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An investigation of the Application of Atomisation to Absorption

by

Mavis X. Polk

on assignment from

U. S. Naval Postgraduate School, Monterey, Cal

A Dissertation

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Approved and recommended for acceptance as a dissertation in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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#### Introduction

The absorption of gases from some carrier by a liquid is one of the most widely used mechanisms in industrial operations. This process plays an essential part in the recovery of light oils, in the purification of domestic gases, in humidification, in the recovery of gasoline from natural gas, and in many other phases of industry.

The research toward an understanding of vapor liquid transfer and hence absorption began about 1900, but it has been only in the last twenty-five years that intensive research has been undertaken on the subject. This research has shown that the rate of mass transfer depends on such physical conditions as contact area, temperature, pressure, equilibrium relations, and upon an arbitrary value called the efficiency of the particular type of equipment.

Early absorption processes consisted simply of bubbling a gas through a liquid but this was found to be unsatisfactory except in the case of very soluble gases. With the recognition of the depth of liquid and size of the gas bubbles upon the absorption the trend developed toward developing conditions giving maximum surface area contact. This led directly to the design, development, and use of packed towers with the purpose of the packing being to increase the contact area or as sometimes called "effective area." Slats, broken quartz, screens, brick, berl sadelles, raschig rings, and spheres are a few examples of the numerous types of packings used.

The design of packed columns is based mainly on experience and to a small extent on theory. The use of packing necessitates a large tower of heavy construction. It is obvious that the pumping costs in such units would be high but probably the greatest objection is that such systems tend to plug either by accumulation of tar-like materials or ly resinification of active materials on such large surfaces. This results in reduced capacity and higher pumping cost and the necessity of periodic cleanings. Nevertheless practically all industrial absorption has been carried out in this type equipment until very recent years. At the present time packed columns and spray columns are being used industrially with the packed column still predominating.

The spray system of absorption was developed to try to overcome the objections of the packed columns and still retain the "effective area" concept of absorption. In this system the liquid is dispersed at the top of the column as a spray and during its fall comes into contact with the rising gas. In the most recent design of spray systems the sprays are collected at different points in the column and recirculated in secondary sprays at velocities in excess of the inlet rates. It has been shown commercially that the decrease of the size of the liquid drop and resulting increase of the liquid-gas contact area gives better absorption. A major advantage of the spray system over the packed column is the tremendous decrease in the size and weight of the equipment necessary to handle the same capacity.

The work done in developing absorption equipment has seemed to have as a principle aim the development of greater contact area between liquid and gas as shown by the use of smaller gas bubbles in bubbling gas through

 $\rightarrow$  2.

liquid, the use packed and bubble plate columns, and the use of spray systems of increasingly finer sprays. With this thought in mind it is only logical to consider the use of atomization for absorption.

Atomization breaks up the bulk liquid into a host of tiny droplets, causing a greatly increased surface area of the liquid to be exposed to the gas, and permits of intimate mixing. As an example of the increase of area consider the droplet formation from one liter of liquid. If the liquid is assumed to have a spherical shape it will have a diameter of approximately 12.4 centimeters and a surface area of  $\mu$ 83 square centimeters. Now if this liter of liquid is broken down into equal sized spherical droplets 1.24 microns in diameter, there will be one billion droplets having a total surface area of  $483,000$  square centimeters.

This thesis was undertaken in an effort to initiate work towards the utilization of atomization for absorption processes. Laboratory size absorption equipment was developed in which the carrier gas, containing the gas to be absorbed, was used to atomize the liquid being used as solvent. This constituted a co-current absorption system.

A slightly soluble system, carbon dioxide and water, and a highly soluble system, benzene and kerosene, were selected for experimental work» The carbon dioxide water system was used mainly to evaluate the equipment and to determine the concentrations of absorbed gas that could be obtained by atomization» The results were also compared with published date on absorption of carbon dioxide by water.

The benzene-kerosene system was selected because of its close resemblance of the commercial process of recovery of coke oven gases with wash oil. In the experimental work it was tried to simulate con

ditions as found in industry in order that comparison could be made between published results and experimental results. The effect of drop size and interfacial area, as produced by atomization, upon absorption was studied and the results noted.

#### Historical

The work of this thesis deals with both absorption and atomisation therefore it is felt proper to give background on these two fields. This will be done by giving a brief review of absorption with special emphasis being placed upon the contact area between liquid and gas, and a brief review of the theory of atomization with related work in measurement of drop sizes.

#### Review of Absorption

Gas absorption, in general, involves the treatment of a gaseous mixture with a liquid which dissolves one or more of the gaseous constituents» Usually the gas phase is composed of two or more constituents, one of which is soluble while the others are inert. Most absorption processes, as found industrially, are carried out in towers with the gas and liquid flowing countercurrent to each other. Packed or bubble-cap towers are generally used in order to attain intimate contact between the liquid and gas, although for some purposes spray towers are used.

The design of absorption equipment, the use of different types of packings, and the use of spray towers are all the result of much investigation. The extent of this work may be realized by reference to standard Chemical Engineering textbooks such as those written by Badger and  ${\tt Mccabe}^{\!1\!}$ , Walker, Lewis, McAdams, and Gilliland $^{\!1\!}$ , Sherwood $^2$ , and Brown and Associates<sup>3</sup>, to mention a few as well as to Perry's CHEMICAL

*5»*

 $ENGINEERING HANDBOOK<sup>5</sup>$ . There are many excellent literature surveys of the subject, two of which the reader may be referred to are Theses by Osborne<sup>6</sup> and Grosse.<sup>7</sup>

For the purpose of this thesis only a few of the highlights in the development of gas absorption, especially as attaining to the importance of surface or interfacial area, will be discussed in the text with a complete bibliography being given for any additional reading desired.

Possibly the first person to give much thought to the theoretical and/or actual aspects of absorption was Hurter $8$ . He used three types of absorption systems in his work:

- 1. Simply bubbling gas through a liquid.
- 2. Spray type, or liquid drops falling through an open tower.
- 3. Packed tower, or use of fillers in a tower.

Hurter favored the third method for absorption and hence possibly started the emphasis on packed and bubble plate type towers. It is interesting to note that his work did include the spray type of tower which is gaining popularity at the present time.

Next of interest was the work done by Partington and Parker<sup>9</sup> on tower design and the definition of absorption efficiency. They defined absorption efficiency as the number of pounds of gas absorbed per minute per square foot of liquid surface and showed it to depend upon the partial pressure of the gas, the nature and rate of flow of the absorbing fluid, the temperature, and the turbulence of the liquid and gas.

Donnan and Masson $^{10}$  proposed that absorption was a transfer of matter and presented a mathematical analysis of an absorption process. They pointed out the importance of turbulence, relative motion, and interfacial area between gas and liquid.

In the early 1920's Whitman<sup>11</sup> proposed the two-film theory of gas absorption which states that the absorption rate is dependent on the rate of diffusion of solute through a gas film, an interface, and then a liquid film. He showed that the transfer of solute through the films was proportional to a driving potential, which could be either a pressure or concentration gradient, and called the proportionality constants diffusion coefficients. This theory has been expanded and substantiated by many other workers among whom may be mentioned Davis and Crandall<sup>12</sup>, Chilton and Colburn<sup>13</sup>. Hanks and McAdams<sup>14</sup>, and Arnold<sup>15</sup>. The complete theory is very well outlined by Sherwood<sup>2</sup> in his textbook with equations being given for the cases where:

- 1. Idquid film controls.
- 2. Gaseous film controls,
- 3• Neither film controls.

These equations state that the rate of transfer of solute from the gaseous phase to the liquid phase is directly proportional to the concentration or pressure gradient and interfacial area and indirectly proportional to the temperature and film thickness» From this it may readily be seen that any factor to influence any of the above will influence the rate of absorption.

This theory of the mechanism of absorption is well received at the present time although the supporting equations contain two quantities which are practically impossible to determine. These are interfacial or absorbing area and film thickness. This difficulty is overcome by using a new factor, called absorption coefficient, which combines the two previous quantities. It is necessary to experimentally determine this new factor.

The overall transfer coefficient, Ka, which, in the English system,

has the units pounds per hour per cubic foot per atmosphere driving force contains the overall all transfer coefficient per unit area K and the interfacial area a. These two values are grouped because of the difficulty to determine either separately.

