AD 741846

AFML-TR-71-161 MRC-DA-305

Final Report

IMPROVING ULTRASONIC TRANSDUCER ASSEMBLIES GENERATING AEROSOLS FOR INSTRUMENTAL TECHNIQUES OF CHEMICAL ANALYSIS

LOUIS E. OWEN

TOMORROW ENTERPRISES

TECHNICAL REPORT AFML-TR-71-161

NOVEMBER 1971

19

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IMPROVING ULTRASONIC TRANSDUCER ASSEMBLIES GENERATING AEROSOLS FOR INSTRUMENTAL TECHNIQUES OF CHEMICAL ANALYSIS

LOUIS E. OWEN

FOREWORD

This report was prepared by Tomorrow Enterprises, 4408 Hickory Lane, Portsmouth, Ohio, 45662, under Subcontract 1132-2 from Monsanto Research Corporation under Air Force Contract F33615-71-C-1132. This contract was initiated under Project No. 7360, "Chemical, Physical and Thermodynamic Properties of Aircraft, Missile and Spacecraft Materials," Task No. 736005, "Compositional, Atomic and Molecular Analysis of Experimental Materials for Advanced Air Force Systems." The work was administered under the direction of the Air Force Materials Laboratory, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, with Mr. Freeman F. Bentley (AFML/LPA) as Project Engineer. The report was submitted by the author in June 1971.

This is the Final Report on this subcontract and summarizes work carried out from . December 1970 to 31 May 1971.

The work at Tomorrow Enterprises was performed by Louis E. Owen, Principal Scientist and Co-owner. A considerable portion of the experimental work was accomplished while Mr. Owen worked as a guest of the National Bureau of Standards in the Coordination Chemistry Section headed by Dr. Oscar Menis. Dr. Radu Mavrodineanu, a member of the section, specifically aided the project with technical advice and personal encouragement.

The contractor's report number is MRC-DA-305.

This technical report has been reviewed and is approved.

Freeman F. Bentley

FREEMAN F. BENTLEY
Chief, Analytical Branch
Materials Physics Division
Air Force Materials Laboratory

ABSTRACT

A search for surface coatings which, when applied to the working faces of active piezoelectric transducers, would prevent their destruction by sample solutions containing HF, was unrewarded. Coatings otherwise suitable for "fountain type" nebulizers could not withstand the great rigor of direct impingement nebulization in which the working surface is only lightly loaded. All failure modes involved loss of coating integrity with subsequent attack of the piezoceramic by the HF.

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SECTION I

INTRODUCTION

The intent of this contract effort was to improve transducers employed in direct impingement ultrasonic nebulizers. Improved mounts providing better cooling were to be devised and superior surface coatings protecting active nebulizing surfaces from chemical attack were to be sought. During the contract period principal effort was given to the development of chemical-resistant coatings for the piezoelectric elements employed as nebulizing devices. Superior cooling, though achieved, is a rather moot point in the absence of transducers capable of withstanding sample solutions of pertinence.

The work of this report is best understood as an essentially integral extension of the work reported in Technical Report AFML-TR-70-220.

SECTION II

SUMMARY

Superior mounting techniques developed for coaxial piezoelectric disks resulted in greater longevity and more predictable performance for direct impingement nebulizers. Nonfluoride containing sample solutions were nebulized with great efficiency and analytical usefulness in atomic absorption spectrophotometry. However, the major goal of the project, the protection of active nebulizing surfaces from attack by fluoride ions, was not achieved.

SECTION III

TRANSDUCER MOUNTS

The transducer mounting (Figure 1) used primarily as a test assembly for evaluating coatings is also amenable to use as a direct impingement device. The piezoelement is either soft soldered or cemented with silver-containing epoxy in the center of a large brass washer. The brass is no more difficult to coat protectively than the silvered electrode face of an active disk and, as a passive surface, more easily retains the protection. Cooling is excellent because of the open construction, though high purity water must be used and its purity maintained.

