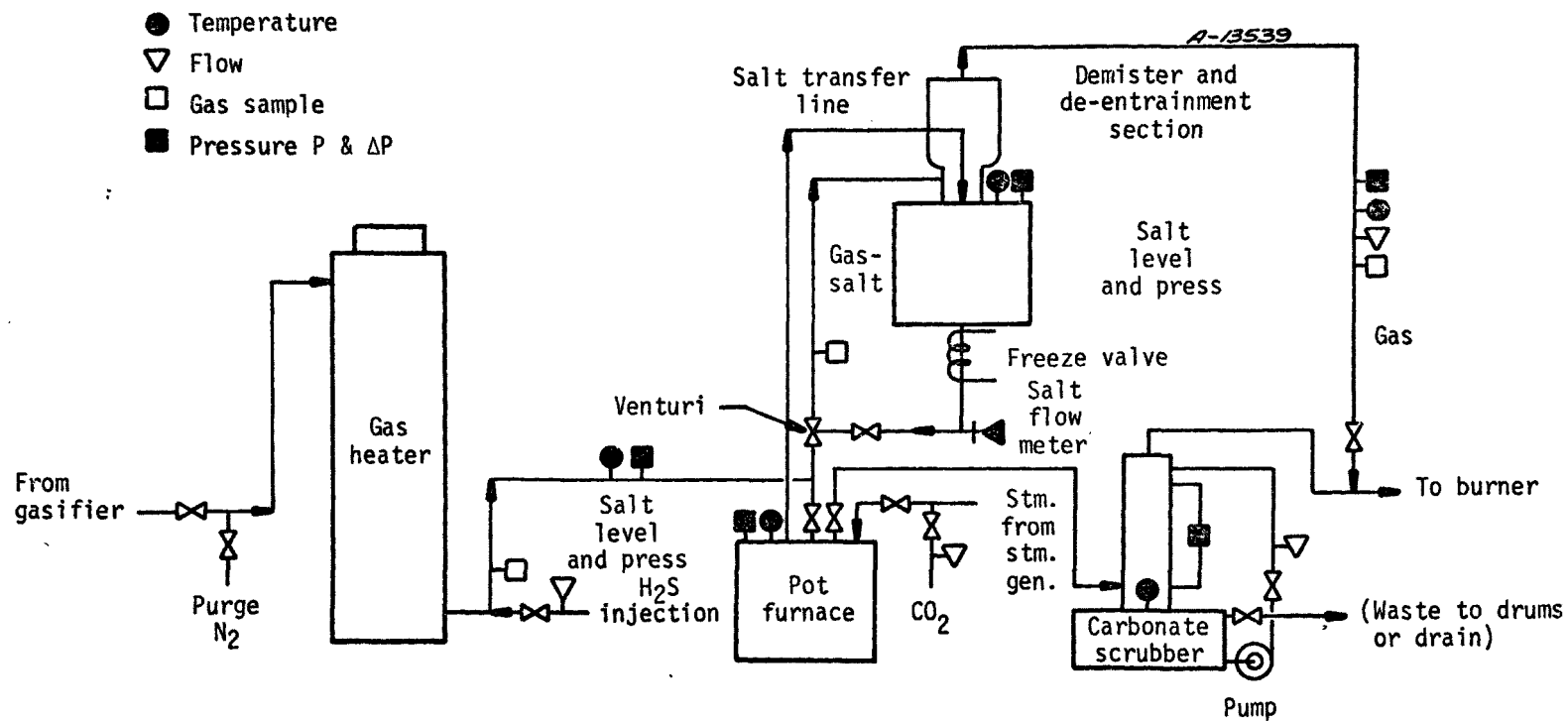


Figure 1. Schematic of Battelle-Northwest molten scrubbing process (Reference 1).

of molten salts entering from a reservoir. Good gas-liquid contact is obtained and both sulfur compounds and particulates are entrained in the liquid droplets. Gas-liquid separation is accomplished in a de-entrainment, mist eliminator section, with the cleaned gas flowing to a burner, and the spent molten salt returning to the reservoir.

The working fluid used in the Battelle scrubber was a salt mixture of approximately 36 wt% Na_2CO_3 , 37 wt% K_2CO_3 , 13 wt% Li_2CO_3 , and 14 wt% CaCO_3 . CaCO_3 was added to facilitate H_2S scrubbing; and the amount of Li_2CO_3 was decreased from the 1:1:1 weight ratio of alkali metal carbonates in a eutectic mixture, since the lithium salt was by far the most expensive of the three carbonates used. The above mixture is a noneutectic and has a melting range of 393°C to 541°C. Above 541°C the mixture consists of one liquid phase.

No provision was made in the Battelle pilot facility for either insoluble particulate removal or the continuous regeneration of metal sulfides to metal carbonates in the spent salts, although batch carbonate regeneration was accomplished by blowing CO_2 and steam through the spent melt. In a continuous regeneration system, insoluble particulates presumably could be filtered from used salts and the batch carbonate regeneration scheme could be adapted to a continuous system.

Gas-liquid contacting in the Battelle pilot plant was accomplished by a venturi atomizer. As discussed in Section 6, particulate removal in a venturi scrubber is effected by particle inertial impaction onto liquid droplets. The Battelle venturi was designed to remove 55 percent of the particles 4.0 microns or larger.

A key consideration in any scrubbing scheme is the final gas-liquid phase separation after the scrubbing is complete. Mist elimination in the Battelle pilot plant was accomplished in two stages. A de-entrainment section consisting of a column packed with 1-inch diameter ceramic (Al_2O_3) balls was designed to remove 50 percent of the droplets 3.5 microns in diameter and 95+ percent of the droplets greater than 10 microns. This section was followed by a demister device consisting of packed rolls, 3/4 inch in diameter, made of 3-inch wide stainless steel screen.

Perhaps the most important consideration in the design of a scrubbing process utilizing molten salts is materials choice. The combination of high temperature (850°C in the Battelle process) and the high corrosive nature of molten salts makes the choice of materials of construction quite difficult. Both AI and Battelle conducted short-term static corrosion screening studies on a wide range of metals, metal alloys, and ceramics. In addition, AI conducted long-term corrosion tests in rotating capsules. AI found that Type 347 stainless steel was suitable for service below 500°C, while Battelle found aluminized Type 304 stainless steel sufficiently corrosion resistant

at 760°C. However, neither material was exceptionally resistant to attack by the hot molten $(\text{Li,K,Na})_2\text{CaCO}_3$; CaS mixture. Alumina ceramics proved quite resistant but these materials were rejected as not being readily amenable to conventional fabrication and assembly techniques.

SECTION 5

PERFORMANCE DATA

Performance data on particulate removal from the Battelle molten scrubber is presented below. This scrubber is the only one for which particulate removal data are available, and even these data are scanty and inconclusive because of the start-up and operating problems experienced.