Baker, Chilton and Vernon<sup>16</sup> made a rather intensive study of liquor distribution over various types of packings in order to try to determine the effective wetted area or interfacial area. With packings large in comparison to the tower diameter they found a decided tendency for the liquid to concentrate along the walls and hence small interfacial area as compared to packing area. They found, however, that with the ratio of the diameter of tower to diameter of packing 8 to 1 or greater and with proper feed distribution reasonably uniform distribution could be expected throughout the column. Mayo, Hunter and Nash<sup>17</sup> expanded this study in a very ingenious manner. They used Rashig rings made of double layer paper as packing and circulated water containing red dye through their tower. After running for 10 to *15* minutes the tower was drained and the packing removed and dried. The rings were divided and the dyed area measured for the inner and outer surface. The tower was fitted with a paper liner so that the wetted wall surface might also be measured. The measurement of dyed surface would be a measurement of wetted area. Their work agreed very well with that of Baker, Chilton, and Vernon<sup>16</sup>.

The criteria for a good packing has been the object of much research and this research is well summarized by Badger and McCabe<sup>1</sup>.

Whitman, Long and Want<sup>18</sup> returned interest to the spray type tower in their studies of the rate of absorption for a small drop of water

falling freely through gas. They found the absorption rate coefficient obtained in this manner to be very much greater than those reported for absorption through flat liquid surfaces, from rising gas bubbles, or in wetted wall columns. This has been explained as being due to the comparatively thin film of stagnant gas associated with the drop. The velocity of fall of the drop and any rotary motion during its fall would be factors contributing to break-up of the gas film.

Hixson and Scott<sup>19</sup> probably started the present era investigation of spray towers. They point out the work done on packed and bubble plate columns and the lack of work on the spray system although spray towers were the earliest type of absorption apparatus $\frac{8}{\cdot}$ . In their work they used a very small tower and apparently did not have an adequate method of condensing their spray, as they make the statement that if drops were too small the gas would carry them out of the tower. In the discussion of their work they tabulate the length of liquid jet from the nozzle prior to spray formation. This they then dismiss as being unimportant, giving the reason that the concentration of gas at the nozzle end of the tower is low and that the surface area exposed to the gas is smaller in comparison with the liquid flowing. It has been shown<sup>20</sup> that actually the highest rate of absorption takes place in the vicinity of the nozzle,

Johnstone and Kleinschidt $^{20}$ , in their investigation of absorption of gases in wet cyclone scrubbers, presented information that is pertinent to this thesis. They pointed out that from a theoretical standpoint it would be expected that atomization of the absorbing liquid would provide a large surface area of contact, and that such a surface travelling

through the gas would be ideal for the absorption of very soluble gases. The work $^{18}$  done on measurements of absorption on single droplets has verified this by showing the existence of a high absorption coefficient. They attributed the lack of favor with which spray systems were received industrially to be caused by "rule of thumb" methods of design, without proper appreciation of such factors as size of droplets, their interfacial velocity, the number of droplets per unit volume of tower, and the mechanism of mass transfer in the liquid phase. It was also pointed out by these authors that measurements made on a large spray tower indicated that the rate of absorption was high immediately in front of the nozzles, but that it dropped off rapidly with distance. No measurements of the size distribution of the droplets from the nozzle were made, but they stated that it was obvious that the smallest particles, which were the source of the major portion of the absorption, lost their initial kinetic energy and could not penetrate more than a few feet in the gas. It should be emphasized that the system used for the above observations employed countercurrent flow.

These investigators used two wet cyclone gas scrubbers designed specifically for spray systems as gas absorbers. From their work they developed a theory of absorption by a spray droplet which seems to agree well with other available references. It was also pointed out that drops of the size of 25 microns supplied 50% of the absorption surface in their work although these droplets amounted to less than 0.1% of the total volume of liquid. They feel that any decrease in drop size increases the absorption in two ways, the increase of the number of drops and hence area of the liquid and the decrease of velocity giving longer contact time.

Johnstone and Silco $x^{21}$  expand the previous work<sup>20</sup> done on cyclone spray towers using laboratory size equipment. They assert that the quantity of gas absorbed by a liquid drop depends in part on the area of the drop, the velocity relative to the gas, and the time of travel. Their system was not countercurrent but across current in nature and mass transfer corresponded to only slightly more than one theoretical plate in a single unit. The number of transfer units available in a short height was found to be quite large and almost complete absorption of a solute gas could be obtained when the proper solvent was selected.

#### Theory of Atomization

Atomization can be defined as the process of subdividing a liquid into droplets of such size that there can be no further size decrease by physical means. This definition, while it may be accurate, does not quite picture atomization as it is recognized in industry. For the purpose of this thesis, atomization will be defined as a process which produces an extremely high ratio of surface to mass of a liquid. It is felt that this latter definition more clearly pictures atomization as recognized by most scientist and engineers. Although spray might be defined by a very similar definition, atomization is usually pictured as producing much smaller droplets and hence more surface area per unit mass.

Rayleigh<sup> $\mathrm{s}^{30}$  observation of the collapse of a round cylindrical</sup> liquid column under influence of surface tension was apparently the first recorded work done on drop formation or atomization. He found that the column is unstable if its length exceeds its circumference.

 $\text{Castleman}^{31}$  described atomization of liquids in air streams as follows : "A portion of the large mass is caught up (say at a point where the surface is ruffled) by the air stream and, being anchored at the other end, is drawn out into a fine ligament. This ligament is quickly cut off by the rapid growth of a dent in its surface, and the detached mass, being quite small, is swiftly drawn into a spherical drop." He stated that, within limits, higher air speeds caused finer ligaments having shorter duration and gave smaller drops. Also he made the observation that a liquid forced under high pressure into still air was atomized. He concluded that this was the reverse of air-stream atomization, but very similar in that the fast moving liquid would lose filaments to the still air in the same manner as the fast moving air drags ligaments from quiescent liquid.

Joyce<sup>32</sup> gives a good picture of the process of atomization. Although his article is devoid of references, it is apparent that his description is based on the observations of earlier investigators. The influence of such investigators as Rayleigh<sup>30</sup>, Castleman<sup>31</sup>, and Schweitzer<sup>33</sup> can be detected. Briefly his description is as follows: To atomize a liquid, force is applied to the liquid to set it in motion and it is constrained in some manner so that it will emerge as an attenuated film. This film is then disrupted into shreds or filaments by the persisting balance of energy in the liquid derived from the force which promoted the initial flow or in some cases by additional applied force. The shreds or filaments break up into smaller fragments. The surface tension of the liquid causes these small fragments to rapidly assume spherical shape.

The atomization of a liquid therefore involves enforced attentuation,

ligament or filament formation, and disruption. The disruption of the filaments, by the action of the surface tension, follows the  $\pi$  d rule. The filaments may vary greatly in diameter and therefore the resulting droplets will also vary in size correspondingly, thus producing a spray composed of droplets of a wide range of size.

It must not be assumed that there is a clear line of demarcation between the attenuation and filament formation. The formation of the filaments is due to the force causing attenuation and to surface tension and there is an interplay of these forces during atomization.

Nukiyama and Tanasawa<sup>38</sup> studied the atomization patterns of liquids by taking stereoscopic instantaneous photograms of liquid jets atomized in high speed air streams. At low air velocities they found that the relative motion between air and liquid produced bead-like swellings and contractions which increased in amplitude until the liquid jet finally breaks up and forms individual drops. If the velocity of the air is increased, a fluttering action occurs in the jet which forms a shape like that of a twisted ribbon. A portion of the ribbon is caught up by the air stream and is drawn out into a fine ligament. This ligament is in turn quickly cut off and the separated mass swiftly forms a spherical drop. A further increase in the air velocity causes the horizontal part of the twisted ribbon to flatten and form a cobweblike film. This film is so thin that it tears apart and diffuses into microdroplets. Upon still further increasing the air velocity the number of films gradually increase until the twisted ribbon disappears and only a large number of the cobweb-like films are seen.

Three common methods of atomizing liquids are centrifugal disks,

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pressure nozzles, and two-fluid nozzles<sup>5</sup>. The centrifugal disks atomize by means of extending the liquid into a fine sheet and discharging it at high speed from the periphery of the rapidly rotating disk. The degree of atomization, as a function of disk speed and diameter, is believed to be effected by the peripheral speed as opposed to the angular speed. Pressure nozzles bring about atomization by forcing liquid under high pressure and with a high degree of spin through a small orifice. The characteristics of the spray or fog is determined by the pressure and orifice size» Two fluid nozzles, characterized by paint spray guns, perfume atomizers, etc., bring about atomization by the impingement of a stream of gas on a slowly moving stream of liquid. They operate at relatively low pressures.