Another test assembly for coated disks was designed to permit simple interchange of the piezoelements. The disks are held tightly against the lip of a Teflon tube by a brass cylinder and a spring-loaded, insulated center contact. Mechanical dampening of the vibrating disk reduces its utility as a direct impingement nebulizer, but coatings may be tested validly and serial evaluations may be made rapidly.

Figure 2 illustrates a transducer mounting assembly complete with nebulizer chamber. The design is derived from the unit shown in Figure 2 of AFML-TR-68-354. Improved piezoelement mounting for more efficient cooling incorporates a thin rod contact cemented to the central electrode on the rear of the disk. The rod and brass sheet are thinly coated with an electrically insulating though thermally conducting epoxy coating. The use of highly purified water is desirable to reduce the significance of inadequately coated areas.

Glass-glazed piezoelectric elements in the mount of Figure 2 have survived hundreds of hours of frequency-monitored operation while nebulizing moderately acidic solutions.

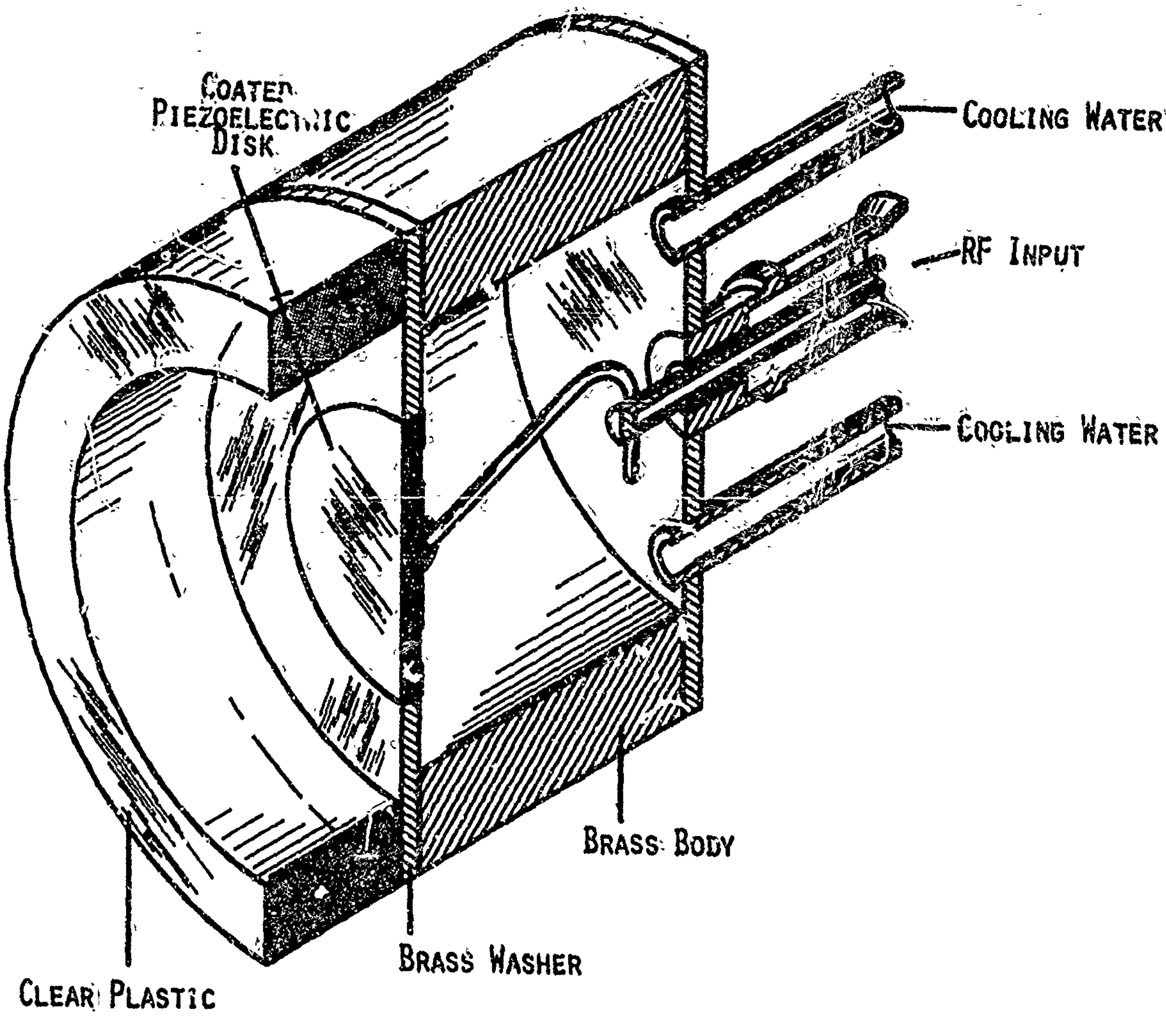

Figure 1. Test Assembly for Coated Disks.

