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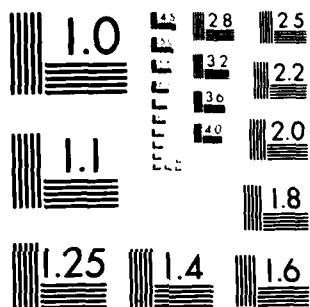
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**EXPERIMENTAL DESIGN FOR THE MEASUREMENT OF ACOUSTIC
PARAMETERS OF MARINE SEDIMENTS CONTAINING GAS HYDRATES**

J. Mark Daniels
Paul J. Vidmar

**APPLIED RESEARCH LABORATORIES
THE UNIVERSITY OF TEXAS AT AUSTIN
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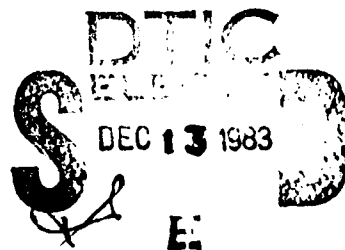
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Technical Report

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and sufficiently low temperature to form gas hydrate crystals. The vessel can be easily modified to accept a variety of transducers for measuring the acoustical, thermal, and electrical properties of hydrated marine sediments. The experimental plan calls for three phases. In the first, the basic apparatus will be constructed and the most difficult aspect of the experiment, actually growing hydrates in sediments, will be perfected. In the second phase, modifications suggested during the first phase will be implemented, transducers for acoustical measurements will be added, and the initial measurements of the acoustical measurements will be taken in which the sediment type, pressure-temperature conditions, and hydrate concentrations will be varied over a wide range in order to determine their influence on the acoustic parameters.

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I. INTRODUCTION

The effect of the ocean sub-bottom on underwater sound propagation is an active field of research. Much theoretical and computational effort has been and is being expended to determine how the acoustical parameters of the ocean bottom and sub-bottom structure (geoacoustic profile) affect acoustical quantities such as bottom reflection loss, mode attenuation coefficients, group velocities, and propagation loss. The result of such efforts is useful only to the extent that the geoacoustic profiles used actually contain the relevant acoustic parameters of the ocean floor.

One sub-bottom feature that is seldom included in geoacoustic profiles is the presence of hydrates in marine sediments. Hydrates are crystalline compounds formed when gas molecules are trapped in an icelike crystalline structure. Hydrates in marine sediments can significantly alter the values of geoacoustic parameters from those expected for nonhydrated sediment structures. The changed values of quantities such as compressional speed, shear speed, and absorption can greatly alter acoustic predictions. The occurrence and acoustical significance of natural gas hydrates in marine sediments has recently been reviewed.¹

The formation of natural gas hydrates requires three conditions: sufficiently low temperature, sufficiently high pressure, and saturation level gas in the water. The pressure-temperature conditions suitable for hydrate formation are found over 90% of the ocean floor and to a considerable depth in the sediment. There is substantial evidence,¹ that both biogenic and thermogenic gases are found in sufficient quantities for hydrate formation in many seafloor areas. Zones of gas hydrates have been detected dispersed in sedimentary sequences, occupying fractures in sedimentary and metamorphic rocks, and as massive units of pure crystal.

Recent numerical studies¹ have demonstrated that bottom loss from hydrated marine sediments can be significantly different from that expected from a nonhydrated sediment. The computed bottom loss was obtained using compressional wave velocities that were consistent with the few available laboratory measurements; basically, however, the computation relied upon estimates of the shear velocity and absorption, not yet measured. The computed bottom loss was found to be particularly sensitive to the value of the shear velocity.

One conclusion drawn from these studies is that hydrates are a potential mechanism for producing the enhanced reflectivity² of the ocean floor observed at many locations in the Atlantic. Whether hydrates can indeed produce the enhanced reflectivity depends on the actual value of the shear speed of the hydrated sediment. If hydrate formation is the cause of the enhanced reflections, this has implications for modeling the ocean floor and for defining geologically similar areas for use in extrapolating acoustic features.

This report describes an experimental apparatus and experimental plan developed during 1982 by ARL:UT for carrying out laboratory measurements of the acoustic properties of marine hydrates. Safety considerations drove the design toward operation at low pressures. During the initial learning phase, a nonexplosive gas, xenon, will be used. The acoustic measurements will use transducers developed at ARL:UT³ that have been successfully used to measure shear and compressional velocities. The experimental plan will first concentrate on the difficult task of producing samples of hydrated marine sediments. Then prototype acoustic measurements will be made, followed by a concentrated series of acoustic measurements using a variety of sediment types and gases.