The Battelle molten scrubber was operated for six performance runs during late 1974. Of these, only three runs yielded H_2S scrubbing data and only two of these yielded particulate removal data with any degree of reliability.

Several shake-down problems were encountered, most of which resulted in salt crystallization and subsequent line plugging throughout the process stream. This occurred in spite of the care taken with trace heating in the pilot plant design.

A gas flowrate of 70 scfm at 750°C - 800°C and 1 to 3 psig was used in the two runs yielding particulate removal data. The salt flow for these runs was estimated at 2 gpm. Particulate gas samples were isokinetically withdrawn using high temperature stainless steel probes both prior to and after the molten salt scrubbing step. For the pre-venturi sample, the total inlet particulate burden was determined by passing the sample through a heated glass cyclone followed by a high-temperature glass wool filter capable of collecting particles 0.3 microns in diameter. Post-venturi samples were collected and sized using an Anderson stack sampling head capable of sizing particles in the 0.3 to 20 micron range. Data for the two productive runs appear in Table 1. During Run No. 3, the wire mesh demister downstream of the alumina balls de-entrainment section was not used.

Table 1 shows that the overall scrubber collection efficiency was 70 to 90 percent, versus a design efficiency of 99 percent for particles > 4 microns. However, most significant was the fact that approximately 30 wt% of the collected particles in the scrubber outlet gas were entrained salt crystals. This observation has ominous implications if turbine quality gas is the desired product. The Westinghouse Company has specified a maximum allowable limit for alkali metals in gases to present-day turbines of 40 ppb. The large quantity of salt

carryover experienced in the Battelle pilot plant suggests that closer attention be paid to mist eliminator design.

TABLE 1. PARTICULATE REMOVAL WITH BATTELLE MOLTEN SALT SCRUBBER*

	Run No. 3	Run No. 6
Inlet gas particulate burden	0.1 gr/scf	0.0674 gr/scf
Outlet gas particulate burden	0.0122 gr/scf 60 wt% < 4 μ m	0.0223 gr/scf 76 wt% < 4 μ m
Overall collection efficiency	88%	67%
*Reference 1		

The Battelle molten scrubber was in operation for a total of about 25 hours. After Run No. 6 it was disassembled, inspected and cleaned. Visual inspection showed no evidence of major corrosion, though some material erosion was evident immediately downstream of the venturi throat. However, it is doubtful that significant material corrosion evidence would exist after such a short period of operation.

SECTION 6

THEORETICAL DISCUSSION

The Battelle-Northwest pilot plant employed a venturi scrubber for gas-liquid contacting. The principles of operation and collection mechanisms for venturi scrubbers, the effects of high temperature and pressure, and the effects of using a molten scrubbing medium are briefly reviewed in this section.

6.1 PARTICLE COLLECTION MECHANISMS

Venturi scrubbers employ gradually converging, then diverging sections of pipe as shown in Figure 2. The venturi throat is the intersection of the converging and diverging sections of pipe. Generally, liquid is introduced into the gas stream at the throat, and is quickly atomized into small droplets by the high velocity gas. Particles entrained in the gas stream are collected by the liquid droplets primarily through particle inertial impaction onto the droplets. This collection proceeds until the droplets are accelerated to the gas (and presumably the particle) velocity (References 4 and 5).

Consider a single spherical collector (liquid droplet) in a uniform velocity field (gas) containing a suspension of small particles as shown in Figure 3. As the fluid stream approaches the collector the streamlines envelop it. However, due to inertial forces, the particles tend to cross fluid streamlines, impact the collector, and stick to it or become entrained within it. Thus a single collector target efficiency may be defined for the spherical collector,

$$\eta = \left(\frac{y_0}{r_d} \right)^2 \quad (1)$$

where y_0 is the distance from the sphere's axis to the limiting streamline for impaction and r_d is the collector (liquid droplet) radius.

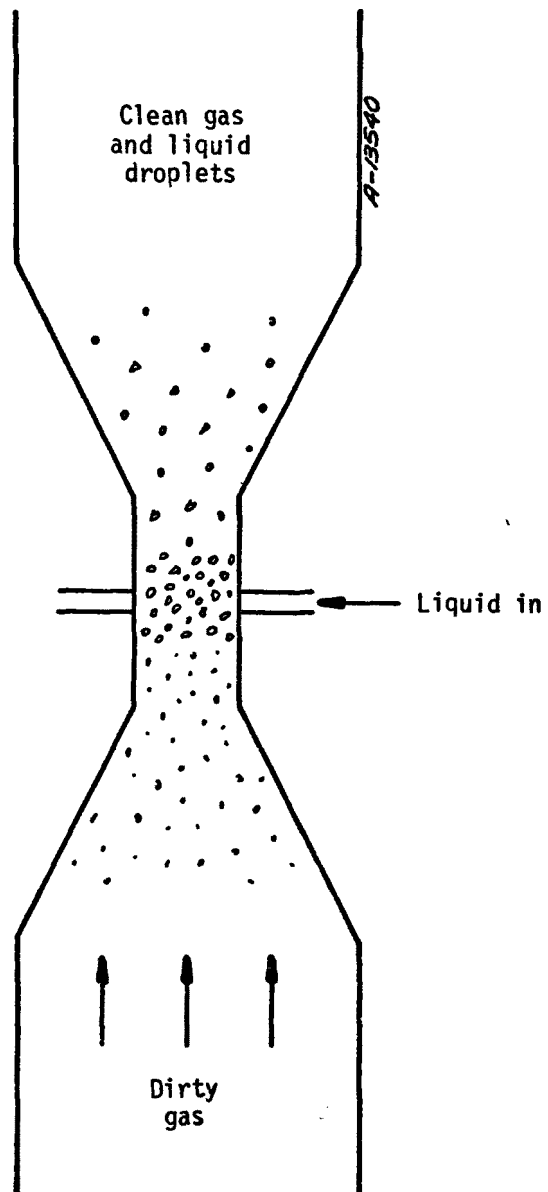


Figure 2. Operation of a venturi scrubber.