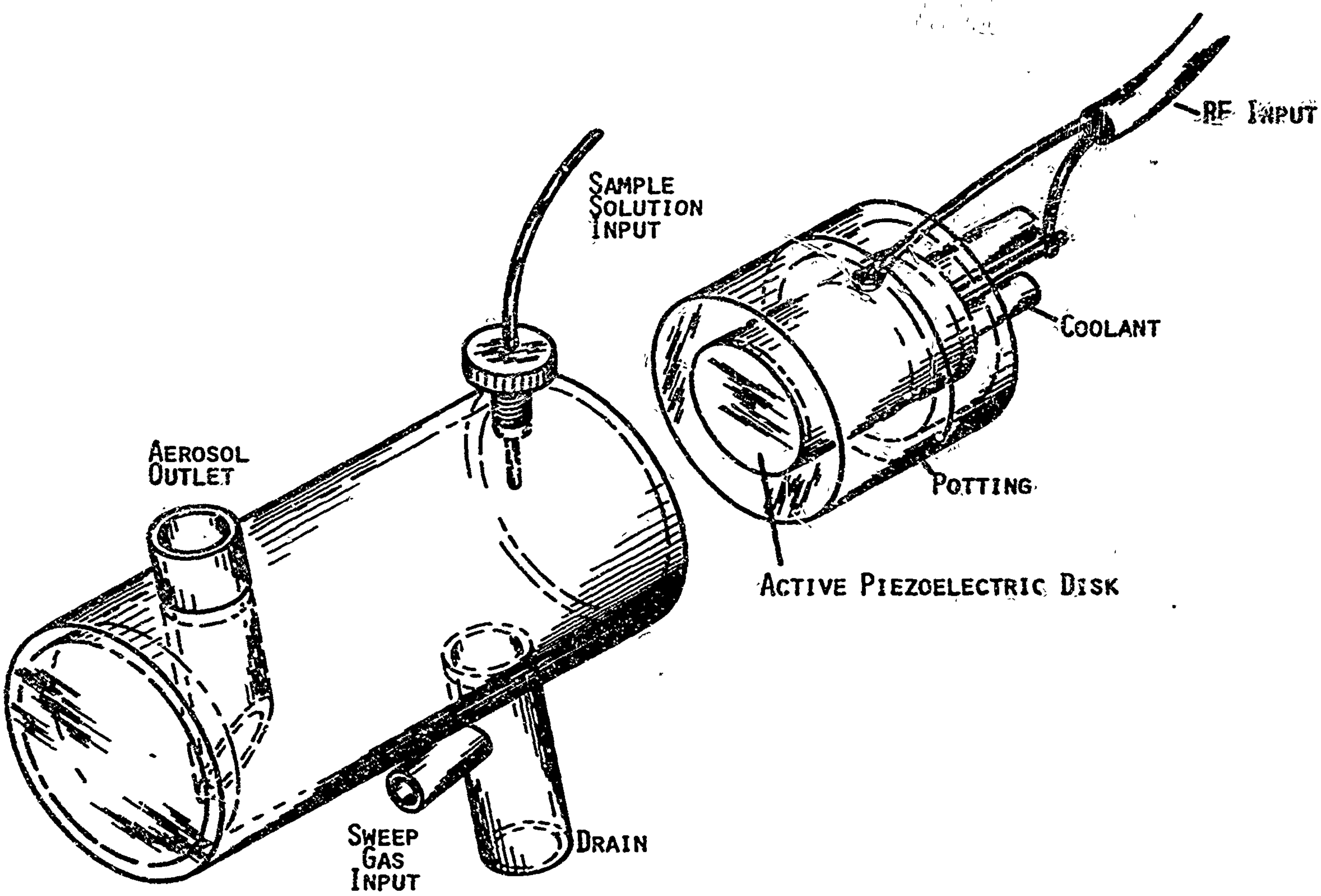

Figure 2. Direct Impingement Ultrasonic Nebulizer Assembly.

SECTION IV

EVALUATION OF COATINGS

During the evaluation of coatings it was noted that coated disks are more suited to "fountain" nebulization than to direct impingement nebulization.

In "fountain" operation, of course, the disks are more uniformly loaded and better cooled. More importantly, perhaps, they are in constant contact with a medium capable of accepting the acoustical energy. If coatings could be of rigid, highly elastic material and of the right thickness (preferably half wavelength), then their working faces would have maximum excursion while their interface with the driving disk would be lightly stressed. A resonant composite would exist for which a gas interface (partially normal in direct impingement nebulization) would introduce no additionaly problem. Available coatings, however, are either inelastic or at best quite difficult to coat in precise thickness. As a consequence they couple well only to a liquid where an energy transfer can take place in spite of inexact impedance matching.

Direct impingement nebulization, in which the working face is incompletely and rather intermittently wetted, is inherently inefficient in transferring its acoustical energy to the solution presented for nebulization. The bond between the piezoelement and its coating is greatly stressed. High power inputs are required while, concomitantly, working surface cooling is limited. It is thus obvious that the requirements are stringent for candidate protective coating materials.

1. FLUORINATED COATINGS

Resistance to HF is an obvious characteristic of materials already fluorinated. Teflon and its homologues therefore rated extensive consideration. Fluorinated coatings may be obtained from commercial applicators or may be attempted using a variety of formulations intended for field application.

a. Sintered Coatings