This report is organized as follows. Section II reviews the nature, occurrence, and properties of hydrated marine sediments. Section III presents the experimental apparatus, and Section IV, the experimental plan. Section V is a summary.

II. GAS HYDRATES IN MARINE SEDIMENTS

Gas hydrates are icelike crystalline compounds in which a water lattice forms a cage that encloses a guest molecule in a gaseous state. Hydrates are formed under a wide range of pressure-temperature conditions typified by low temperature, high pressure, and saturation level gas in the water. These pressure-temperature conditions exist over 90% of the ocean bottom and to a considerable depth into the sediment, while gas concentrations are suitable in many seafloor areas.⁴

Most occurrences of hydrates have been inferred from the observation of bottom simulating reflectors (BSRs) or seismic bright spots. In both cases, the reflector is believed to be at the bottom of the hydrate zone where the temperature has increased sufficiently to prevent hydrate formation deeper into the sediment. It is believed that hydrates act as an impermeable cap, trapping free gas below the hydrate thermal stability region. The large impedance contrasts which result cause the observed phase reversals characteristic of BSRs. Gas hydrates have been recovered from the ocean floor at one site on Leg 76 of the DSDP near a bottom simulating reflector. The recovered cores indicate that the hydrates in this region exist in thin layers, dispersed throughout the sediment, and probably do not act as a gas trap.⁵ Hydrates have also been observed as a massive zone of pure crystal.

Natural gas hydrates occur in two basic structures. The structure I hydrate is associated with the lower molecular weight gases such as methane, ethane, carbon dioxide, and hydrogen sulfide, and may be formed from the noble gases such as xenon and argon. The higher molecular weight hydrocarbons form a different lattice (structure II). In biogenic production of natural gas in marine sediments, low molecular

weight methane is the primary product and hence biogenic gas will form structure I hydrate crystals. Thermogenic gases are composed of a wide variety of hydrocarbons and will thus form both structure I and structure II hydrates in sediments. An analysis of the gas content of hydrate crystals is useful in revealing the origin (biogenic or thermogenic) of the hydrate zone, as well as indicating the dominant lattice structure.

Preliminary analyses of gas content of the hydrates recovered on DSDP Leg 84 reveal that they are of a biogenic origin, rather than resulting from upward migrating thermogenic gases.⁶

Among the physical properties observed in situ in sediments containing hydrates are increased compressional velocities, reduced thermal conductivity, and increased electrical resistivity. A limited set of laboratory measurements of compressional velocities of hydrated sediments by Stoll and Bryan⁷ observed compressional velocities as high as 2.8 km/sec in sediments expected to have velocities between 1.6 km/sec and 1.8 km/sec in the absence of hydrates. Recent borehole sonic logs⁸ of a massive hydrate zone revealed compressional velocities as high as 3.8 km/sec within the hydrated sedimentary sequence.

A number of questions concerning the acoustic behavior of hydrated sediments remain to be answered. Some of these questions are:

- (1) How does the compressional velocity of hydrated sediment change with changing sediment type and concentration of hydrate within the sediment hydrate within the sediment?
- (2) What are the corresponding shear velocities of hydrated sediments?
- (3) What are the compressional and shear wave attenuations of hydrated sediments?

- (4) How are these and other parameters affected by changes in hydrate concentration within the sedimentary sequences?

III. EXPERIMENTAL APPARATUS

In FY82, ARL:UT designed an apparatus for use in addressing the questions concerning the acoustical behavior of gas hydrate bearing marine sediments. The design of the experiment is based upon several considerations. Pressures appropriate for safe and efficient work under laboratory conditions require the use of gases (or mixtures of gases) which have relatively low dissociation pressures. The dissociation pressure of a gas in a hydrate, which can also be considered as the vapor pressure of the hydrate, is the value when the hydrate is in equilibrium with water and the gas. Xenon was chosen for structure I hydrate gas because of its low dissociation pressure (1.5 atm) and its nonreactive character. Propane was chosen for the structure II hydrate because of its low dissociation pressure (1.74 atm).

As a result of discussions with previous researchers,⁹ it was determined that the procedure for growing hydrates was the most difficult aspect of the experiment and should be developed prior to concentrating on acoustical measurements. It was also realized that the pressure vessel should be constructed of a material that could be adapted as the experiment proceeds. The material chosen was acrylic, which is relatively easy to adapt and which meets the low pressure requirements of the experiment.