The distribution of drops in a sprayed jet, the effect of the quantity of air, and the effect of the relative velocity between air and liquid in the case where water was atomized by air in a convergent nozzle were studied by Nukiyama and Tanasawa $^{34}$ . They concluded that:

- 1. With the exception of the extremities, the mean drop diameters are uniform in the sprayed jet and that atomization was finished completely at the throat of the nozzle.
- 2. With the relative velocity constant, the ratio of liquid to air controls the drop size and not the velocity of the water, the size of the water orifice, or the nature of flow of the water. When the ratio of volume of air to volume of water exceeds  $5,000$  the drop size remains constant.
- 3» When the ratio of volume of air to volume of water is greater than  $5,000$ , the drop size is inversely proportional to the relative velocity.

In another paper<sup>35</sup> these investigators studied the effect of the above properties, as well as the effect of different size air and water orifices, on sharp edged and cylindrical nozzles. They concluded that the effects were the same as found for the convergent nozzle and that varying the size of the air and water orifices did not change drop size if the relative velocity and liquid-gas ratio remained constant.

The effect of viscosity, density, surface tension, relative velocity, and liquid-gas ratio may be expressed by the empirical equation  $37$ 

$$
d\mathbf{o} = 585 \sqrt{\frac{\sigma}{\rho}} + 585 \left(\frac{\mathcal{A}}{\sqrt{5\rho}}\right)^{0.45} \cdot (1,000 \frac{q}{q_a})^{1.5} \qquad 1
$$

where  $\omega =$  mean diameter of drops in microns.

- $v =$  relative velocity between liquid and air stream in meters per sec.
- $\rho$  = density of the liquid in grams per cubic centimeter.
- $\sigma$  = surface tension of the liquid in dyne per centimeter.
- $\mathcal{U}$  = viscosity of the liquid in dyne seconds per square centimeter.
- $Q<sub>a</sub>$  = quantity of air in cubic centimeters per sec.
- $Q_L$  = quantity of liquid in cubic centimeters per sec.

This equation is valid under the following conditions:

- 1. Air is used as atomizing gas.
- 2. Density between  $0.8$  and  $1.2$ .
- 3. Surface tension between 30 and 73.
- 4. Viscosity between 0.01 and 0.30.

The effect of viscosity, surface tension, and density, as well as velocity and quantity ratios, may be deduced from the equation. When the ratio of air to liquid is large the second term will be small and the first term will govern the drop size, thus showing viscosity to be of minor importance. When the air to liquid ratio is small the second term will be large and thus govern the drop size. In this case the surface tension would have only slight influence. The density effects the size of drops

inversely at all times.

The dimensions of the above equation are inconsistent unless the authors had assigned dimensions to the constants, nevertheless its validity has been substantiated by the "work of many other investigators as will be explained below.

Drop, size determination has been the object of much more research than the theory of atomization and a few methods will be mentioned.

The most recent method of determining drop sizes seems to be that proposed by Geist, York, and Brown<sup>29</sup> in which they use an electronic spray analyzer. This consists of a charged wire, inserted into a moving suspension, connected to electronic circuits which count the electrical pulses created by the interception of drops and wire. The size of the electrical pulse is a function of the size of the drops. The investigators<sup>29</sup> are doing further research on this method, but at the time of their report it was not suitable for small droplets.

Shallow sampling cups, coated with non-wetted material and having a flat transparent bottom, are filled with a liquid that is non-miscible with, and having a density slightly less than, the sprayed fluid. The drops of sprayed fluid will settle through the liquid in the cup and may be photographed<sup>39</sup>.

Direct photography of the drops in the sprayed jet by actual insertion of the camera into the spray has been tried by many different investigators and is fully explained by York $\overline{^{40}}$ . This method avoids physical sampling.

The theory of light scattering<sup> $\mu$ </sup> has been used for measuring particle size. This is done by measuring the intensity of light scattered at right angle to a beam passing through the spray and calibrating this

against the number of drops. The decrease in light transmission through the spray is measured and calibrated to give the number of drops of given sizes.

Drop sizes are determined by a series of four oil-coated slides in the cascade impactor  $\frac{1}{2}$  method of analysis. A sample of the spray passes through a small opening and impinges on a slide placed perpendicular to the opening. Some of the spray is not collected on the slide and passes through another opening and impinges on a second slide and so on until drops have been deposited on all slides. The theory is that the larger drops deposit on the first slide and decreasing size drops on the next three slides.

The wax method of drop size determination as described by  $Jogce<sup>32</sup>$ consist of atomizing liquid wax into a flowing water bath, then collecting and sizing the solid wax particles.

Microscopic slides or plates are used to determine drop size and the technique varies greatly. For example, the method used by Lewis, Edwards, Goglia, Rice and Smith $43$  consisted of simply waving a microscopic slide through the spray at a velocity of several feet per second and then counting the drops. The method used by Nukiyama and Tanasawa<sup>34</sup>. as described below, was somewhat more complicated.

It appears, from the literature, that the work done by Nukiyama and Tanasawa<sup>34,37</sup> is the most reliable to date. They measured drop size in gas stream atomization, both with convergent nozzles and sharp edged orifices. Air was used to atomize water, as well as water glycerine, water-methanol, water-ethanol, and water-ethanol-Glycerine solutions. The drops were collected on a specially prepared oil slide by means of

a shutter arrangement and photographed. The number of drops were counted and the mean diameter, do, calculated from  $34$ 

$$
\text{do } = \sum x^3 \quad \text{do } \Delta n / \sum x^2 \quad \text{do } \Delta n
$$

where  $\Delta$  n represents the number of drops, the diameters of which lie between x and  $\triangle$  x. It was found that the mean diameter of the drops were uniform everywhere in the sprayed jet except in the extremities.

The diameters of the water orifices were varied in these investigations from 0.2 to 1 mm and those of the air orifices from 1 to 5 mm. The air velocity, water velocity, and air-water volume ratio were also varied as noted above in the explanation of equation 1. This empirical equation has been well substantiated by later investigators and as the equation is based on all other work performed by Nukiyama and Tanasawa $34*38$ , It would seem in turn to substantiate all of their work.

These investigators also presented an empirical equation for determining the mean drop diameter of gasoline, ethyl, alcohol, and heavy oil<sup>37</sup>, as well as an empirical equation for particle size distribution. The latter equation, which is similar to the Maxwell equation for velocity distribution in a perfect gas, contains four constants that are dependent upon the type nozzle and conditions of atomization. Lewis, Edwards, Moglia, Rice, and Smith $^{43}$  comment on this equation and derive several different sets of constants.

The latter investigators<sup>43</sup> conducted a very detailed investigation of atomization and drop size determination for the "Office of Scientific Research and Development." They concluded, from a literature survey, that the best correlation of available data was the work by Nukiyama and Tanasawa<sup>34:38</sup>, and therefore their efforts were to test the validity of the Japanese equations. Besides their own experimental data and

results, these investigators subject data of Houghton, Sauter, and Lee to analysis by equation 1. They find rather good agreement and point out what they believe to be the causes for descrepencies. For example, Sauter, using an air Venturi, assumed all drops to be spherical and distributed uniformally across the spray. Both of these assumptions have been shown to be erroneous. Houghton's measured drop sizes were smaller than those predicted by the equation but this was thought to be an error due to sampling technique. It was pointed out that failure to measure just one large drop among as many as a 1,000 small drops would seriously effect the mean drop diameter. The data of Lee's experiments were found to agree well with calculated values from equation 1.

It was specifically pointed out<sup>13</sup> that the work done by Nukiyama and Tanasawa appeared valid not only for small gas atomizing nozzles, but also for other types such as pressure nozzles and large Venturi nozzles.

 $\text{York}^{40}$ , in his work on drop size determination, used a standard paint spray nozzle which is much different from the nozzles used by Nukiyama and Tanasawa, but his data showed a definite tendency to extend the curves presented by the Japanese and thus further substantiate their work.

An excellent literature survey of atomization and spray is DeJuhasz's $^{\frac{l_1 l_2}{l_1 l_2}}$ "Bibliography on Sprays," and supplement number one to this Bibliography by DeJuhasz and Meyer.  $45$ 

Bevans<sup>46</sup>, Pierce<sup>47</sup>, and Limper<sup>48</sup>, in individual reports, give evidence supporting the validity of the empirical equations proposed by Nukiyama and Tanasawa.