Custom application by commercial concerns incorporates sintering of the coating compound at temperatures exceeding the Curie point of the most heat resistant piezoceramics. It was necessary, therefore, to have disks coated after fabrication including electrode application but before poling (being made piezoelectrically active). It takes more than six months before coatings may be evaluated, but we did complete tests for two different ceramic compositions.

Failure modes for all disks tested appeared to result from bond failure between the coating and the silvered electrode face. Eventual dramatic attack of the ceramic by the test solution showed that film integrity was also lost. The sequence involved is not well established.

It is possible that the sintering temperatures for the fluorinated coatings are high enough that partial ceramic-silver solutioning exists. In order to establish this point another series of test disks were being coated commercially and poled at the end of the contract period. Evaluations completed were ambiguous enough to justify continuation of the approach.

b. Nonsintered Coatings

A large variety of fluorocarbon formulations, advertised primarily as release and lubricating agents, are available for field application. They require curing conditions ranging from room temperature drying to baking at temperatures (300°C) which approach the sintering temperatures used by commercial applicators.

We worked unsuccessfully with four different formulations and applied a variety of multiple coats in attempts to produce a complete barrier to the test solution. Multiple, very thin coats had a better appearance than equivalent thickness single coats. None, however, was able to withstand high power driving long enough to be analytically useful. Lack of coating integrity accounted for failure more frequently than bond failure to the electrode silver. As in the case of sintered coatings, sufficient promise was shown that experiments are continuing beyond the contract period.