The apparatus is shown schematically in Fig. 1. Detailed engineering drawings have been included in the Appendix. The acrylic pressure vessel will be slightly larger than the apparatus used by Stoll and Bryan⁷ in their earlier experiments. The pressure vessel will be held at a constant temperature (2-4°C) above the freezing point of water to prevent the formation of ice. Gas is introduced from a 600 psi

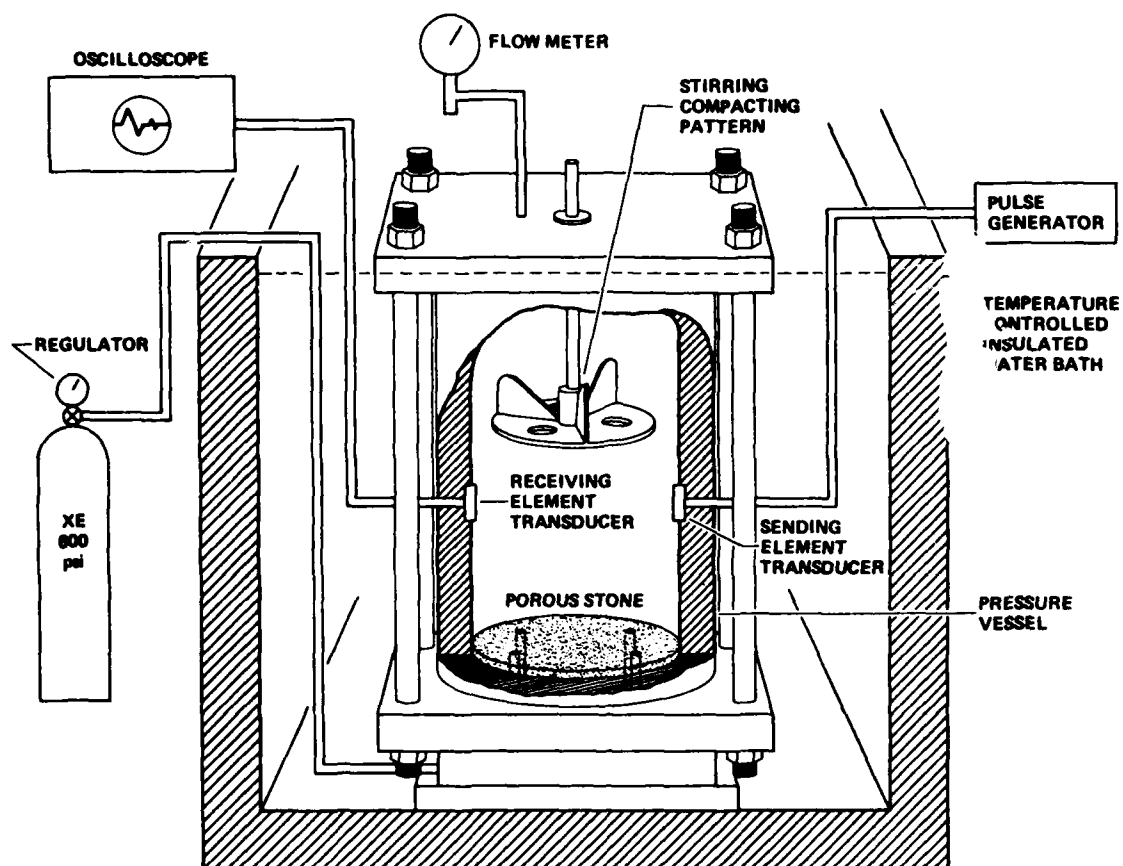


FIGURE 1
SCHEMATIC REPRESENTATION OF HYDRATE
EXPERIMENTAL APPARATUS

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cylinder through a regulator into four ports in the bottom of the pressure vessel. Above the ports is a porous stone, which will enable the equal diffusion of gas into the chamber containing water saturated sediment. The gas will be allowed to diffuse through the sediment while being stirred with the platen. Gas will leave the vessel through a port at the top, and the column will be monitored with a calibrated flowmeter.

Acoustical transducers will be mounted on the inside of the pressure vessel chamber next to the walls. A number of mounting and transducer configurations will be tried to maximize coupling to the hydrate and to minimize coupling to the apparatus. The transducers will be similar to those developed earlier at ARL:UT³ and will be capable of exciting and receiving both shear and compressional waves.