#### Nozzle Design

One of the first, and perhaps most important, things considered for this thesis work was the design of a suitable nozzle. A search of the literature and of spray equipment catalogs yielded numerous types of nozzles for consideration. These included such types as the standard paint sprayers, perfume atomizers, pressure nozzles, high-speed rotating disks, and two fluid type nozzles to mention only a few. Although it has never been clearly defined as to where this transition is that enables one to differentiate between sprayed liquid and atomized liquid, the two fluid type .nozzles seemed capable of giving drops of such a size that no question would arise concerning this point. Nukiyama and Tanasawa $^{37}$ , using this type nozzle, were able to obtain, and measure, drops having a mean diameter of less than 20 microns.

It was decided to try the two fluid nozzle for the present investigation and a nozzle as shown in Figure 1 was designed. This nozzle is much like those used by Nukiyama and Tanasawa<sup>37</sup>, in the work on drop size determination. Fundamentally the nozzle consists of a gas inlet, a liquid inlet, a liquid orifice, a mixing chamber, and a gas-liquid mixture outlet orifice. Actually the design and machining of the nozzle was more critical than the above would suggest. The liquid inlet entered the rear of the nozzle and terminated 0.39 inches from the inside wall of the nozzle's frontspiece. A 0.0135 inch orifice, exactly centered, was drilled in the liquid inlet. The frontspiece was drilled, exactly center, with a





0.150 drill and this hole was machined to taper outward at an angle of 60 degrees, thus giving a 0.150 inch sharp edged orifice. The criterion for selection of the size of the liquid nozzle was the smallest that would allow maximum anticipated flow of liquid. No special criterion was used for selection of the size of the outlet orifice except that it has roughly about eleven times the diameter of the liquid orifice.

The location and size of the gas inlet did not appear critical other than it be placed behind the outlet of the liquid orifice in order not to impinge upon the liquid stream, and be large enough to accommodate anticipated flow. It was located  $3/\mu$ -inch behind the liquid inlet and was a standard  $1/l$ -inch brass tube fitting. Likewise the size of the mixing chamber apparently was not critical as the liquid rate, gas rate, pressure, and size of the outlet orifice would control the degree of atomization.

The nozzle as described above proved to be satisfactory, but in order to note the effect of different orifice sizes another nozzle was designed as shown in Figure 2. This new nozzle was basically the same as the first nozzle, but was so designed that orifices of different sizes could be installed. In other words, it was what could be called "convertible" as opposed to "permanent. " The liquid inlet of the second nozzle was fitted with a cap with a  $1/l$ -inch holed drilled in the center. Liquid orifices were drilled in small plates which would fit inside of the cap. The end of the liquid outlet tube and inside of the cap were machined so that, when assembled, the cap would seal the plate containing the liquid orifice firmly against the liquid outlet tube. Three such orifice plates were constructed, the sizes of the orifices being 0.0146, 0.0256, and 0.033 inches with maximum capacities being 160, 385, and 650 cc per min. respectively

# FIGURE 2 CONVERTIBLE TYPE GAS LIQUID ATOMIZING NOZZLE



under the conditions used.

The discharge end of the nozzle was also modified to enable the interchange of outlet orifices. This was done by threading the outside of the nozzle to accommodate a cap, drilling the cap, and fitting the cap with orifice plates in the same manner as was done for the liquid outlet. Three plates were machined to fit into the cap, drilled, and tapered, in the manner described for the first nozzle, to give orifices of 0.150, 0.120, and 0.073 inches. Dimensions of the body, mixing chamber, gas inlet, and liquid inlet as well as location of gas and liquid inlets were retained the same as in the original nozzle described above and shown in Figure 1.

Absorption of Carbon Dioxide with Water

The carbon dioxide-water system was used for an initial investigation of the effect of atomization on absorption. This system was used mainly to evaluate the nozzle design and to determine the nearness of approach to handbook<sup>22</sup>, equilibrium values obtainable by means of atomization. Some comparison of the actual absorption was made with published results for packed columns, but these were not as conclusive as desired. The published results were not of recent date and also their validity could not be substantiated.

#### Experimental Apparatus

The experimental apparatus used for the first part of the investigation on the carbon dioxide-water system is shown diagrammatically in Figure 3. This apparatus was used for all runs shown in Tables  $1-6$ . The carbon dioxide supply was a fifty-pound cylinder of compressed gas. The gas line from the cylinder to the junction with the air line contained a standard carbon dioxide reducing valve and a calibrated gas rotameter for controlling rate and pressure and measuring the volume of the gas. The air supply was an Ingersoll Rand air compressor. The line from the compressor to the junction of the air and carbon dioxide lines contained a needle throttling valve, a pressure gauge, and a gas rotameter placed in the order mentioned. Upon leaving their respective rotameters, the air and carbon dioxide lines were joined and a single line was led to the nozzle.

**V-l,W2.V-3,V-4y-5- CONTROL VALVES**

**P-l, <sup>P</sup>t <sup>2</sup> — PRESSURE POINTS**

**<sup>T</sup>-<sup>1</sup> ----- TEMPERATURE POIMT**



FIGURE 3

ORIGINAL CO<sub>2</sub> APPARATUS

The water supply was taken from the city water main and filtered through a glass wool filter which was placed in the supply line. The water rate was measured by a liquid rotameter and controlled by means of two control valves, placed before and after the rotameter in the line. A bleed-off valve was placed in the water line for the purpose of purging and of blowing out the nozzle. The water was introduced into the nozzle from the back, Figure 1, and mixed with the gas mixture in the mixing chamber. The mixture was discharged into the collector as a fog.

Several different sizes and types of collectors were tried as listed in Tables 3 and 4 with a 2  $3/\mu \times 48$  inch pipe being selected. The collector was placed in a vertical position, sealed to the nozzle at the nozzle end, and open to the atmosphere at the discharge end.

The atomizing nozzle used was a sharp-edged gas atomizing nozzle shown in Figure 1 with a 0.0135 inch water orifice and a 0.150 inch gaswater mixture orifice.

Several faults were found with the apparatus as described above. The two most outstanding were insufficient air capacity and unsatisfactory results due to the use of city water<sup>23</sup>. In order to correct these faults, along with several minor ones, the apparatus was modified as shown in Figure  $l_+$ . The Ingersoll Rand air compressor was replaced by a Worthington compressor having a capacity of 17 c.f.m., and a source of distilled water was added. This source consisted of a three and a half gallon pressure tank, in principal an acid egg, which was pressurized by means of the Ingersoll Rand compressor. Also the collector was changed to a  $5$  1/2 x 36 inch glass column to enable observation of the fog pattern.


APPARATUS 4 FIGURE MODIFIED CO2

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The collector was now sealed at both ends and discharged into a Cotrell precipitator. Some runs, as noted in the tables, were made without the collector, but with the nozzle discharging directly into the Cotrell,

The Cotrell, Figure  $5$ , was made of a 1 3/8 inch inside diameter copper tube  $2\mu$  inches long with a platinum wire centered in the tube as an electrode. A potential of  $10,000$  to  $15,000$  volts was supplied from a high voltage power pack, The vapor entered the side' of the tube near the bottom and the condensate was discharged from the bottom by gravity with the gases being swept out of the top of the tube.

The nozzle was also modified as shown in Figure 2 and described under "Design of Nozzle,"

# Experimental Procedure

The experimental procedure differed somewhat during the course of the investigation. For all runs shown in Tables  $1-6$  the separate streams of carbon dioxide and air were metered by means of gas rotameters, then mixed and fed into the atomizing nozzle, Rates of both streams were maintained constant by means of manually operated control valves.

Tap water from the Bethlehem city water main was introduced to the nozzle through a liquid rotameter with the rate being maintained constant by manually operated control valves. The temperature of the water was that of the underground lines and therefore varied somewhat from day to day.

The water entered the atomizing nozzle from the back and then into the chamber of the nozzle through a 0,0135 inch orifice, The gas mixture entered the chamber of the nozzle from the side, picked up the water and

FIGURE 5-COTRELL



the mixture was then dispersed into a collector through a 0.150 inch orifice as a fog. Pipes of various sizes and lengths were tried as collectors so as to note the effect that each had on the fog pattern with a  $5 \frac{1}{2}$  inch x 3 foot glass column being selected. The end of the collector through which the nozzle fitted was sealed around the nozzle to prevent air from being drawn in by the fog leaving the nozzle. Samples for analysis were taken from the discharge end of the collector which was left open to allow the system to remain at atmospheric pressure. The temperature of the mixture was taken in the collector.