2. GOLD

Gold is theoretically insoluble in sample solutions of interest to the Air Force but gold plating is notoriously imperfect, exhibiting pin holes and other defects. Gold surfacing is used, however, for lining pots used for molten fluorides by some AEC installation. In particular, lab personnel at the Ames Laboratory, Iowa State University, felt that they could obtain nonporous, purportedly sputtered, gold surfacing. High efficiency piezo-ceramic disks were obtained and sent to Iowa State for gold coating. After being coated they were returned to their original manufacturer for poling. Following a 12-week aging period the disks were tested in the standard HF solution. They failed after operating six hours at moderate power level.

It is possible, of course, that the gold coat was impaired by mechanical piercing during the poling process. High pressures and temperatures are used in the poling operation.

Gold coating by sputtering is precluded for previously-poled ceramics because their Curie point might well be exceeded. Electroplating, however, is applicable for otherwise completed disks and claims are made by plating supply firms that nonporous platings are possible for plate over fifty millionths of an inch thick.

3. PROPRIETARY COATINGS

a. Unknown Composition

The industrial world is replete with proprietary materials designed to protect metal surfaces against the ravishment of hostile environments. From one highly-touted line of "clear sealers" we selected four different sprayable solutions for test. They all provided static protection but apparently absorbed enough energy under insonation so that the coating eventually evaporated. The pattern associated with thin liquid films on active piezoelectric disks was well duplicated in the pattern of fluoride attack. The protection provided by such air-drying sealants is limited, in our experience, to nonworking surfaces.

b. Polysiloxanes

The thermosetting organopolysiloxanes, typified by Owens-Illinois' "glass resin" series, continue to provide hope and frustration as chemically resistant coatings for ultrasonic nebulization. They produce hard coatings which occasionally are of the correct thickness to resonate as half wavelength plates. On the other hand, it was impossible to attain such thickness reproducibly, and sufficiently long-lived resistant transducers were not produced. The class of compounds is still considered promising and efforts to use them will continue.

SECTION V

PUMPS

The complete nebulizing systems desired for analytical usage require sample solution pumping to the active face of the piezoelectric disks. In consequence, interest in the low flow rate pumps continues. During the project period, the three-roller peristaltic pumps available were fitted with a six-roller shaft. With smaller volume entrapped between successive rollers, the pumps required rpm's 150% greater than the three-roller pumps for equal flow rates. As a result, flow pulsations have lessened amplitude and appear with three times the frequency. Flow across a piezoelectric transducer was appreciably smoother.

The original designer and manufacturer of the pumps expressed interest in the work and promised to make the six-roller pumps commercially available.

SECTION VI

CONCLUSIONS

Although coating work during the contract period did not result in the development of a coating for piezoelectric disks capable of withstanding HF, commercially applied Teflon, field applied fluorinated compounds, and organopolysiloxanes offer sufficient hope for success that experimental work continues.

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UNCLASSIFIED
Security Classification

DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Monsanto Research Corporation 1515 Nicholas Road Dayton, Ohio	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b. GROUP

3. REPORT TITLE
IMPROVING ULTRASONIC TRANSDUCER ASSEMBLIES GENERATING AEROSOLS FOR INSTRUMENTAL TECHNIQUES OF CHEMICAL ANALYSIS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)
Final Report - 1 December 1970 - 31 May 1971

5. AUTHOR(S) (Last name, first name, initial)
Owen, Louis E.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
October 1971	14	5

8a. CONTRACT OR GRANT NO. AF33615-71-C-1132 Subcontract 1132-2	9a. ORIGINATOR'S REPORT NUMBER(S) MRC-DA-305
b. PROJECT NO. 7360	
c. Task No. 736005	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) AFML-TR-71-161
d.	

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11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Air Force Materials Laboratory Air Force Systems Command Wright-Patterson AFB, Ohio 45433

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DD FORM 1 JAN 64 1473

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
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nebulization						
resistant coatings						
fluorinated coatings						
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