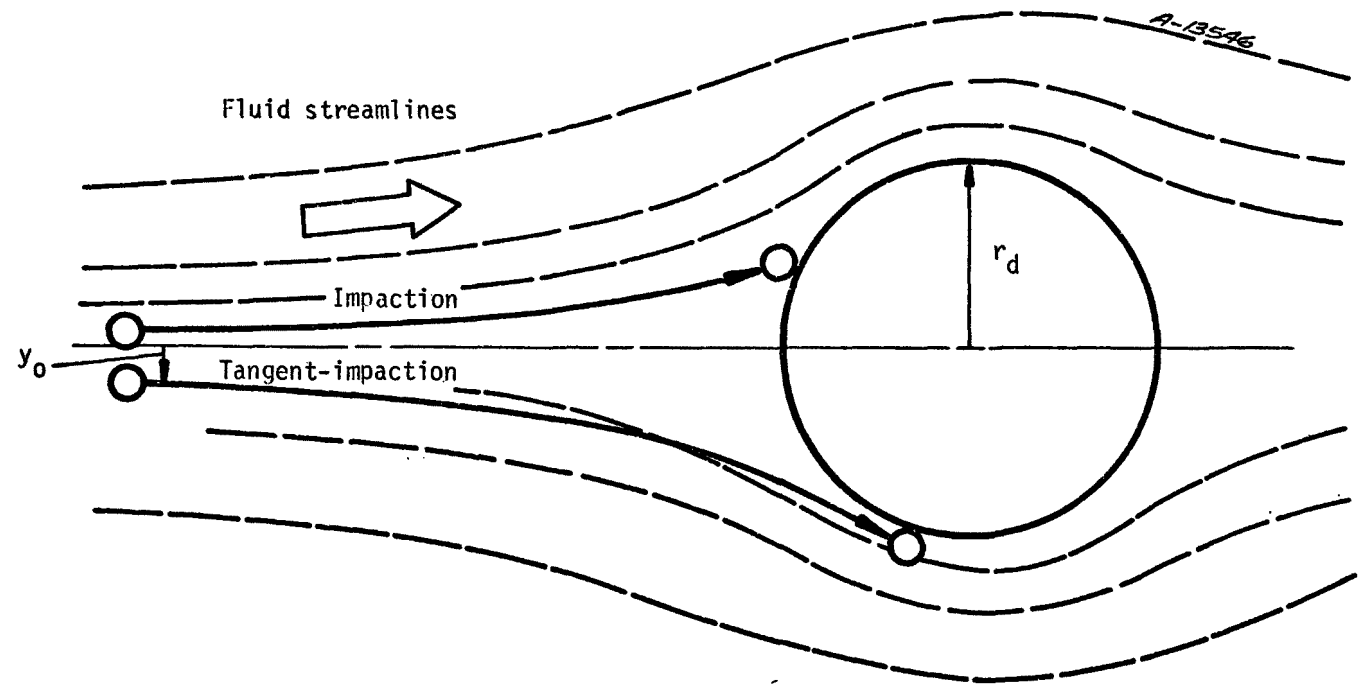


Figure 3. Fluid streamlines and particle trajectories around a sphere (Reference 4).

The dimensionless equations of motion for particles obeying Stoke's law are:

$$K_p \frac{d\tilde{u}_p}{d\tilde{t}} = (\tilde{u}_g - \tilde{u}_p) \quad (2)$$

$$K_p \frac{d\tilde{v}_p}{d\tilde{t}} = (\tilde{v}_g - \tilde{v}_p)$$

where $\tilde{u}, \tilde{v} = u/u_0, v/u_0$; u_0 is a reference velocity,

$$\tilde{t} = \frac{u_0 t}{r_d} \quad (3)$$

and

$$K_p = \frac{C' \rho_p d_p^2 u_0}{18 \mu_g r_d} \quad (4)$$

The quantities u and v are the x and y components of velocity, respectively, t is time, K_p is termed the inertia parameter, and C' is the Cunningham correction factor for the drag on a sphere in steady motion. Equation (2) may be solved for a variety of collector shapes and imposed velocity fields allowing, eventually, a calculation of y_0 , and hence η . Walton and Woolcock (Reference 6) experimentally determined η for $K_p > 0.2$ and found that

$$\eta = \left(\frac{K_p}{K_p + 0.7} \right)^2 \quad (5)$$

approximated the data very well. A comparison of Equation (5) with the theoretical prediction of η vs. K_p for potential flow appears in Figure 4.

If it is assumed that:

1. Particles move only with the gas stream and are collected only by liquid droplets
2. The liquid drop diameter is given by the Sauter mean diameter from Nukiyama and Tanasawa (Reference 7),

$$d_d = \frac{58,600}{u_g} \left(\frac{\sigma}{\rho_l} \right)^{1/2} + 597 \frac{\mu_l^{0.45}}{\sqrt{\rho_l \sigma}} \left(1000 \frac{Q_g}{Q_l} \right)^{1.5} \quad (6)$$