Runs were made in the following manner: The inlet gas composition and the liquid and gas rates were fixed. The cooling effect due to the throttling of the carbon dioxide caused fluctuations in the rate of  $f$ low<sup>23</sup>. These fluctuations were eliminated by allowing the carbon dioxide to flow long enough to maintain essentially constant temperature and hence constant flow rates through the throttling valve before taking readings. After rates had become steady, liquid samples were taken from the end of the collector and analyzed. Readings of rates of flow, temperature, and pressure were taken at the time of collecting the sample.

The procedure used for the remainder of the investigation of the carbon dioxide-water system differed only slightly from that as described above. Air for all runs listed in Tables  $7-10$  was supplied by a larger compressor, but it was treated in the same manner as before. Distilled water was used instead of water from the city main and it was introduced into the system in the same manner as described above. The only difference was the source, which was now a pressurized tank. The major difference was the collection of the sample. Where before the sample was collected

at the open end of the collector, it was now collected from a discharge line on the bottom of the Cotrell precipitator. The end of the collector was sealed, with an outlet leading the gas to the Cotrell» The procedure for manipulation of controls, readings, and analysis remained the same.

For all runs listed in Table 10, discharge from the nozzle was not dispersed into a collector and thence into the Cotrell, but instead was put directly into the Cotrell with the sample being collected from the bottom of the Cotrell; manipulation of controls, readings, and analysis remained the same.

## Method of Analysis

Since the composition of the two inlet streams were determined by metering known quantities to the system, only the outlet streams remained to be determined» It was decided to analyze the outlet liquid stream and to determine the outlet gas stream by method of difference.

The first method of analysis of the liquid stream consisted of straight titration of the water-carbon dioxide solution with a standardized base» Ten ml» samples were collected from the collector in a pipette and introduced into a flask. The  $H_2CO_3$  of the solution was neutralized by titrating directly with  $0.0527N$  Ba(OH)<sub>2</sub>. All runs of Tables 1-4 were analyzed in this manner.

It was noted that reproducible results could not be obtained from the same sample if only a few minutes were allowed to elapse between the original analysis and a check analysis. Presumably this was due to carbon dioxide being lost to the surrounding air, and, if this were true, it was logical to assume that some of the gas would escape during sampling

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and titration, as well as upon sitting.

A second method of analysis<sup>23</sup> was tried in an attempt to decrease the possibility of escape of the gas. In this method the sample was collected, as before, in a 10 ml. pipette, but was immediately neutralized by introducing it into a flask containing an excess of  $0.0527N$  Ba(OH)<sub>2</sub>. The end of the pipette was placed beneath the surface of the  $Ba(OH)$ <sup>2</sup> when transferring the sample. The excess Ba(OH)<sub>2</sub> was neutralized by titrating with standardized HC1. Phenolphthalein was used as the indicator in all titrations. Runs shown in Table 5 were made to check these two methods of analysis. From the results it was apparent that carbon dioxide had been escaping in the first method of analysis so the second method was adopted for the remaining experimental work.

To insure accurate analysis of the dissolved carbon dioxide, and to preclude the error due to the acidic or basic quality of the water, blanks were run on each batch of water used. This was accomplished by titrating samples of the water obtained from the tank prior to starting actual runs, and making necessary corrections in the analysis of the carbon dioxide-water sample.

### Absorption of Benzene with Kerosene

A study of the effect of atomization on absorption was made on the benzene-kerosene system» It was necessary to produce an air-benzene mixture of known composition and to use this mixture as the atomizing gas. The method adopted, as described below, for this study contained many variables and efforts were made to note the effect of all the obvious ones.

## Experimental Apparatus

The experimental apparatus used for the investigation of absorbing benzene vapors in kerosene is shown diagrammatically in Figure 6. A three and a half gallon tank, pressurized by an Ingersoll Rand air compressor was used to supply kerosene to the system. The air line, containing a diaphram reducing valve for maintaining constant pressure on the tank, entered the tank at the top and the liquid outlet was  $2 \frac{1}{2}$ inches from the bottom. The tank was equipped with a pressure gauge and a thermometer. The liquid line from the tank to the nozzle contained a rotameter and two quarter inch, stainless steel, needle valves. The valves were placed before and after the rotameter.

The benzene tank was a 6-foot length of standard 2-inch pipe fitted at the top for an air inlet line and a filling plug. A fitting for a



ABSORPTION APPARATUS KEROSENE **BENZENE**  $\biggl\{$  $\overline{O}$ FIGURE liquid outlet line was placed four inches from the bottom and the bottom end of the pipe was sealed with a standard pipe cap. Compressed air to pressurize the tank was furnished by a Worthington Air compressor. The air line contained a Conover reducing valve for the purpose of maintaining constant pressure on the benzene system and a shut-off valve. The liquid line from the benzene tank was equipped with a rotameter, a quarter-inch stainless steel needle valve, a standard quarter-inch T with a bleed-off line and valve, a quarter-inch valve, a check valve, and a vaporizer which were placed in the order mentioned. The bleed-off line and valve were placed in the liquid line to enable "in place" calibration of the rotameter. This was accomplished by closing the quarter-inch valve between the bleed-off and check valve to direct the benzene flow into the bleed-off. During actual runs, the valve on the bleed-off was closed and the quarter-inch valve mentioned above was opened full. The purpose of the check valve was to preclude the possibility of vapor backing into the liquid line as the benzene was vaporized. Sufficient pressure was maintained on the benzene supply tank to reduce this possibility to a minimum.

The vaporizor consisted of nine feet of eighth-inch copper tubing would around a half-inch core in a single layer. This was covered with asbestos as insulation and wound with a 26 ohm resistor consisting of number 22 Nichrome wire. The whole assembly was covered with eighthinch asbestos to insulate against heat loss. The vapor line leaving the vaporizer led to a T which was also wound with resistance wire to act as a booster heater. The resistance wire was insulated from the T by sheet asbestos and also covered with asbestos lagging. A thermometer was

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placed in one outlet of the T, and the vapor line from the T was connected through the back of an elbow into the air line. The heating elements of the vaporizer and booster heater were connected in series and controlled by a standard Variac.

Air was supplied by a Worthington air compressor having a rated capacity of 17 C.F.M. at 85 pounds pressure. The air line leading from the compressor to the junction with the benzene vapor line contained a flexible connecting line, a Conover pressure regulating valve, a pressure gauge, a thermometer, a rotameter, and a valve, all placed in the line in the order mentioned. The purpose of the flexible connection was to damp the vibration of the equipment caused by the air compressor.

At the point of junction of the air and benzene vapor lines the air had to make a 90-degree turn with the vapor joining the air stream during this time. Just downstream from the junction a union was installed to permit breaking of the line for the purpose of checking possible benzene condensation. The line carrying the gas mixture was connected to the nozzle by a quarter-inch copper tube.

The nozzle, described under "Design of Nozzle," was connected into a collector consisting of a glass column  $5 \frac{1}{2}$  inches in diameter and 3 feet long. The collector was fitted at both ends with standard column flanges and placed just off the horizontal to allow drainage. The flange on the upper end of the column was tapped and fitted with the nozzle and a thermometer. The flange on the lower end of the column was tapped and fitted with a sampling tube, a pressure gauge, and a gas outlet line. The gas outlet line was led out of the building to prevent accumulation of benzene vapors in the work space.

All rotameters, thermometers, and pressure gauges were calibrated.

# Experimental Procedure

It was necessary to "warm up" the equipment for forty-five minutes to an hour prior to each new series of runs. The compressors and Variac were started and allowed to run about thirty minutes and then the liquid benzene flow was started into the system. After the benzene flow had started, another fifteen to thirty minutes was allowed to enable the vaporizor and vapor lines to attain constant temperature. This warm-up period was necessary as the air compressors tended to heat for awhile after starting, thus causing the air temperature to change, and also benzene would condense in the vapor lines prior to entering the air stream when the equipment was cold. When the temperature of the benzene vapors, measured at the T just prior to entering the air line, became steady at 120° centigrade, the air-benzene line was broken and checked for condensed benzene. If no liquid benzene was present, the benzene, air, and liquid rates were set and the system was allowed to run for several minutes longer before runs were started. All of the above precautions were taken to insure representative and reproducible samples when the actual runs were started.

