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DESIGN CONSIDERATIONS FOR A SYSTEM TO INVESTIGATE SHORT-LIVED NUCLEI PRODUCED AT A REACTOR

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Abstract: A system for the precision study of the nuclear decays of short-lived nuclei produced by neutron irradiation is described. The system as envisioned will employ continuous introduction of irradiated material into the ion source of an electromagnetic isotope separator and subsequent "on-line" operation of the separator in connection with nuclear decay analysis apparatus. The results of an empirical study are presented regarding the characteristics of a vapor transport line used for the extraction of irradiated material from the vicinity of the reactor core and transport to the isotope separator ion source. Of particular interest in this study are the transit times achieved with the system and properties of the system associated with the presence of electrical discharge in the vapor transport line. Extensions of the results of this study to an actual system are discussed.

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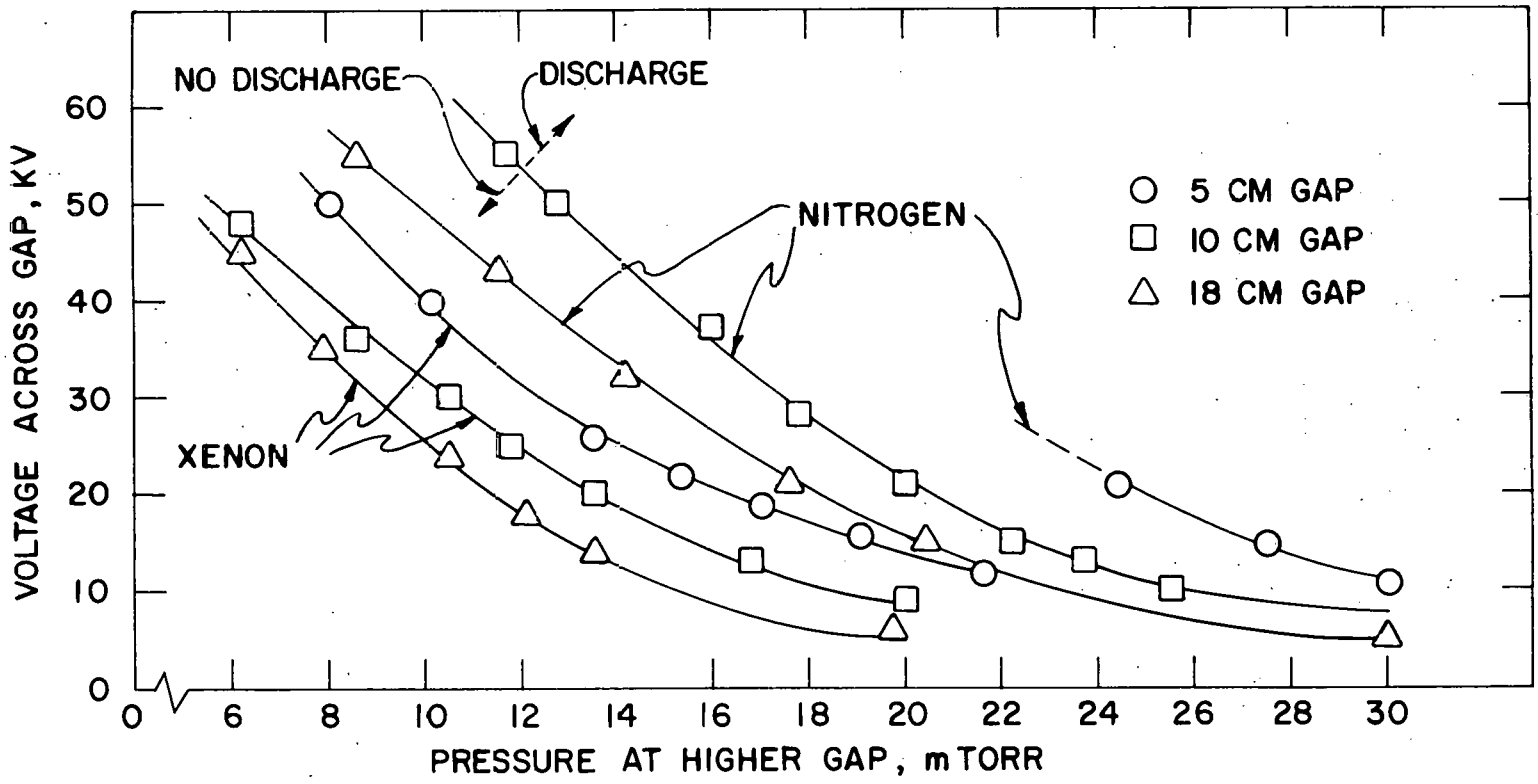
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1. Introduction