The ARL:UT design is based, in part, on the use of xenon gas, which has the advantage of having a low dissociation pressure (1.5 atm) for the structure I xenon hydrate crystals. The acrylic pressure vessel will also withstand pressure (1.74 atm) suitable for forming propane gas structure II hydrate crystals. Xenon and propane form the structures of hydrate crystals that are found in marine sediments.

While safety and convenience led to the choice of low pressure xenon and propane gases, the vessel is capable of safely handling pressures in excess of 14 atm. This will allow, in the final stages of experimentation, the introduction of gas mixtures in order to vary the pressure-temperature conditions through a wider range. While the apparatus will not confine the nearly 30 atm needed to form pure methane type I hydrates at about 0°C, it will support the formation of hydrates from a 98% methane and 2% propane mixture at about 14 atm. In the initial phase of growing hydrate crystals and measuring their properties, xenon gas will be used. This will avoid potentially dangerous concentrations of hydrocarbon gases while procedures are being developed.

IV. EXPERIMENTAL PLAN

The experiment will be performed in three phases. First, the apparatus will be constructed and the procedure for growing hydrates in sediments will be developed. Discussions with previous investigators⁹ indicated that the most difficult aspect of the experiment will probably be actually growing hydrates. In this first phase, the goal will be to vary pressure-temperature conditions, as well as rates of introduction of gas, in order to optimize the nucleation and growth of hydrate crystals. Initially, growth in pure water will be attempted, followed by introduction of a standard medium sand, 20-30 Ottawa sand, which has been used successfully in growing hydrates.⁷ Emphasis at this stage will be on observing the effects of stirring rates of the compacting platen and of the rate of introduction of gas into the chamber. Direct visual and microscopic examinations of the physical structure of hydrated sediments will be made for use in developing a composite media theory of propagation in hydrates. This will be accomplished by installation of an optical port in the wall of the vessel.

In the second phase of the experiment, the apparatus will be adapted for acoustical measurements. Acoustical transducers will be installed and procedures developed to measure shear and compressional velocities. The modifications suggested during the first phase will also be implemented to optimize hydrate growth. Preliminary acoustical measurements will be made and the results submitted for publication.

In the third phase, more extensive acoustical measurements will be taken in which the sediment type, pressure-temperature conditions, and hydrate concentrations will be varied. During this phase, the system

will be adapted to more directly investigate hydrate growth in sediments. Hydrocarbon gases, such as propane, which occur in marine sediments will be used. Measurements comparing the acoustical behavior of the structure I hydrated sediments (xenon) with that of the structure II hydrated sediments (propane) will enable a determination of the differences between hydrate zones of biogenic origin and those of thermal origin. Special attention will be devoted to studying the acoustical variability of sediments differing in type and size as they are hydrated with varying concentrations of hydrate crystals.

V. SUMMARY

One feature that is rarely included in geoacoustic profiles of the ocean floor is the presence of natural gas hydrates in marine sediments. Hydrates are icelike crystalline structures that can significantly alter geoacoustic parameters such as shear and compressional velocities and absorption. The acoustic behavior of hydrated marine sediment can differ greatly from that of nonhydrated sedimentary sequences. Recent calculations indicate that bottom loss from hydrated marine sediments is particularly sensitive to the as yet unknown value for shear velocity.

Potentially favorable conditions for the formation of hydrate zones exist over 90% of the ocean floor and to a considerable depth in the sediment. Recent evidence indicates the presence of massive quantities of gas hydrates in a variety of sedimentary sequences.

In FY82, ARL:UT designed an experimental apparatus and developed an experimental plan to measure the acoustical behavior of marine sediments bearing gas hydrates. The goal of the experimentation is to obtain values for shear and compressional velocities in a variety of hydrated sediments. The apparatus consists of an acrylic vessel designed to contain sediments and gases under pressure-temperature conditions for the formation of gas hydrates. Xenon gas will be used initially to form the structure I gas hydrate, followed by the use of propane gas which forms the structure II hydrate. Both of these structures occur in marine sediments. The low dissociation pressures of xenon hydrates (1.5 atm) and propane hydrates (1.74 atm) will enable a safe and efficient performance in the laboratory environment. The vessel is designed for installation of instrumentation capable of measuring the acoustical, thermal, and electrical properties of gas hydrates in sediments.