Runs were made in the following manner: After all rates were steady, the collector was drained and then washed, by allowing about 200 - 300 c.c. of liquid to collect and draining, before the sample was taken for analysis. The sample was taken for analysis and analyzed immediately by the Interferometer as described under "Analytical Methods." Readings

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of air rate, temperature and pressure; benzene rate, temperature, and vapor temperature; kerosene rate and temperature; and the temperature of the mixture inside the chamber were taken and recorded at the time of sampling..

The line carrying the air-benzene vapor mixture was broken just beyond the point of mixing before and after each run to check for possible condensation of benzene vapors, in the event of benzene condensate being found the run concerned was thrown out and a new run made When a change in any of the fixed rates occurred during a run, the run in question was thrown out and a new run made.

The calibration of the Interferometer was checked each day using kerosene and benzene samples taken from their respective tanks prior to starting the equipment.

## Analytical Methods

Originally it was planned to analyze the inlet and outlet gas streams and to meter the inlet liquid, thus tying down three of the four streams. The composition of the fourth stream, the outlet liquid, was to be derived by difference and checked frequently by distallation analysis. This procedure would necessitate an accurate analysis of the benzene-air and benzene-air-kerosene mixtures.

A modified Orsat analysis, in which the samples of the gas were burned and the products of combustion measured accurately, was tried. Although this method takes a long time to run, it is simple and also gives a check for accurateness in that the decrease in volume has to be balanced against the carbon dioxide formed. Using air-benzene

mixtures it was found that this balance could not be made, The decrease in volume always exceeded the carbon dioxide formed. The most plausible explanation for this seemed to be incomplete combustion with part of the benzene burning to carbon instead of carbon dioxide. After repeated trials with the equipment, a coating of carbon could be seen in the combustion tube. The intensity of the heat on the ignition wire was increased and the rate of passing the sample over the wire was decreased, but still there seemed to be incomplete combustion.

The Orsat combustion method was abandoned and a method using *20%* Oleum and Bromine was tried. The sulphuric acid solution of the Combustion Orsat was replaced with a solution of *20%* Oleum and Bromine thru which the sample was passed repeatedly until no further reduction in volume was discernable. Although this method had reportedly been used with very satisfactory results it was noted that the sample led out of the Oleum could be caused to burn in the combustion chamber. As benzene was the only constituent present which should burn, this indicated incomplete absorption of benzene in the Oleum, identical samples were analyzed by the combustion method and by the method using Oleum. In all cases the combustion method indicated a higher percentage of benzene in the mixture, so the second method was also abandoned as being unsatisfactory.

Next a gas Interferometer was tried. Gas cells were placed in a standard portable Interferometer and benzene-air mixtures were led through one cell and air through the other. This method proved satisfactory for very low concentrations, but two faults were found. The first was the narrow range of the instrument due to its great precision and secondly

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the difficulty of calibration at concentrations in excess of 1% benzene in air. The solubility and physical properties of benzene precluded the accepted methods of collecting gas mixtures and of moving the mixture through the Interferometer.

Numerous other methods of benzene-air analysis were considered, with the most promising being the use of "cold fingers," dew point determination, and thermal conductivity cells, but none was found which would give the accuracy desired. In view of the difficulty encountered in the analysis of the gas stream, a method was sought in which the constituents entering the stream could be determined without analysis. This was accomplished by adding a benzene vaporizer to the equipment. Thus the benzene was metered through an accurately calibrated rotameter, then vaporized, and led into the air stream in the gaseous state. The air was metered just prior to mixing with the benzene vapor. In this manner the composition of the inlet gas mixture could be accurately determined.

With the above modification the composition of the two inlet streams could be determined, but the two outlet streams were still undetermined. As efforts to analyze the inlet gas stream had failed to yield a satisfactory method, it was decided to determine the outlet gas stream by difference and to analyze the outlet liquid stream. Several methods of analysis were considered for this stream, with analysis by Interferometer being used. The gas cell of the portable Interferometer was replaced by a one centimeter liquid cell. Although the Interferometer is basically an instrument for measuring refractive indices it works on a basis of comparison with a known solution and this property was utilized for analysis.

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The manufacturers of the Interferometer state that, due to the high precision of the instrument, generally the calibration will be a straight line function for the first third of the range. In this range of the instrument the solutions are so dilute that Raoult's law holds in most cases, and it was found to be true in the case of benzene-kerosene solutions. The instrument was carefully calibrated for each new batch of kerosene and each time a straight line function was obtained for solutions which did not differ more than 1.3% by volume. Since the Interferometer proved to be an accurate and highly reproducible means of analyzing the benzene-kerosene solution it was adopted as the method of analysis for this work.

### Experimental Results

The carbon dioxide-water system was used mainly to evaluate the equipment and nozzle design. The data collected for this purpose is given in Tables 1 - 10. Air rates and carbon dioxide rates in all the tables have been corrected to cubic feet per minute at one atmosphere and 700 F. The pressures listed in the tables are of the metered gas and not that of the gas-water mixture. The pressure of the gas-water mixture was one atmosphere.

Table 1 was prepared to show the time necessary for a constant sample to appear at the sampling end of a pipe 11 feet 10 inches long. This time appeared to be used for the liquid sample to travel the length of the pipe as it was later found that constant samples could be obtained almost instantaneously right at the nozzle.

Table 2 indicates the effect of varying gas rate upon absorption using pure carbon dioxide. The increase of gas rate decreases the size of the liquid droplets thus exposing greater surface area for absorption.

Table 3 shows the results of using a carbon dioxide-air mixture for the gas and varying the rates and composition of the gas. All subsequent runs for the carbon dioxide-water system were made using air and carbon dioxide.

Table  $\mu$  was prepared to show the effect of different type collectors on the absorption. It was noted that the collector did not seem to help

 $\mu$ <sub>3</sub>.

the actual absorption but worked mainly as a device for breaking down the fog pattern. The best collector appeared to be a  $\frac{1}{4}$  foot by 2 3/ $\frac{1}{4}$ inch pipe in a horizontal position. The use of a smaller collector resulted in a high gas velocity and hence possible blowing out of the absorbed carbon dioxide. A larger sized collector would not adequately condense the fog, thus leading to smaller observed values of absorption. Runs 12 - 16 of Table 4 were made one day and runs 17 - 36 were made the next day.

Table 5 shows a comparison of analytical methods. Two samples were taken on each run and analyzed by the two methods described under analytical methods. On the basis of the results shown in Table  $5$  the second method of analysis described was accepted for the remainder of the experimental work. All runs shown in Tables  $1-\mu$  had been analyzed by titration with barium hydroxide.

Table 6 indicates the effect of changing gas composition on the amount of carbon dioxide absorbed. A glass column was used as a collector to observe the break up of the fog pattern.

All runs shown in Tables 7-10 were made using distilled water and with a modification to the original equipment. The procedure for this work and the equipment modificatlpns are explained under experimental procedure and experimental equipment.

Table 7 shows the results of varying gas compositions and rates at low liquid rates. The collector consisted of a  $5$  1/2 inch by 3 foot glass column and the gases from the collector was discharged into a Cotrell precipitator. The nozzle used was the "convertible" nozzle shown in Figure 2 with a 0.150 inch outlet orifice and a 0.01 $\mu$ 6 inch liquid

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orifice» The total gas rate varied from that which would barely break up the liquid stream to rates which produce very fine fog.

Table 8 shows the results of varying gas rates and compositions at higher liquid rates than used in Table  $7$ . The nozzle used was the same as described above with a  $0.150$  inch outlet orifice and a  $0.0256$  inch liquid orifice. All other conditions were as described for Table  $7.$ 

Table 9 shows the results of varying gas rates and compositions at very high liquid rates. A  $0.033$  inch liquid orifice used was in the nozzle with all other conditions remaining the same as above.

The data of Table 10 shows the results obtained when using no Collector and thus is an indication of the absorption actually obtained due to the nozzle. The mixture from the nozzle was discharged directly into a Cotrell precipitator and collected. Runs  $1 - 15$  were made using the original nozzle shown in Figure 1 and runs  $16 - 43$  were made using the nozzle shown in Figure 2 with a  $0.150$  inch outlet orifice and a 0.0146 inch liquid orifice. Rates of all streams were varied, as noted by the tabulated data, in order to compare the results with those obtained using collectors and different sized orifices.