At the Ames Laboratory Research Reactor, one of the major experimental facilities being developed is intended to make possible precise and detailed analysis of the nuclear decays of short-lived radioisotopes with half-lives down to the order of one second. In the reactor it is possible to produce many such activities through various neutron activation processes, including fission. With the exceptions of (n, p) reaction products, all such activities differ in mass from the parent atoms; thus, they can be separated in an electromagnetic isotope separator from the parent and also from possible interferences resulting from competing reactions.

Although the recently-developed techniques of nuclear decay analysis employing semiconductor detection devices have made possible investigations of short-lived nuclei which previously could not be approached satisfactorily, to date few of the activities in the half-life region between one and 180 seconds have been studied with high-resolution analysis systems such as high-resolution beta-ray spectrometers. Such analysis is very difficult since the source preparation requirements become quite stringent for the high-resolution study of electron spectra, and conventional source preparation techniques result in lengthy processes which then prohibit the careful study of short-lived beta activities.

The system described below has been conceived as an attempt to extend the half-life range of detailed nuclear spectroscopy of reactor-produced activities. A description of the experimental arrangement is presented, followed by a study of the important features of a vapor

transport line which will be employed to extract irradiated material from the reactor for introduction into the remainder of the system.

2. Proposed Experimental Arrangement

The system being developed at the reactor to provide the capability of analyzing short-lived nuclear decays is outlined in fig. 1. A sample containing the parent isotope from which the desired activity will be produced is placed in the neutron flux near the core of the reactor at the end of a tube 1.27 cm in diameter and about 4 m long. This tube will transport molecules of parent and activated material (at saturated activity) from the reactor to the ion source of an electromagnetic isotope separator. It is proposed that heating elements surround the tube to make it possible to vaporize many samples which are solid at room temperature and transport the activity in the vapor state.

The philosophy used throughout the conception of the system has been to use conventional techniques where possible. Thus, the electromagnetic isotope separator is of the Scandinavian type[1-4]) which was obtained from Nuclear Engineering and Equipment S. A. of Geneva, Switzerland, and was constructed at the Nobel Institute of Physics, Stockholm, Sweden. Following the separator is an ion beam switching magnet system obtained from Magnion, Inc., Burlington, Massachusetts. The purpose of the switching magnet is to permit directing the separated ion beam to one of five experimental arrangements and to refocus the beam in the ± 45° deflection directions with a deflection radius of 45 cm. At one of the 45° deflection

ports, a high-resolution beta-ray spectrometer will be situated such that the ion beam is deposited at the source position of the spectrometer. The spectrometer is a magnetically-shielded double-focusing type and follows the design of Bartlett[5]). At other ports of the switching magnet will be located nuclear radiation detector systems, as yet not specified, but designed for specific investigations of activities of interest. At the time of writing, the isotope separator and switching magnet system have been tested as an ion optical unit and appear to perform as desired.

A study of the activities available by neutron irradiation which are amenable for study with the system being developed reveals a virtual plethora of alternatives. Of particular interest are fission products or activities produced by other neutron reaction processes resulting in proton and neutron number regions away from the magic numbers. The activities which can be studied with the system being developed are limited in principle by the current capacity of the isotope separator, for given activation conditions (neutron flux, activation cross-section, and decay constant). An equivalent activity of at least 150 μ Cur is available under the present current limitation of the separator (imposed by conditions to give excellent focus), even for a ratio of activated-to-parent abundance of 10^{-8}.

The isotope separator current will be adjusted to give a "steady-state" activation product deposit which is replenished at the rate of decay, according to the common ideas of "on-line" operation. In this manner, sophisticated analysis can be made of the nuclear decays without regard to limitations of analysis imposed by half-life. Several advantages of the

scheme are apparent: the radioactive deposit will be isotopically pure, of high specific activity, and of ideal thickness for high resolution studies.

The portion of the proposed arrangement for which there is the least past applicable experience is the transport line through which vapors of activated material are extracted from the reactor and introduced into the isotope separator. Since this portion of the system represents the major development project in the proposed system, the balance of the discussion presented here will be concerned with the characteristics of such a transport system.