The experiment will be conducted in three phases. In the first phase, the apparatus will be constructed and the procedure for growing hydrates will be developed. In the second phase, any modifications suggested during the first phase will be made and the apparatus will be adapted for acoustical measurements. Procedures for measuring the acoustical parameters will be developed and initial results obtained. In the final phase, an extensive set of measurements will be taken in which the sediment type, pressure-temperature conditions, and hydrate concentrations are varied over a wide range in order to determine their influence on the acoustic parameters.

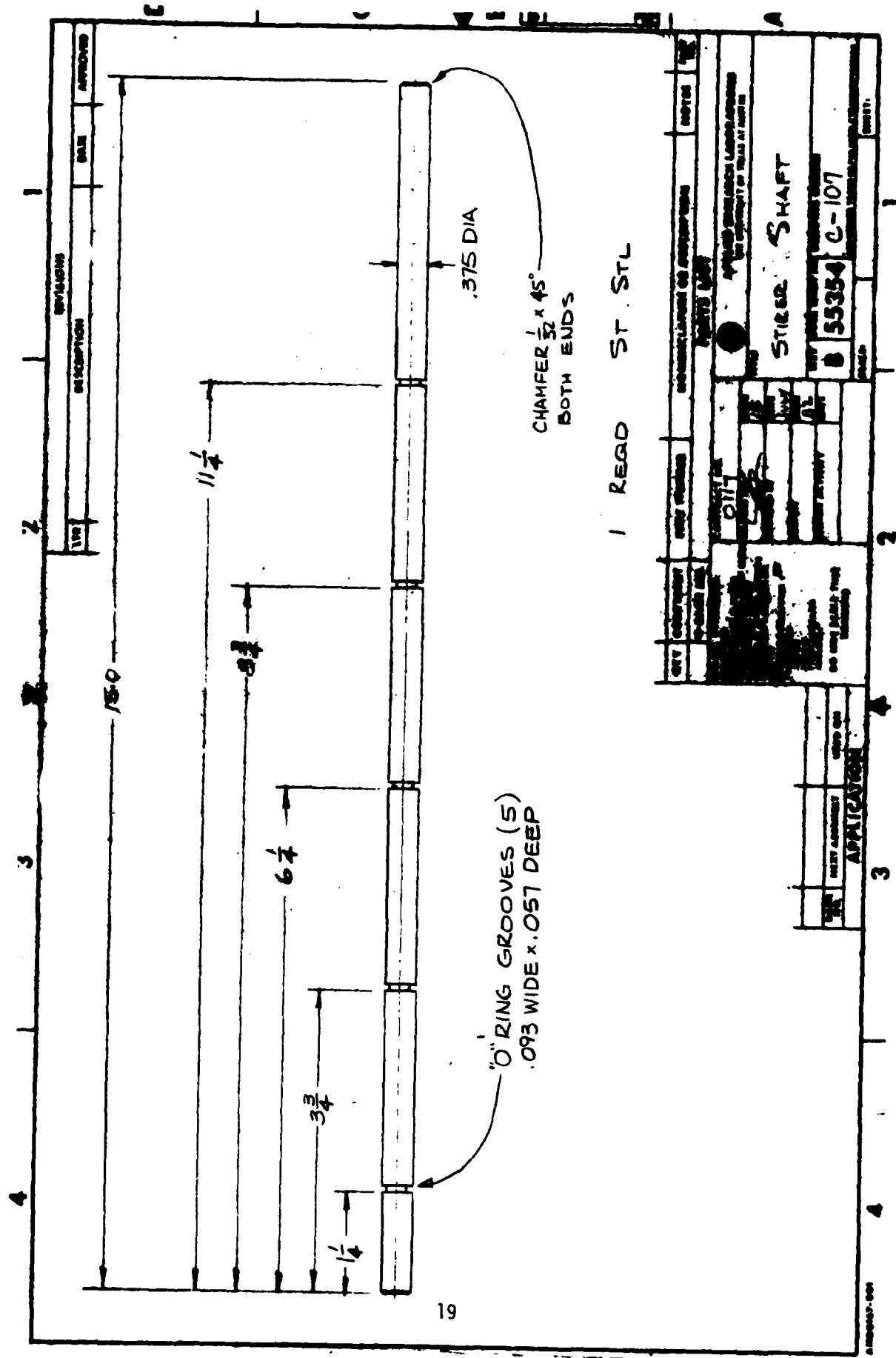
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APPENDIX