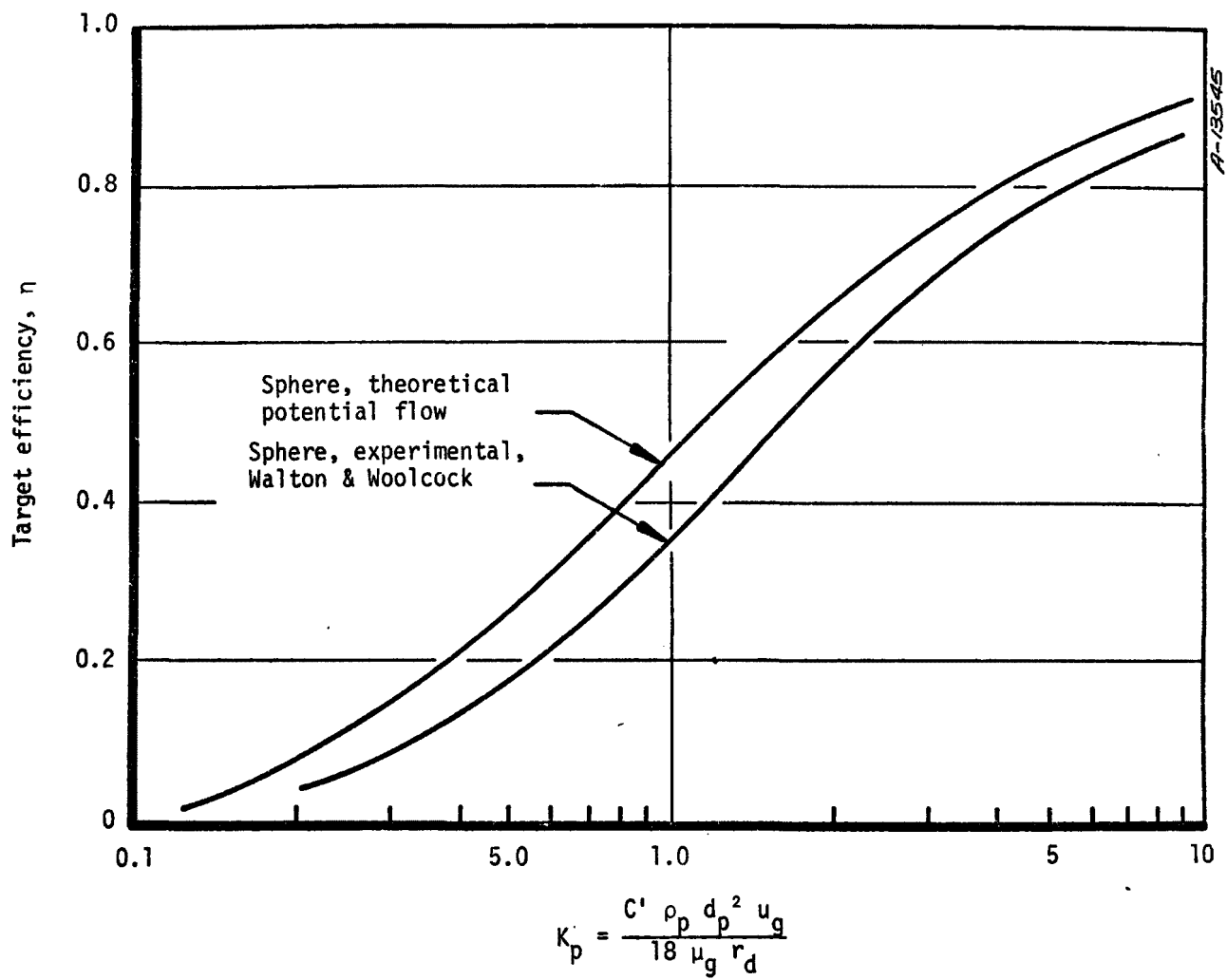


Figure 4. Experimental and calculated target efficiencies for spheres (Reference 4).

where the relative velocity between gas and liquid is assumed to be the gas velocity u_g (cm/sec), σ is the liquid surface tension (dyne/cm), μ_l is the liquid viscosity (poise), ρ_l the liquid density (g/cm³), and Q_l and Q_g are volume flowrates of liquid and gas, respectively. Drop diameter, d_d , is in microns.

3. The liquid droplets' drag coefficient (C_D) is given by Ingebo's (Reference 8) data in the drop Reynolds number (Re_d) range of 6 to 400.

$$C_D = \frac{55}{Re_d} \quad (7)$$

4. Particle collection is by inertial impaction only.
5. Particle concentration is uniform in any plane perpendicular to the gas flow.
6. Liquid is not atomized and distributed over a cross section until

$$u_r = f u_g . \quad (8)$$

where u_r is the relative velocity between gas and liquid and f is an empirical proportionality constant.

then the single particle size penetration (one minus efficiency) may be derived for a venturi scrubber in the following manner (References 4 and 5).

For venturi scrubbers the inertial impaction parameter is defined in terms of the relative velocity between gas and liquid droplets. Thus,

$$K_p = \frac{C' \rho_p d_p^2 u_r}{9 u_g d_d} = \frac{u_r d_{p_a}^2}{9 u_g d_d} \quad (9)$$

where the aerodynamic diameter of a particle is defined as

$$d_{p_a} \equiv (\rho_p C')^{1/2} d_p \quad (10)$$

Taking into account the continuous change in u_r from $f u_g$ to zero, the average target efficiency ($\bar{\eta}$) is expressed in terms of $K_{pt} = K_p(u_g)$ by integrating Equation (5) over K_p to obtain

$$\bar{\eta} = \frac{f K_{pt} + 0.7 - \frac{0.49}{f K_{pt} + 0.7} - 1.4 \ln \left(\frac{f K_{pt} + 0.7}{0.7} \right)}{K_{pt}} \quad (11)$$

The single particle size penetration, P_t , is obtained from a material balance over a differential volume and then integrating. Thus,

$$P_t = \exp \left(- \frac{Q_l \bar{n} Z}{2 d_d Q_g} \right) \quad (12)$$

where Z is the length over which scrubbing occurs. Since particle impaction is operative only when a relative velocity exists between particles (gas) and liquid droplets, Z will be given by the length required to accelerate a liquid droplet to the gas stream velocity. Assuming a constant acceleration given by C_D , then

$$P_t = \exp \left[- \frac{2 Q_l u_g \rho_l d_d}{55 Q_g \mu_g} \bar{n} (K_{pt}, f) \right] \quad (13)$$

where \bar{n} is given by Equation (11). A plot of \bar{n} vs. K_{pt} for $f = 0.25$ is shown in Figure 5.

Standard operating conditions for the Battelle molten scrubber were $Q_l \approx 2$ gpm and $Q_g = 70$ scfm at $750^\circ\text{C} - 800^\circ\text{C}$ and 1 to 2 psig through a 1.5 inch venturi throat (Reference 1). For these values, $Q_l/Q_g = 1.19 \times 10^{-3}$, and throat gas velocity, $u_g = 9310$ cm/sec. Shown in Figure 6 is a plot of predicted particle penetration for $f = 0.25$ as a function of aerodynamic particle size with gas velocity as a parameter. Note that P_t is a strong function of particle size in the 1 to 10 micron range, and that scrubber efficiency ($1 - P_t$) is low for submicron particles. Figure 7 shows P_t as a function of d_{pa} with Q_l/Q_g as a parameter. Note here that P_t is a very strong function of the liquid-to-gas ratio. Again, scrubber efficiency is observed to be quite low for submicron particles at all liquid/gas ratios.

6.2 HIGH TEMPERATURE/PRESSURE EFFECTS

The effects of temperature and pressure on predicted venturi performance are quite important (References 9 and 10). A wealth of experience has been gained with venturi operations at temperatures $< 100^\circ\text{C}$ using water, however, molten scrubbing requires operation at temperatures up to $900^\circ\text{C} - 1000^\circ\text{C}$. Referring to Equations (5), (9), and (13), it is clear that the effects of temperature and pressure on P_t are exhibited largely through changes in μ_g and C' . Strauss and Lancaster (Reference 9) have shown that changes in C' with temperature and pressure are much less significant than changes in the gas viscosity, so only viscosity effects will be considered here.

The viscosity of a gas increases with both temperature and pressure, so the effects of increased temperature or pressure will be to decrease the inertial impaction parameter K_p , and