3. Properties of the Vapor Transport Line

3.1 Character of Vapor Flow in the Transport Line

Under the concepts of the system being developed, it is necessary to transport activated vapor from the core of the reactor to the ion source of the isotope separator. Since the ion source of the separator operates in the pressure region of about 1 mTorr, it is logical to consider a transport line which terminates at the ion source with this pressure. A study[6] was made of the vapor flow characteristics in a tube simulating the transport line, observing the flow of gases at room temperature.† The results of this study can be applied to other vapors if (1) the transport tube is heated sufficiently to prevent condensation of vapor; (2) the sample is heated to establish the necessary pressure at the reactor end to provide the required flow of vapor down the line with the proper pressure

†For details of the study, the reader is referred to ref. [6]. The essential results of the study are presented in the present work.

maintained at the ion source end; and (3) if there is no other process taking place; e.g., chemical reactions between the vapor and the walls of the transport line. The most interesting feature of the vapor flow is the time required for transport of the vapor to the isotope separator ion source, and in order to provide a basis upon which the transport time can be calculated, it was first necessary to determine the nature of the flow of vapor down the line.

A vacuum system was built to simulate the transport line, including connection to the ion source of the isotope separator. A study of the expected flow through the system was made using the assumption of molecular flow. It was determined that for the dimensions of the line, and using an exit hole diameter of 1.5 mm (assuming the flow through the line was the result of pumping through the exit hole), a pressure of 4 mTorr would be required at the reactor end of the transport line to maintain a pressure of 1 mTorr at the ion source end. Under these conditions, the mean free path of the molecules is indeed not much greater than the characteristic dimensions of the transfer line (e.g., the diameter), and thus the assumption of free molecular flow is not completely justified.

To determine what effect any deviation from molecular flow has on the actual nature of the flow, measurements were made of the pressures on each end of the transport line as a function of the mean pressure in the system, as shown in fig. 2. For pure molecular flow, the ratio of the pressures at each end (P_1 refers to the pressure at the reactor

end of the line, P_2 to that at the ion source end) is a function of the geometry of the flow line alone, and hence should be a constant, independent of the mean pressure in the system. As can be seen from the data plotted for nitrogen, there is no significant variation from what is to be expected from the molecular flow prediction. For xenon, however, a significant deviation is apparent for mean pressures above about 2 mTorr. The solid curve passing through the points is calculated for xenon using the viscous correction for conductance by Knudsen[7]). It is apparent, then, that although the situation is surprisingly well described by molecular flow, there are small, but significant, viscous effects which may be important in the determination of transit time through the line.

3.2 Transit Time

In using the vapor transport line, it is important to know the time required for vapor to be transported down the line. The vacuum system employed above was adapted to make measurements of this time, as a function of equilibrium flow conditions. The technique employed was to introduce at the reactor end a small pressure pulse superposed upon the equilibrium flow and to record the arrival of this pressure pulse at the ion source end.

Under the assumption of molecular flow, a δ-function pressure pulse superposed upon equilibrium flow will have the appearance of fig. 3 after the molecules involved in the pressure pulse have been

transported to the opposite end of the transport line. The most probable time of arrival, denoted t_{peak}, is a characteristic time of the flow conditions and shall be referred to in subsequent discussion as the "transit time". The curve presented in fig. 3 is calculated, and the corresponding experimental results for nitrogen are shown in fig. 4, which illustrates typical recorded responses from the pressure transducer at the ion source end of the transport line. The curve f(355, t) is a calculated time-dependent response after a transport of 355 cm using the assumptions of molecular flow. Curves #1 and #2 were recorded with a mean pressure in the transport line of 0.23 mTorr, while the maximum response at the reactor end (where pressure pulse was introduced) was 0.8 mTorr and 2.7 mTorr, respectively. As can be seen, there is a slight effect on the tail of the time response as viscous effects are increased, but the transit time is unaffected by moderate disturbances to the equilibrium flow. Curve #3, on the other hand, is the response at a mean pressure of 4.8 mTorr, and the pressure pulse peak was higher than 10 mTorr, the maximum calibrated response of the gauges used. With these conditions, the nature of the equilibrium flow is clearly intermediate between free molecular and viscous, so the viscous corrections to the molecular transit time are significant.