ENGINEERING DRAWINGS FOR ARL:UT
HYDRATE EXPERIMENTAL APPARATUS

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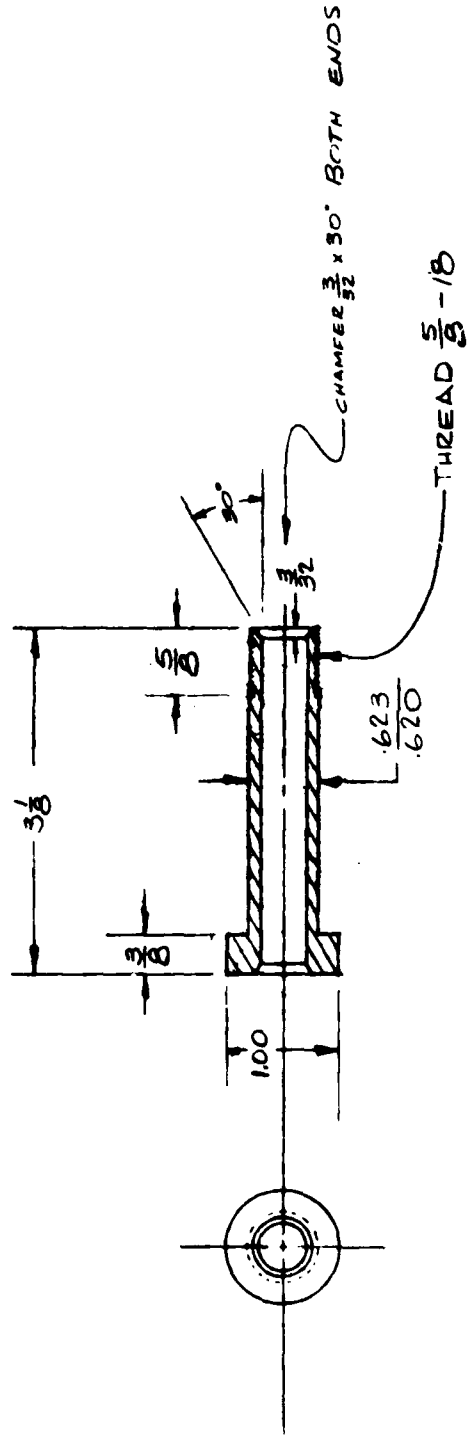