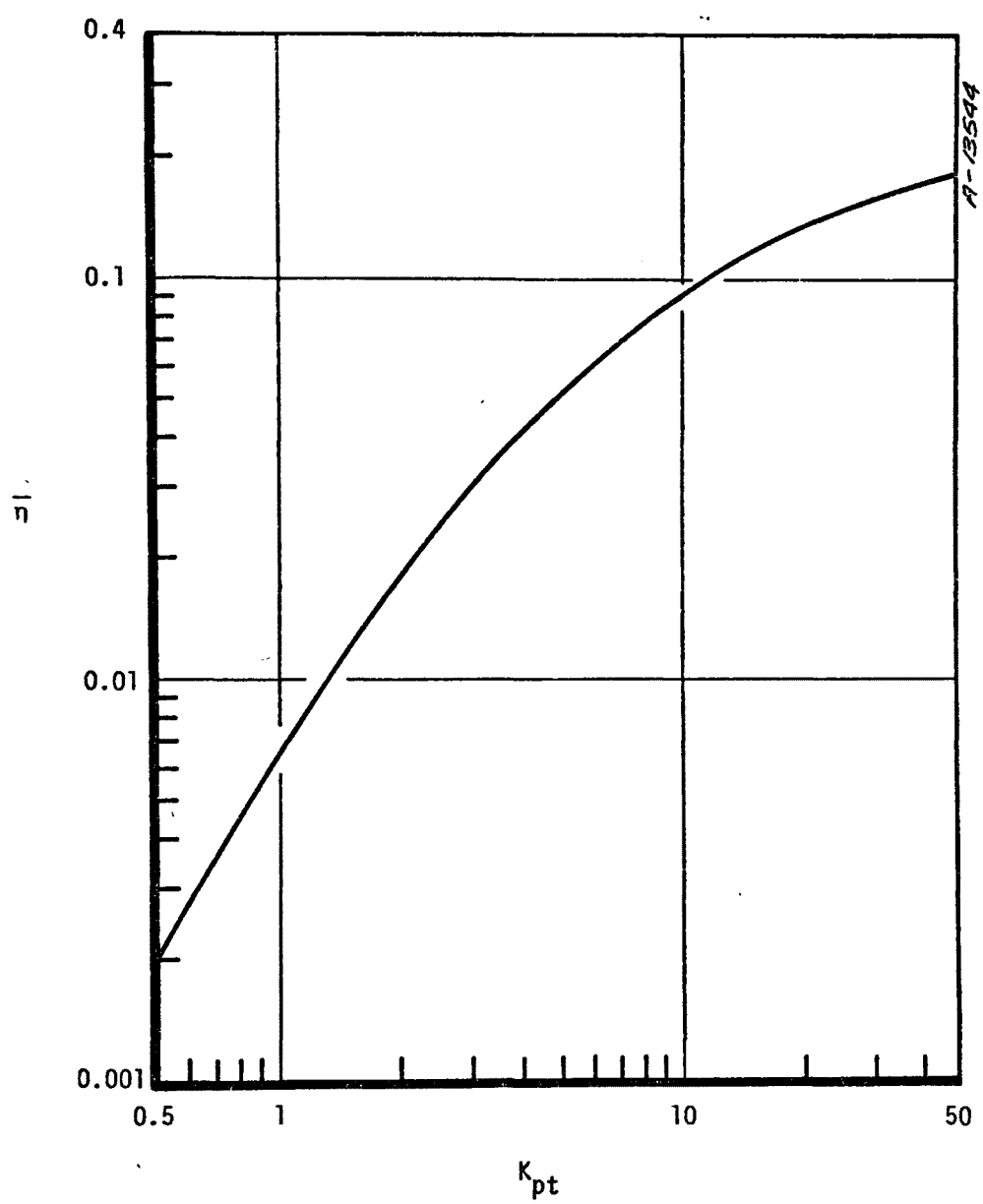


Figure 5. η versus K_{pt} for $f = 0.25$ (Reference 4).

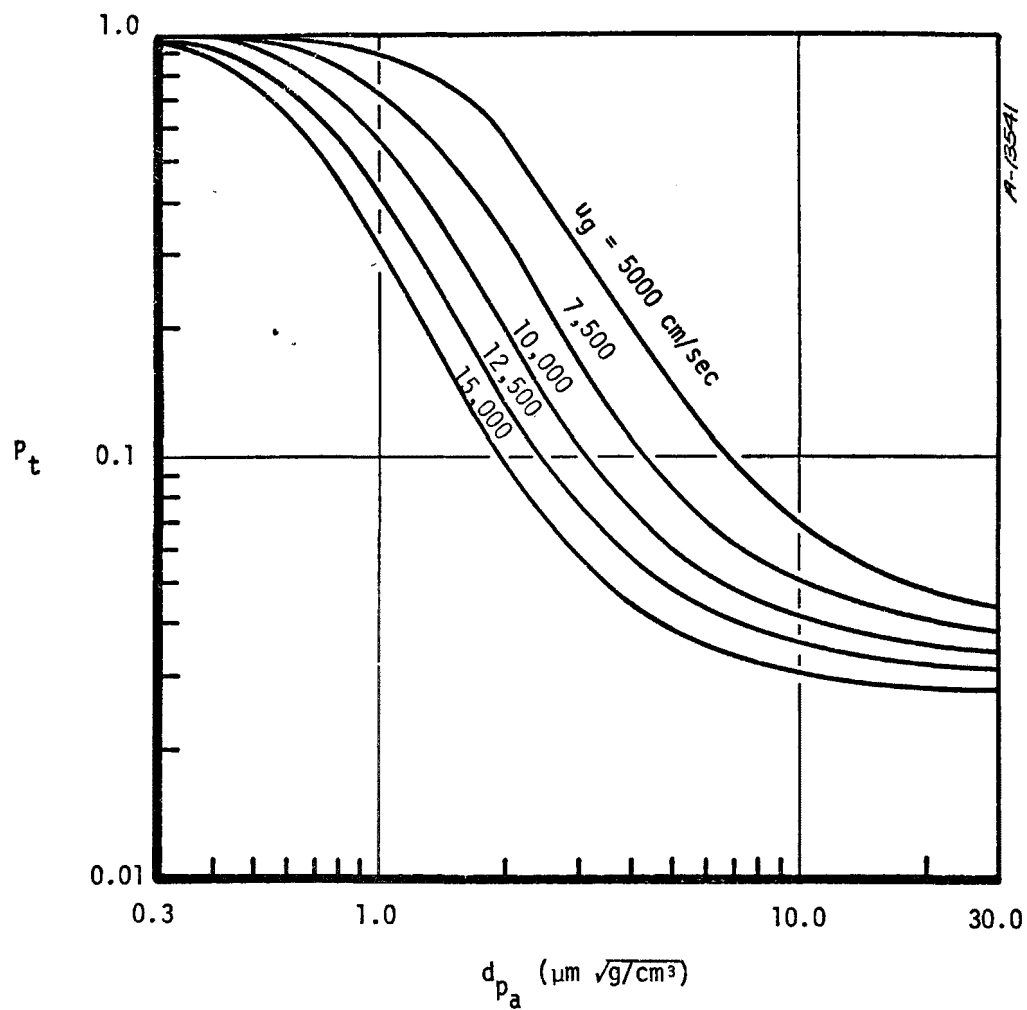


Figure 6. Penetration versus aerodynamic particle diameter for $Q_\ell/Q_g = 1.19 \times 10^{-3}$.