The transit time desired is that obtained as the pressure pulse is made arbitrarily small--the time resulting is then a true measure of the transit time for equilibrium flow. Experimental values of the transit time were measured for a variety of equilibrium flow conditions,

and the results are shown in fig. 5, plotted as a function of peak ion source pressure. The results are presented for both xenon and nitrogen. As can be seen from the nitrogen data, so long as the total pressure at the ion source end of the transport line remains below about 3 mTorr (satisfying the conditions for ion source operating pressure), the transit time is independent of the size of the exit hole and is apparently not strongly dependent upon the exact nature of the flow. However, as the mean pressure in the system is increased to introduce significant viscous effects, the transit time is decreased, as shown by the dashed line.

The results shown for xenon show very strongly the effects of viscous flow, and the dashed line represents the locus of transit times measured with different equilibrium flow mean pressures. As expected from the results of fig. 2, only at low mean pressures does the transit time approach a constant value in accordance with molecular flow.

It is interesting to note that the transit time is actually decreased as viscous effects are first significant in the equilibrium flow. This can be explained by noting that as the mean free path becomes comparable to the dimensions of the transport line, a significant number of collisions are no longer diffused reflections from the tube wall, but are collisions between two molecules in which their total momentum is conserved. Since the net flow of molecules is down the tube in the direction opposite the particle density gradient, there is a net momentum in this direction. Thus, if there occurs some momentum conservation for the molecules, it is predominately in this direction, and the transit time will be decreased compared to pure molecular flow. This argument is, of course, limited to pressures

well below those for which the flow is purely viscous, for in the latter case, the transit time for a molecule will be determined by the time required for all molecules in front of it to precede it through the exit hole.

Using the molecular flow assumptions, however, the transit time is expressed by

$$t_{peak} = \frac{3x^2}{2D} \left[\frac{\pi m}{8kT} \right]^{\frac{1}{2}} , \qquad (1)$$

where x is the length of the tube, D is the diameter of the tube, m is the mass of the molecule, and T is the absolute temperature. Calculation for the transit time in the experimental system yields the results 3.1 sec for nitrogen and 6.7 sec for xenon, which agree very well with the observed transit times for low mean pressures shown in fig. 5. It is thus to be concluded that the molecular flow treatment of the dynamic characteristics of the equilibrium flow down the transport tube results in an excellent representation of the actual flow for the pressure region of interest.

3.3 Electrical Discharge

Under the conditions of the proposed system, the transport line is most suitably arranged with its two ends at different electrical potential, namely the reactor end at ground potential and the ion source end at the acceleration potential of the isotope separator. Since the mean pressure in the transport line is of the order of a few mTorr, this situation results in an exceedingly favorable condition for electrical discharge through the length of the tube. Although the general problem of electrical discharge has been throughly treated, there was found to be no information in

existence for the combination of geometry and pressure region of interest in this study. Hence, an empirical study was made to determine what configuration could be employed to eliminate electrical discharge in the transport line.

A section of the transport tube was replaced by another section employing an isolated conducting tube with pyrex glass spacers at each end. A positive potential of 0 - 60 kV was placed on the isolated portion of the transport line and adjusted with equilibrium flow conditions until a discharge was evident. It was found that for a given length of pyrex spacer the onset of the discharge was a sharp and reproducible function of the pressure in the higher-pressure gap. The results of the study for xenon and nitrogen gas flow are shown in fig. 6 with the observed differences between the gases correlated to the differing ionization potentials. As can be seen, a 10 cm gap appears to be sufficient to suppress any discharge if the gap occurs near the ion source, where the pressure will be about 1 mTorr. Several measurements of transit time were repeated with the gradient imposed, and no effect was noticed, so long as there was no discharge.

It appears, then, that as a result of this study it will be possible to suppress electrical discharge in the transport line in a rather simple fashion. The actual effect of the small gap has been to make the gradient restricted to a distance which is not much greater than the mean free path of the molecules in order to avoid ionizing collision avalanches between the molecules and background ionization electrons.

4. Conclusions

From a consideration of the properties of the vapor transport line which was simulated in the above studies, it appears that there is a direct application of the results of these studies to the design of the high-temperature vapor transport line desired under the conditions of the system being developed. In particular, the measurements of transit time confirm that the system can be used to study the decays of reasonably short-lived activities. It is difficult without further study to determine several important effects which are unique to the transport of radioactive vapors. For example, the materials of construction should be carefully chosen to avoid any change in the chemical designation (and thus the vapor characteristics) of the molecules undergoing transit. The effects of different chemical elements in the same transit line (for example in fission product transport) are difficult to determine without further study. Finally, the character of electrical discharge suppression may be changed with the presence of radioactive atoms in the field gradient region.