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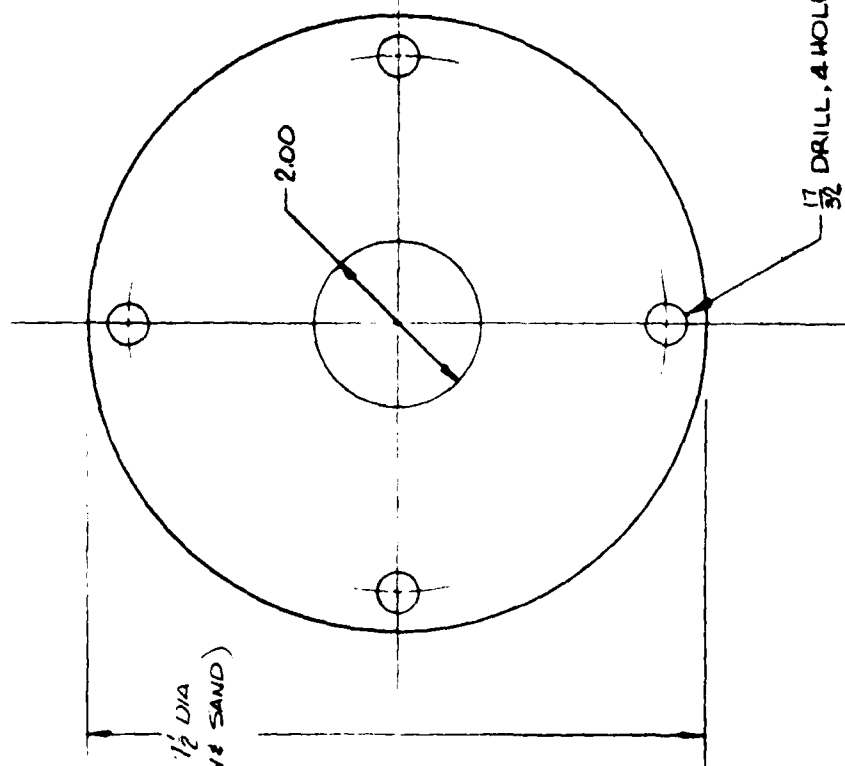
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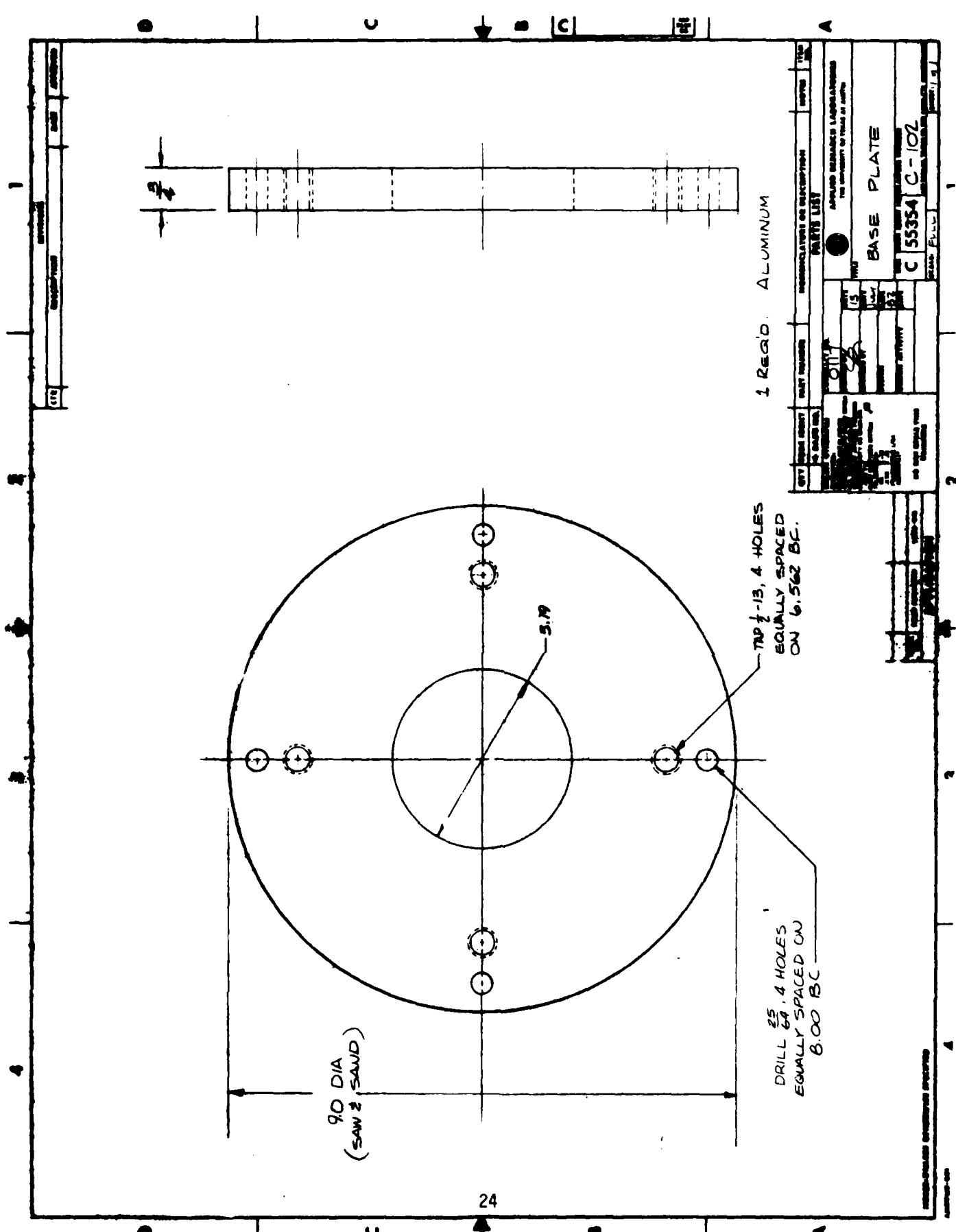