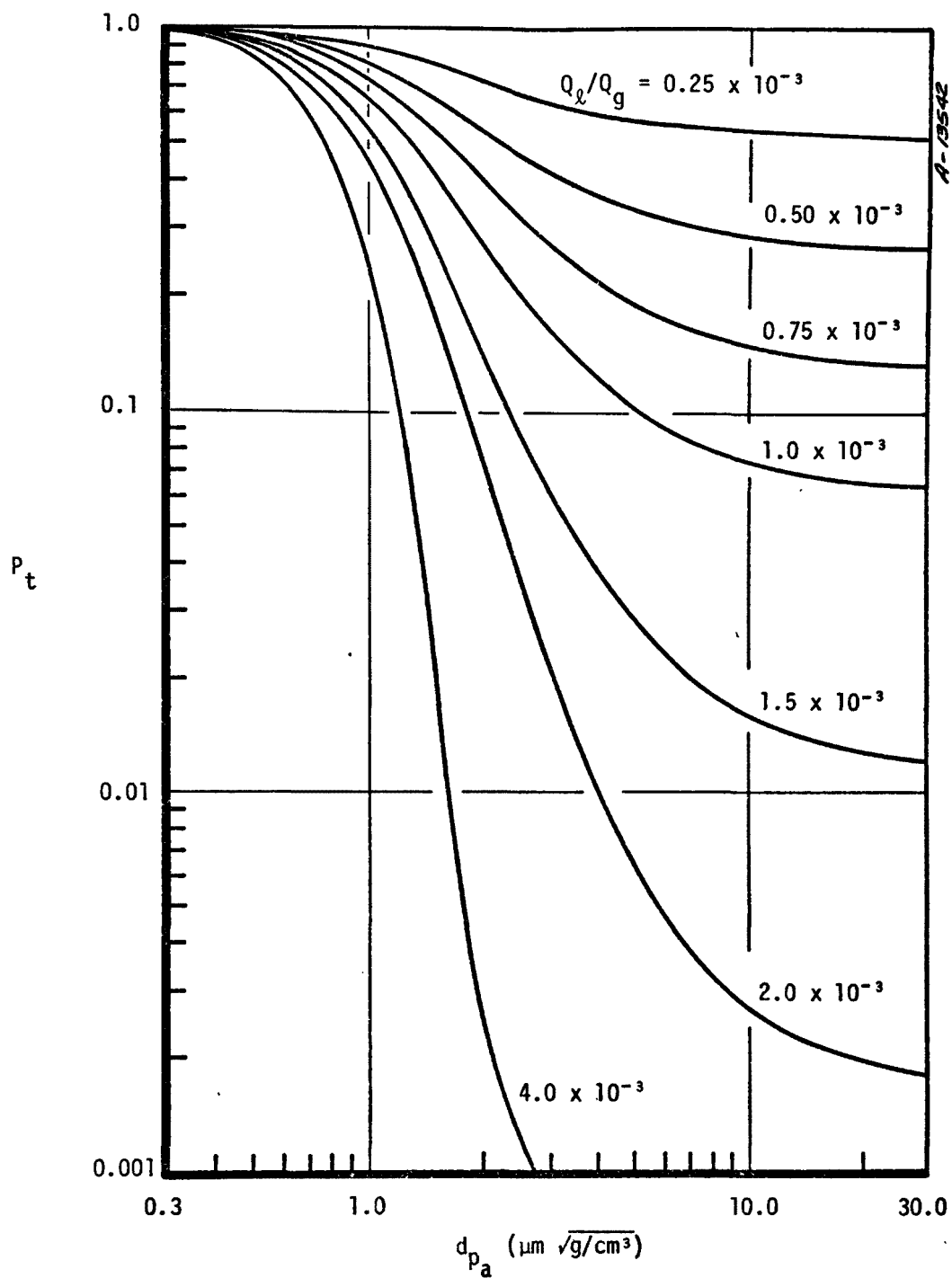


Figure 7. Penetration versus aerodynamic particle diameter for $u_g = 9310$ cm/sec.

hence, decrease the target efficiency η . Consequently, $\bar{\eta}/\mu_g$ decreases and P_t increases with increasing pressure or temperature. The viscosity of air is roughly directly proportional to temperature, however, pressure effects on viscosity are significant only at very high pressures (> 100 atm) or at low temperatures ($< 300^\circ\text{C}$). The effect of temperature on predicted single particle penetration is illustrated in Figure 8 for the Battelle molten scrubber. This figure shows P_t for air as a function of temperature with the aerodynamic particle diameter as a parameter. The associated calculations considered only variations in gas viscosity with temperature; changes in the Cunningham correction factor and in the liquid physical properties were neglected. It is clear from this figure that penetration increases significantly with increasing temperature for all particle sizes.

6.3 PRESSURE DROP AND PARTICULATE COLLECTION EFFICIENCY

Assuming the pressure drop across a venturi scrubber is determined solely by the energy required to accelerate liquid droplets to the gas velocity (wall frictional losses are negligible) and assuming the gas velocity is constant, the pressure drop is given by

$$\Delta P = 1.03 \times 10^{-3} \frac{u_g^2 Q_l}{Q_g} \quad (14)$$

where ΔP is in cm H_2O and u_g is in cm/sec (Reference 4). Since pressure drop is the direct measure of the energy consumed in the scrubbing process, it is clear from Equation (14) that additional energy would be required to increase the collection efficiency for a given particle size and to significantly increase the efficiency of collecting fine particles (≤ 3 microns). This becomes quite evident after examining the curves in Figures 6 and 7. This is discussed further in Subsection 7.5.