The kerosene-benzene system was used to study the effect of atomization on absorption and the results obtained are tabulated in Tables 12 — 20. Air rates have all been corrected to atmospheric pressure and  $70^{\circ}$  F. The volume of benzene vapors was calculated using the perfect gas  $law$ . Drop size was calculated using Equation 1 with the assumption being made that the density and viscosity of the benzene-air mixture was the same as that of air. This assumption is justified because of the very low concentrations of benzene used. The pressure in the collecting system

was atmospheric at all times•

A commercial grade benzol was used in the early part of the investigation and the results are shown in Tables  $12 - 14$ .

The gas mixture used for the runs shown in Table 12 was approximately 3% by volume benzol. Gas and liquid rates were varied in order to obtain different liquid gas ratios and different drop sizes.

Table 13 shows the results of varying liquid rate upon the drop size and upon the absorption. The gas used was 1% by volume benzol.

Runs 1 - of Table 14 were made using a gas approximately *1%* by volume benzol and runs  $16 - 23$  were made using a gas approximately  $1\%$  by volume chemically pure benzene.

Tables  $15 - 20$  depict the results obtained using a pure benzene-air mixture for the gas mixture with the composition ranging from  $0.4%$  to 3» *2%* as indicated. The liquid orifice for runs 13 - 20 of Tables 16 and 17, runs  $11 - 18$  of Tables 18 and 19, and runs  $10 - 20$  of Table 20 was 0.0226 inches in diameter»

The diameter of the liquid orifice for all other runs shown in Tables  $12 - 20$  was  $0.01\mu$ 6 inches and that of the outlet orifice for all runs was 0.150 inches.

Determination of time necessary to obtain constant sample using iron pipe  $10'11''$  x  $2$   $1/2''$  as collector.



# TABLE 2

### Effect of variation of COg rate.  $\text{Collection} - 10'11'' \times 2 1/2''$ iron pipe.



# TABLE 3

# Effect of variation of gas composition. Temperature 70°F.  $\text{Collection}$  -  $10$ 'll" x 2  $1/2$ " iron pipe.



# Effect of varying size of collector. CO<sub>2</sub> rate 1.26 C.F.M. air rate 2 C.F.M. Pressure  $32.7$  psia.  $38.6%$  CO<sub>2</sub> in inlet gas



Comparison of titration methods. Gas rate and composition, pressure and temperature were held constant. Water rate was 92 cc/min. for runs 37 & 38 and 85 cc/min. for runs 39 - 47. Collector was  $4' \times 2 \frac{3}{4}$ " glass column in a horizontal position.



# TABLE 6

# Effect of variation of gas composition. Water rate constant at 100 cc/min. Collector  $\mu$ <sup>\*</sup> x 2 3/ $\mu$ <sup>n</sup> glass column



# Absorption of  $\infty_2$  with Water



# Absorption of  $CO_2$  with Water



# Absorption of  $CO_2$  with Water



### Absorption of CO<sub>2</sub> in Water. Discharge from Nozzle Direct to Gotrell.



# Comparison of Absorption



Absorption of Benzol with Kerosene

Approximately 3% Benzol



# Absorption of Benzol with Kerosene

# Approximately 1% Benzol



Absorption of Benzol with Kerosene

Approximately 1% Benzol



# Absorption of Benzene with Kerosene

# Approximately 1% Benzene



Absorption of Benzene with Kerosene

Approximately 1% Bensene<br>(Varying Liquid Bate)



Absorption of Benzene with Kerosene

Approximately 2% Benzene<br>(Varying Liquid Rate)

 $\bullet$ 



# Absorption of Bensene with Kerosene

Approximately 3% Benzene<br>(Varying Liquid Bate)



**TAHLE 19** 

Absorption of Benzene with Kerosene

Approximately 0.5% Benzene<br>(Varying Liquid Rate)



TAHE 20

# Absorption of Benzene with Kerosene

Approximately 1% Benzene<br>(Varying Liquid Rate)


## Discussion of Results

The carbon dioxide-water system was used primarily to evaluate the design of the equipment and will be discussed chiefly in connection with this evaluation.

As this system consisted of atomizing a liquid and then collecting the spray it was essential to have some means of condensing the atomized liquid. The results shown in Table  $\mu$  may be used to assess the proper design of a collector and to note some of the characteristics of the system. In the use of collectors of small diameter the velocity of the gas mixture was excessive and seemed to desorb some of the absorbed gas. It was concluded that this constituted two systems, an absorption system in the nozzle and a desorption system in the collector. The use of a long collector of larger diameter seemed to produce the same effect. A collector of rather large diameter also produced what appeared to be low absorption. This result can be explained as being due to insufficient condensation, or the sampling of the larger drops only. The actual absorption was not being measured as the finer drops, which would contribute greater surface area, were being swept out of the collector by the escaping gases.

The validity of the above assumptions were based on visual observations of the phenomena taking place and on the results as listed. It can be noted from Table  $l_1$ , run  $l_1$ , that the absorption obtained when the sample was taken directly from the nozzle was greater than that obtained

with any collector other than the 4 foot by 2  $3/4$  inch pipe and 8 foot by 2  $3/\mu$  inch pipe in a horizontal position. The use of a larger collector in conjunction with a Cotrell precipitator which condensed the finer drops gave excellent results but it can be seen by comparing Table 7 and Table 10 that the Cotrell alone was sufficient to condense the fog.

The criteria for design of a suitable collector for an absorption system using atomization would seem to be one in which the gas was not allowed to sweep through the liquid containing the absorbed material and one in which complete condensation took place. A Cotrell precipitator meets the above requirements and can be used for any non-combustible system.

The design and characteristics of the nozzle used was a main item of study. In the preliminary study the nozzle shown in Figure 1 was used. The data shown in Tables  $7-9$  was collected using the convertible nozzle, Figure 2, with different sized liquid orifices. As shown by the results, apparently the size of the liquid orifice does not materially affect the design of the nozzle was long as the relative velocity and liquid-gas volume ratios remain constant. This result was to be expected as it follows from the fact that drop size would remain constant under these conditions and hence surface area of the liquid would remain constant. The actual nozzle design seems to be immaterial providing the conditions of high relative velocity and/or high gas-liquid volume ratios are obtained. Thus, given the conditions of a system, a nozzle may be designed to fit the system.

All other parts of the equipment were auxiliary to the nozzle and

collector. With the modifications, noted under "Experimental Apparatus" they proved to be adequate.

Although the primary consideration was the evaluation of the equipment several features of absorption noted are worth mentioning. Paramount among these is the nearness of approach to recognized equilibrium values of the solubility of carbon dioxide in water attainable almost instantaneously by the method of atomization when pure CO<sub>2</sub> was used as the gas. For example in run 6 of Table 2 the absorption was 96.*2%* of handbook values<sup>22</sup>, in run 1 of Table 1 it reached  $9\nmid 6\n$ , and in run 2 of Table 1 it was 88%. When an air-carbon dioxide mixture was used as the atomizing gas it was found that the equilibrium value calculated by using the partial pressure of carbon dioxide and Henry's Law was exceeded at times. This discrepancy between calculated values of expected solubility and actual solubility obtained by absorption had been noted previously by Koch, Stutzman, Hlum, and Hutchings.<sup>23</sup> Two possible explanations may be given for this discrepancy if it is recognized that the system used in deriving the tabulated values of solubility and of Henry's constant was pure carbon dioxide and water. Thus the change in partial pressures of carbon dioxide was caused by a change in the total pressure of the system and not by the addition of an inert gas. From this it follows that the system used, air-carbon dioxide-water, is actually a different system. The total pressure of the actual system used was always 760 mm and this fact may well have changed the solubility of the carbon dioxide. Another possibility would be that the supposedly inerts in the air, nitrogen and oxygen, affected the solubility of the carbon dioxide. It is true that both oxygen and nitrogen are only very slightly soluble in

water but even this slight solubility may affect the activity of the carbon dioxide in this system. Neither of the above reasons are being stated as the cause of the noted discrepancies but only as possibilities.

Although drop sizes have not been calculated for this system the result of decrease of drop size on absorption may be noted by reference to Table 2. The increase of gas rate, with liquid rate and nozzle constant, increases the relative velocity and gas-liquid volume ratio and decreases drop size.

In order to judge the effectiveness of an absorption system using atomization a comparison of the results obtained in this experiment was made with the results obtained from an experiment using a packed column.  $24$ Runs were selected from the Tables on page 991 of reference 24 which had percentage compositions nearly the same as runs in the present experiment and the data was converted to grams of carbon dioxide per hundred grams of water as a basis of comparison. Table 11 was prepared to depict the compared results.