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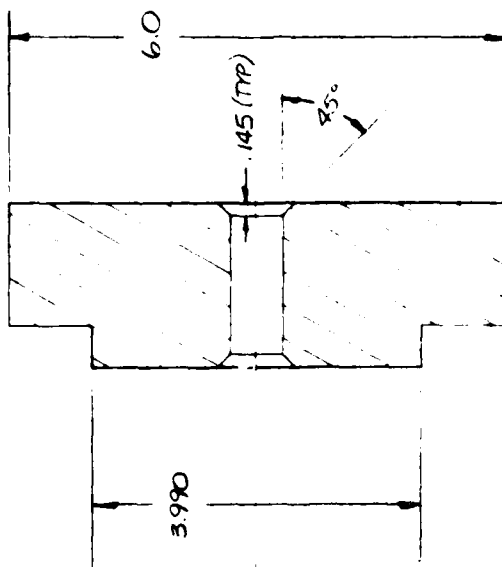
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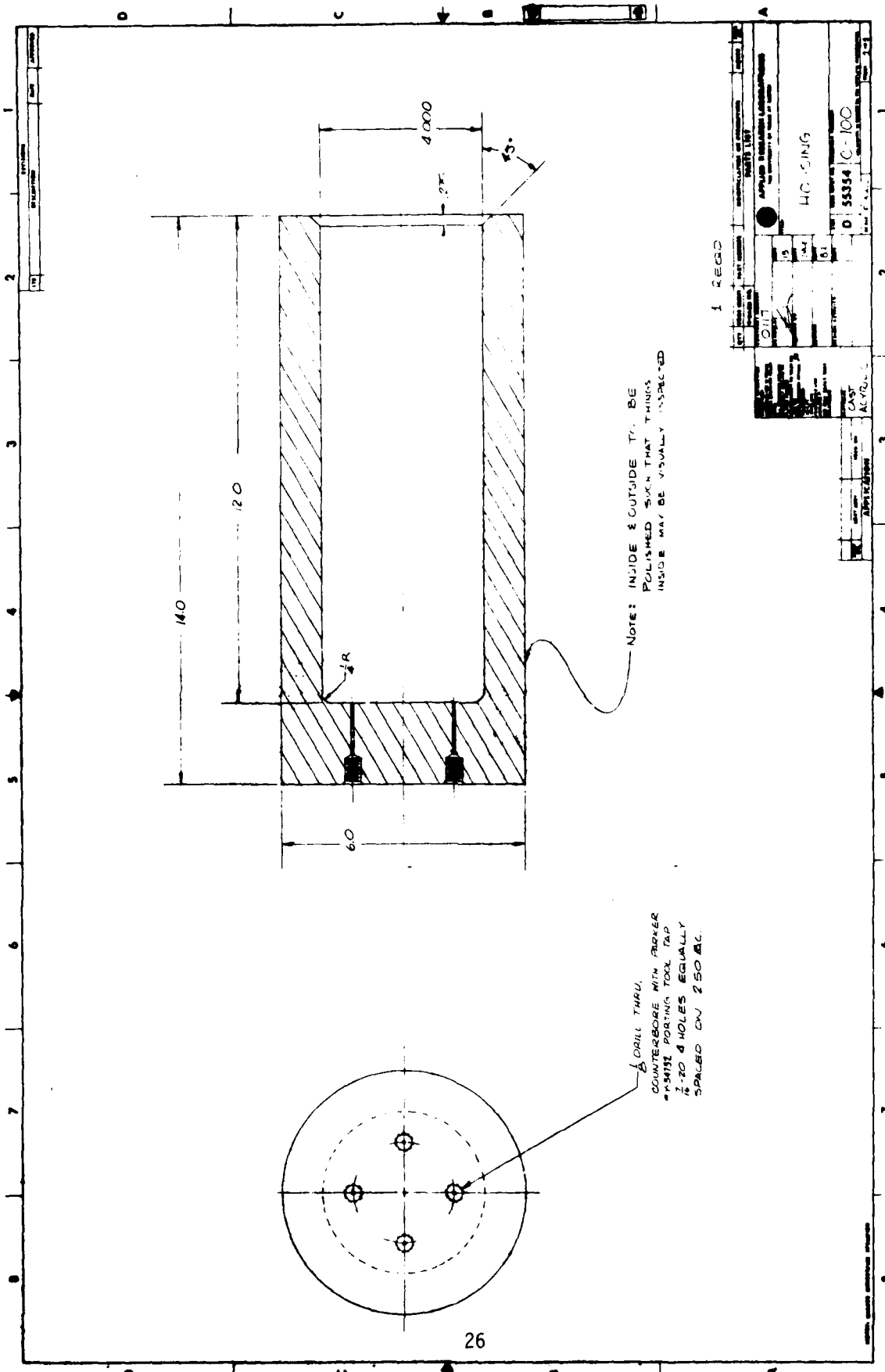
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