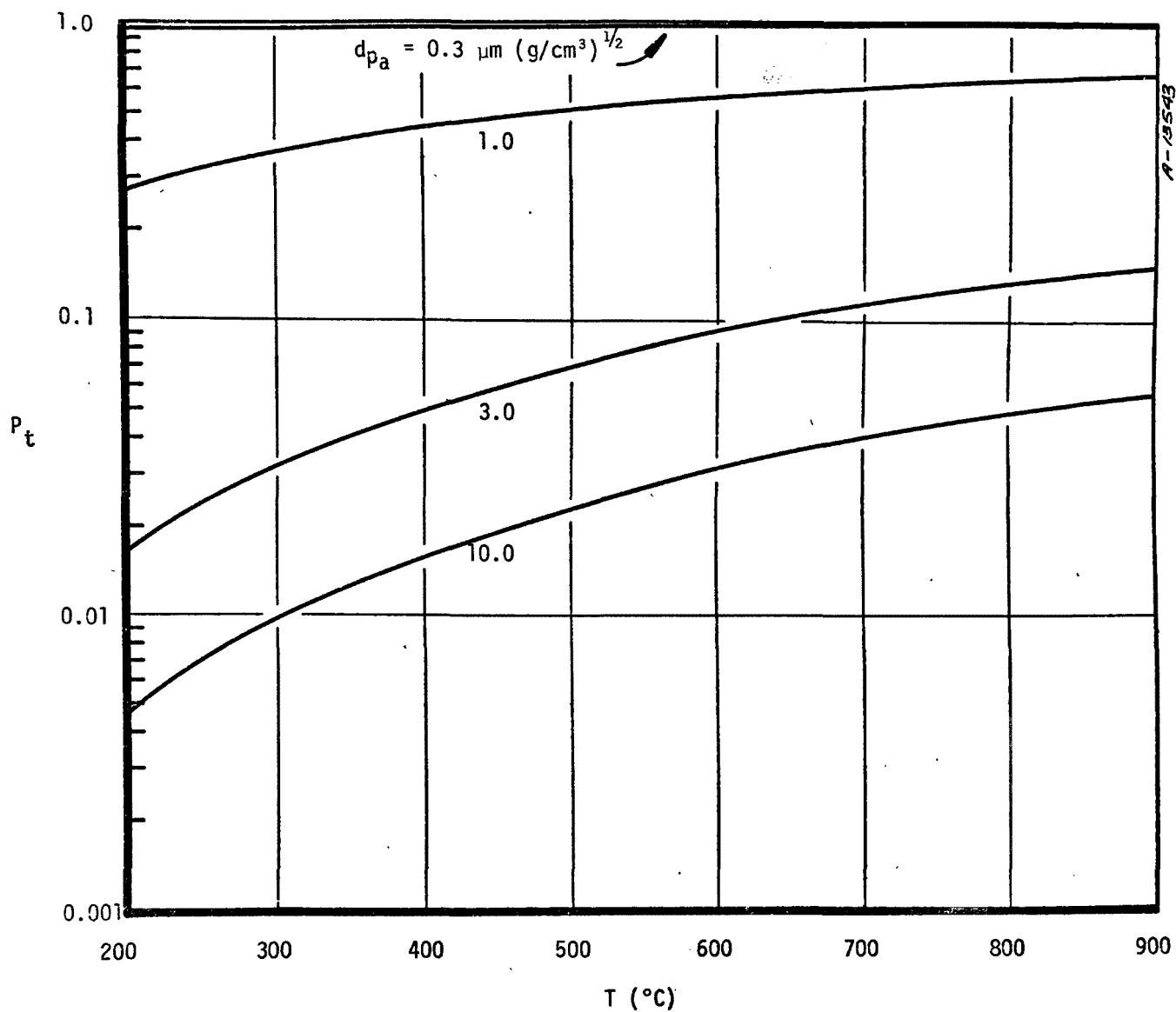


Figure 8. Penetration versus temperature for $Q_\ell/Q_g = 1.19 \times 10^{-3}$; $u_g = 9310 \text{ cm/sec}$.

SECTION 7

PROCESS EVALUATION

Although no molten scrubbing schemes have yet been suggested with the primary goal of particulate removal (Battelle's scrubber treated particulate removal as a secondary consideration and the AI scheme considered particulates to be an unavoidable salt contaminant), this technique may be advanced as such in the future, especially in instances where sulfur compound removal is simultaneously desired. Venturi scrubbers (as well as all scrubbing methods which rely on particle-droplet impaction for particulate collection) require considerable energy consumption for efficient removal of submicron particles. For particles larger than 2 or 3 microns, gas-liquid scrubbing is proven to be an efficient particulate collection method. On the whole, this method remains effective even at the elevated temperatures required for fuel gas scrubbing with molten salts. However, the use of molten salts at elevated temperatures presents a new set of problems which warrants further attention. Some of these points of concern are discussed below.

7.1 MATERIAL CORROSION

Perhaps the greatest drawback to the use of molten salts as a scrubbing liquid lies with the high corrosiveness of the medium. Molten alkali metal carbonates at high temperatures present quite a severe environment to virtually all materials commonly used in the construction of processing equipment. Moreover, the presence of metal sulfides in the gas compounds this problem significantly. Based on corrosion tests, Battelle found aluminized stainless steels to be an acceptable material and reported no problems with severe corrosion during pilot plant operations, however, the pilot plant was in operation for less than 30 hours. Based on longer term dynamic corrosion tests, AI used an austenitic stainless steel as a balance between cost, availability, and corrosion resistance in the construction of their stack gas scrubbing system. However, during operation of their pilot plant AI experienced many problems with stress corrosion cracking of piping and transfer lines, even though the AI process operated at a relatively low (455°C) temperature. AI also considered using aluminized stainless steel but rejected this material because (a) the aluminum is difficult to apply uniformly thus yielding uncoated patches and (b) the aluminizing technique is quite expensive, especially for parts with large exposed

surface areas (Reference 12). At the present time there is no material that is highly resistant to corrosion by hot molten salts and readily amenable to the common process equipment fabrication and assembly techniques at an acceptable cost.

7.2 MIST ELIMINATION

An integral part of any gas-liquid scrubbing scheme is the final separation of entrained liquid from the cleaned gas. This is a special concern when cleaning fuel gas for eventual turbine expansion using molten alkali metals since the allowable concentration of alkali metals in turbine inlet gases is quite low. Both Battelle and AI experienced significant problems with mist elimination (References 11 and 12). The Battelle data on salt carryover was presented and discussed in Section 5. AI reported similar problems with salt crystallization and subsequent salt carryover in wire mesh demisters.

An associated problem is that of salt vaporization and subsequent salt loss in the clean gas. Although the vapor pressures of alkali carbonates are quite low, the vapor pressure of alkali chlorides are significant (e.g., the vapor pressure of NaCl at 800°C is 395 ppm). The use of alkali metal chlorides and other salts with significant vapor pressures must definitely be avoided.

7.3 MOLTEN SALT REACTIONS

Alkali and alkaline earth metal carbonates begin to decompose to their oxides at temperatures around 500°C. Since the oxides of these metals have higher melting points than the carbonates, this decomposition increases the possibility of salt crystallization and line clogging.

Another concern is the possibility of gas reactions with the molten salt liquids. Even though Battelle found no change in the CO, H₂, CO₂, O₂, N₂ or CH₄ composition of fuel gas passing through their molten carbonate scrubber, equilibrium thermodynamics suggest this could be a problem under other test conditions.

7.4 PARTICULATE BUILD-UP IN THE MOLTEN SALT

For a molten particulate scrubbing process to be economically and environmentally feasible, the particulates must be removed from the spent salt melt. AI found flyash sparingly soluble in molten carbonates at 450°C, however, at 800°C Battelle found that 45 to 50 wt% of carbon-free flyash dissolved in the molten carbonates. Certainly the effects of a long-term build-up of these soluble inorganics on the properties of the melt should be studied in greater detail in any future process development work.

Filtering of the used salt melt is required to remove the insoluble components. Other than corrosion, AI reported no problems in filtering insolubles from the molten carbonates using sintered stainless steel filters. However, they concluded that continuous removal of the filter cake was necessary.

7.5 PROCESS APPLICATIONS AND ECONOMICS

Molten scrubbing has been applied to two types of emission problems over the past few years. Atomics International developed the process for removing SO_2 from power plant stack gases, and both IGT and Battelle-Northwest proposed the use of molten scrubbing for H_2S and particulate removal from hot fuel gas from a coal gasifier. We have concluded from the present study that molten scrubbing seems most ideally suited and most appropriately applied to H_2S and particulate removal from hot fuel gases, since an additional step in the regeneration of the molten liquid is required for SO_2 removal.