The results of the above comparison show that a higher concentration of gas was consistently obtained from atomization than from the packed column absorption. The actual difference is even more pronounced than the tabulated figures indicate as the packed column was operating at temperatures ranging  $10^{\circ}$  to 15° Centigrade below those of the present experiment.

As previously stated the primary reason for using the kerosenebenzene system was to be able to compare experimental results with published results in order to judge the effectiveness of atomization in an absorption process. This comparison and also several different

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aspects of the process as noted during the experimental work will be discussed.

The published results selected for the comparison was that presented by Glowoeki $^{149}$  for a packed column equivalent to four transfer units. The data obtained by Glowoeki<sup>49</sup> was shown as a plot of gallons of wash oil per ton of coal versus percentage recovery of benzol. Glowoeki's curves, as reproduced in this thesis, are plotted as cubic centimeters of wash oil per cubic centimeter of benzene. These are the same curves as the conversion was made using the authors data of 2.21 gallons of benzol per ton of coal. Although the published results were obtained using a gas composition of approximately 1% benzol by volume, the statement was made by the author that the composition of the inlet gas did not materially affect the percentage recovery. Figure 7 is a comparison of data obtained in this experiment using compositions of gas of 1% by volume berfzol or benzene and of Glowoeki's curve. The experimental data used for the plot was obtained from Tables 13,  $14$ , 15, 16, and 20. Only those runs which were made using a  $0.0146$  inch liquid orifice and which had mean drop diameters of less than 190 microns were used. The reasons for this choice will be given below in the discussion of Figure  $3_*$ It can be noted from Figure 7 that the results obtained by atomization absorption compare most favorably with those obtained using a packed column of four transfer units.

The effect of drop size on absorption appears to become pronounced when the mean diameter of the drops are about 190 microns. This is shown in Figure 8 which is a plot of the data of Table  $1\mu_{\bullet}$  The percentage recovery can be predicted for this system at any point up to the critical





drop size, but beyond that size the results become erratic and unpredictable. This result was as anticipated when the experiment was undertaken, although the actual critical size could not be predicted without experimental results. It was felt that atomization would produce large surface area and provide intimate mixing thus making a co-current system feasible for absorption. It was recognized that this would necessitate small droplets and the results as explained above bear out the original assumptions.

A discrepancy in the results was found to exist when the absorption obtained using a  $0.0256$  inch liquid orifice was compared with that obtained using a 0.0146 inch orifice. Although, for the same kerosenebenzene ratio, the droplets were smaller for the larger liquid orifice, according to Equation 1, the percentage recovery was less. A close observation of the process gave an explanation for this discrepancy. It was noted that some of the liquid was dripping from the nozzle and was not being atomized when the larger orifice was used. This resulted in a lower, kerosene-benzene ratio in the atomized liquid than that tabulated. Figure 9 was plotted with data obtained from both orifices and although the above discrepancy exists it can be noted that the same tendency of the curve persists for both orifices.

Temperature apparently does not have as pronounced an effect on the atomization absorption process as it does on the packed column absorption No definite conclusions can be drawn on this point as the temperature differences used in the present experiment were not large. Nevertheless, the proposal seems valid as may be seen in Figure 10 which is a plot of experimental data having a temperature differential of  $6^{\circ}$  F<sub>\*</sub> and three





curves presented by Glowoeki. The data obtained in this experiment all falls very close to the same curve in spite of the temperature difference.

The concentration of benzene in the inlet gas has been reported to have very little effect on the percentage recovery at any given kerosenebenzene ratio in packed column absorption. 49 In the present experiment it was found that an increase of concentration of benzene in the inlet gas increased the percentage recovery of benzene. This increase was not particularly large, but as can be noted from Figure 11 it existed.

The concentration of benzene in kerosene is not tabulated in the tables but can be readily calculated from the percentage recovery, benzene rate, and kerosene rate. It was found to increase with the decrease in drop size thus giving further evidence of better absorption by the smaller droplets. The fact that percentage recovery seems to depend partially on the kerosene-benzene ratio may be explained in part by this concentration. The rate of transfer of the benzene from the gas to the liquid has not been determined but it is logical to assume that it depends in part on the concentration of benzene in the liquid. Another thing that would have an effect on the percentage recovery and the rate of transfer would be the surface area of liquid exposed per unit time per unit volume of gas. In order to check this factor the total surface area per unit time was calculated from the data of Table 13 where the volume of gas per unit time is constant. The area calculated was found to remain almost constant as the decrease in liquid rate offset the decrease in drop size. These two factors, the concentration of the benzene in the liquid and the fairly constant surface area of liquid exposed per unit time to unit volume of gas, seem to offer an explanation for higher



percentage recovery at higher kerosene-benzene ratios. The concentration of benzene in the larger liquid drop is less and thus provides a greater driving force for benzene to go from the gas into the liquid, When the droplets exceed 190 microns in size these effects seem to diminish and less efficient absorption is obtained,

A most important consideration of any process is the cost, not only of the process but also of the equipment. The results of this experiment show that atomization absorption can be accomplished in a system consisting simply of an atomizing nozzle and a collector. Since the system is co-current no large tower or chamber is needed to provide liquid gas contact, therefore the cost of equipment should be much lower for a commercial absorption system using atomization than for those now in use. The power and operating costs would be the basic criteria for selection of this system. Pumping power should be low but the necessity of compressing the gas would be important. In the present experiment the pressures of the gas used to atomize the liquid varied, in general, from one to 15 pounds, in a few cases higher pressures were used, bit were not found to increase the absorption enough to warrant their use. The pressures used in any system would have to be determined by such factors as design of the nozzle and capacities to be treated and also the power requirements for compression would be determined by the same factors.

Another important aspect of cost in commercial absorption system is the "down time" or time lost due to cleaning or repairing the equipment. This factor is often large in packed column systems but should be practically eliminated in an atomization absorption system. The latter system, could easily be designed with a number of atomizing nozzles that could be used for standby purposes. In this way if an operating nozzle became plugged or inefficient due to wear it could be removed without stopping the absorption process.

Commercial systems for the recovery of benzene in wash oils are in use at the present time that consist of series of towers which total as much as 300 feet in height. The cost of such a unit, both operating and initial, is great. This experiment shows that the same recovery can be accomplished by atomization absorption and the results compare most favorably with those obtained from the packed columns. Also, although laboratory sized equipment was used, this experiment shows that atomization absorption would require much smaller equipment as it precludes the use of large towers to hold the packing and provide liquid-gas contact space.

## Conclusions and Recommendations

The conclusions that may be drawn from this experiment serve mainly to justify the original premises concerning an atomization absorption process. The work was undertaken with the idea that the increase of area of liquid that would be possible by atomization, in a process which used the gas to be absorbed as the atomizing gas, would yield an efficient co-current absorption system. This system, as conceived, would consist simply of atomizing nozzles and a collection system, with necessary auxiliary equipment, thus resulting in a tremendous decrease in size and weight over present equipment.

From the results obtained in this experiment it can be concluded that absorption ty atomization is feasible and practical and that the system, being co-current, would require much smaller equipment than is required for present absorption processes. The need of large towers for contacting the gas and liquid counter-currently is eliminated as well as is the use of packing. Atomization absorption would need only atomizing nozzles discharging into a collector and thus the size would be determined by the size of a collection unit necessary to accommodate the capacity of the system. A Cotrell precipitator serves as a good collector for a non-combustible system.

It was originally thought that decrease of drop size would allow greater concentrations of gas to be absorbed in the liquid almost instantaneously and the results of the experiment verified this. The

equilibrium conditions of solubility can be very nearly approached simply by use of the two fluid atomizing nozzle. The time required for this condition to be attained is so short that it could be said to be instantaneous as the liquid and gas are travelling at the rate of about 100 meters per second and are in contact for a distance of only 0.01 meters thus a contact time of 0.0001 seconds.

In view of the results obtained in this experiment it is recommended that a larger system be built for the study of atomization absorption with the ultimate goal being commercial use. From the present findings it is easy to conceive of the tremendous decrease in the size of commercial equipment that could be realized over packed columns. Other important considerations from a commercial viewpoint is the fact that an atomization absorption system could easily be designed to be continuous and not have to be stopped for cleaning and repair of nozzles, and the high concentrations of solute in solvent possible.

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