At present there are no process cost estimates for a molten salt fuel gas scrubber. The only cost estimates available were prepared by AI for their flue gas SO_2 scrubbing concept. These, of course, would not apply to a proposed fuel gas particulate scrubber because of the different gas volumes and salt regeneration requirements. However, for an order of magnitude illustration AI estimated in 1971 (Reference 3) that, for a 800 MW power plant (3.6×10^6 cfm) capital requirements would be \$17/kW and annual operating costs would be 0.95 mills/kWh (3.53 mills/Mcf).

In the absence of other data, the relative operating costs for gas cleaning systems may be estimated from the energy requirements of these systems. For atomized liquid scrubbers such as venturi scrubbers the energy required for particle scrubbing is essentially proportional to the gas pressure drop through the scrubber. Calvert (Reference 13) has developed a relationship between scrubber pressure drop and the aerodynamic particle diameter at which penetration is 50 percent ($d_{p_{a50}}$) for a variety of gas scrubbing schemes. Figure 9 (Reference 14) shows $d_{p_{a50}}$ as a function of gas pressure drop for a sieve plate, impingement plate, packed column, and venturi scrubber. Also shown are the theoretical power requirements for each of these techniques. The plot shows that the energy efficiency of venturi scrubbing is comparable to that of other scrubbing schemes, and is, in fact, more energy efficient for collecting smaller, submicron, particles. The figure indicates that, for collecting particles in the 0.5 to 2.0 micron range, energy requirements for venturi scrubbers are 1 to 10 kW/Mcfm. It is indeed significant that the amount of energy required for scrubbing increases quite strongly as the particle size decreases.

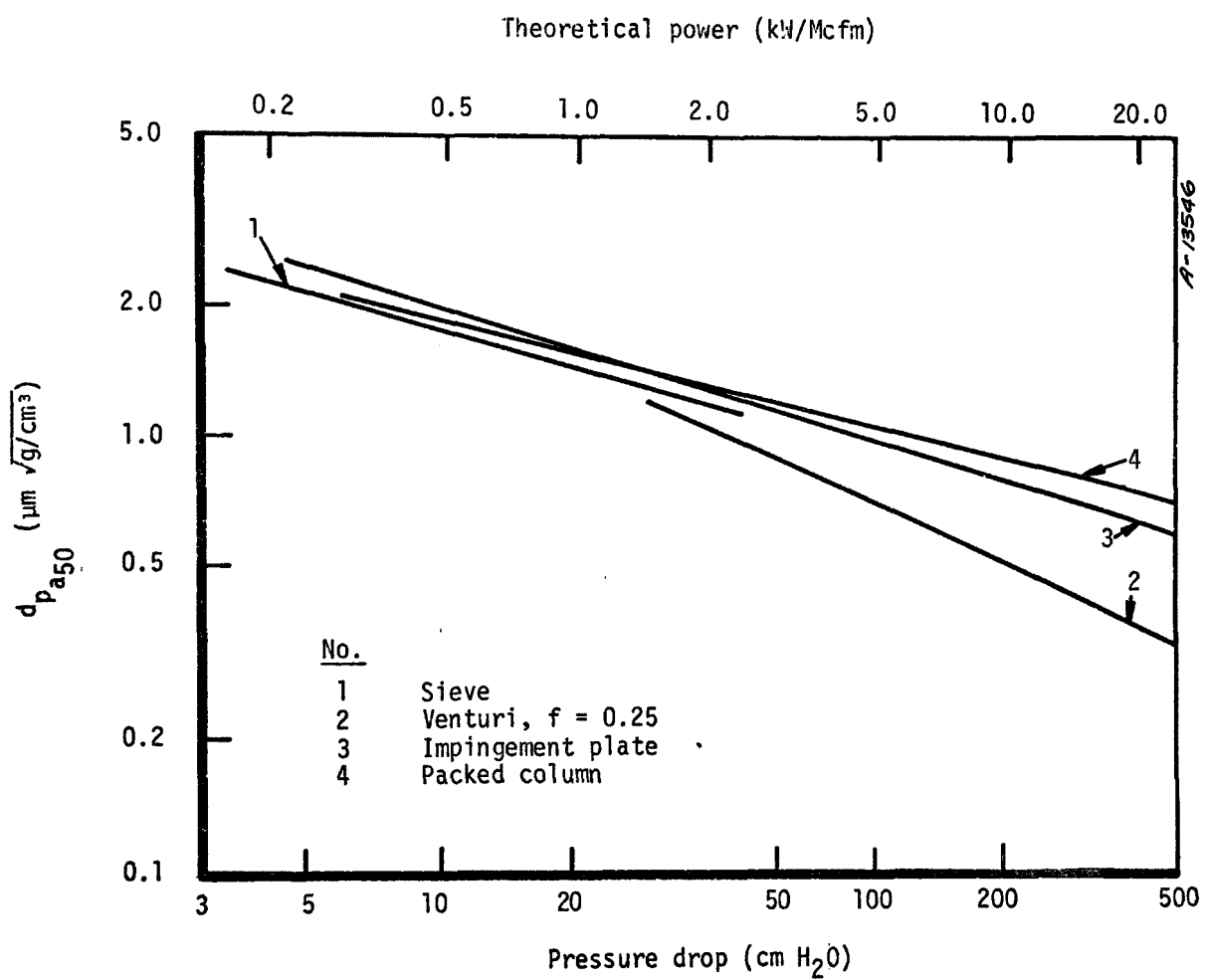


Figure 9. Theoretical power and pressure drop versus aerodynamic cut diameter (Reference 14).

Typical energy usages for other particulate control systems are (Reference 15):

- Cyclones — 1 to 7 kW/Mcfm
- Fabric filters — 3 to 4 kW/Mcfm
- Electrostatic precipitator — 1.5 to 2 kW/Mcfm

Indeed, the energy costs for venturi scrubbing are comparable to those of other gas cleaning methods, and may be lower for smaller particles. Therefore, from an economic standpoint, molten scrubbing appears promising as a high temperature fine particulate clean-up process.

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16. ABSTRACT The report gives results of an evaluation of molten scrubbing for fine particulate control, a concept that study results indicate as seeming to be feasible. Application of the concept to fine particulate clean-up in advanced energy processes seems possible. Molten scrubbing is especially well-suited to processes where simultaneous removal of sulfur compounds is desired. However, before effective molten scrubbing systems can be developed for particulate removal, two important problems need to be solved: (1) finding construction materials at an acceptable cost which can adequately withstand the highly corrosive scrubbing medium presented by hot molten liquids; and (2) improving gas/liquid separation and mist eliminator designs so that liquid carryover satisfies emission standards or gas turbine inlet specifications. Based on the report's observations and on the above conclusions, it appears that considerable development work would be required to investigate the aforementioned problems before a final assessment of the feasibility of this concept could be made.		
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