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FIGURE CAPTIONS

Fig. 1. Layout of system to study short-lived nuclei.

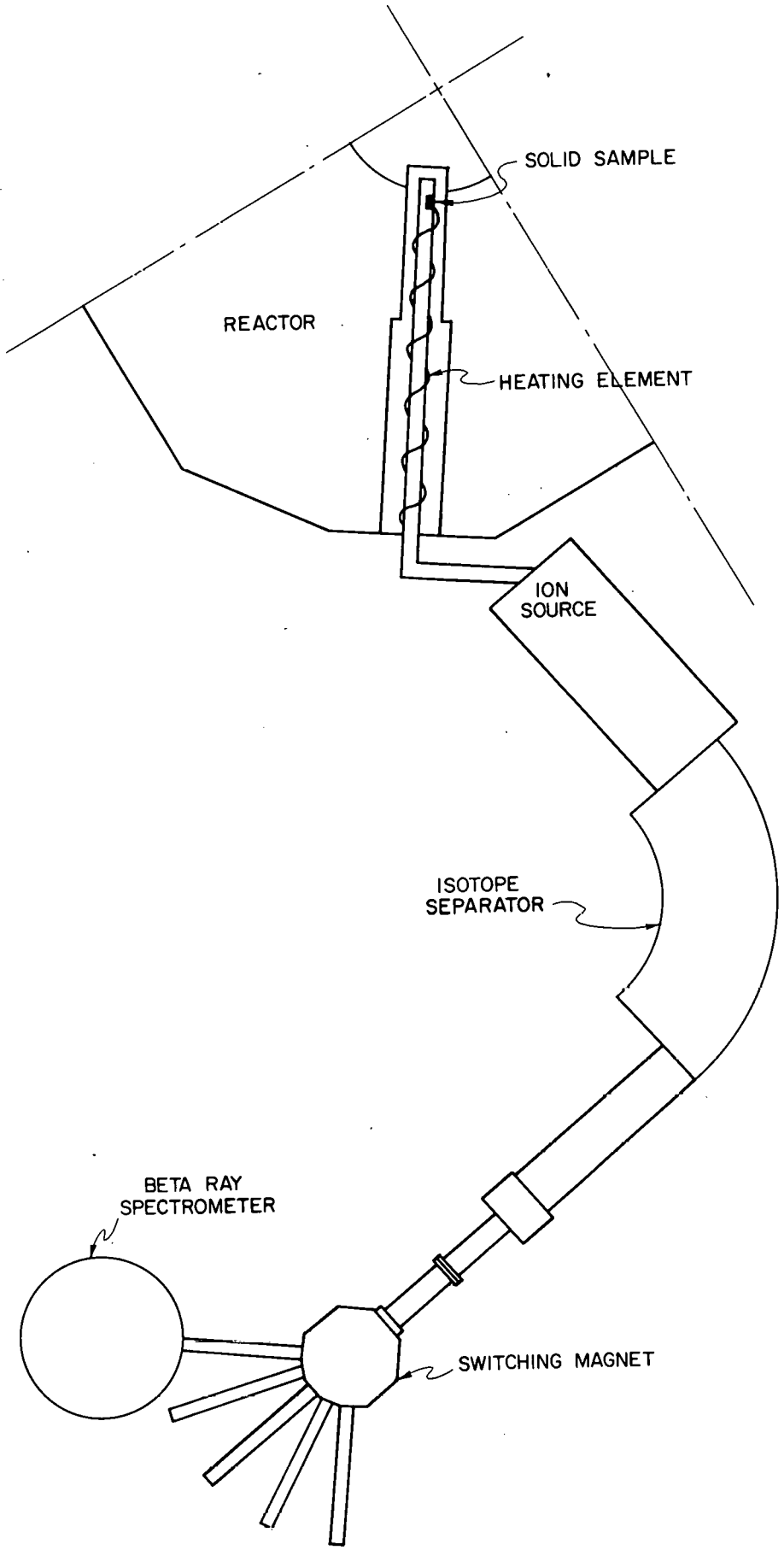
Fig. 2. Pressure ratios for flow in transport line.

Fig. 3. Time response of pressure pulse at ion source end (nitrogen).

Fig. 4. Measured pressure responses at ion source end (nitrogen).

Fig. 5. Transit time as a function of mean pressure.

Fig. 6. Gap voltage vs. pressure at onset of discharge.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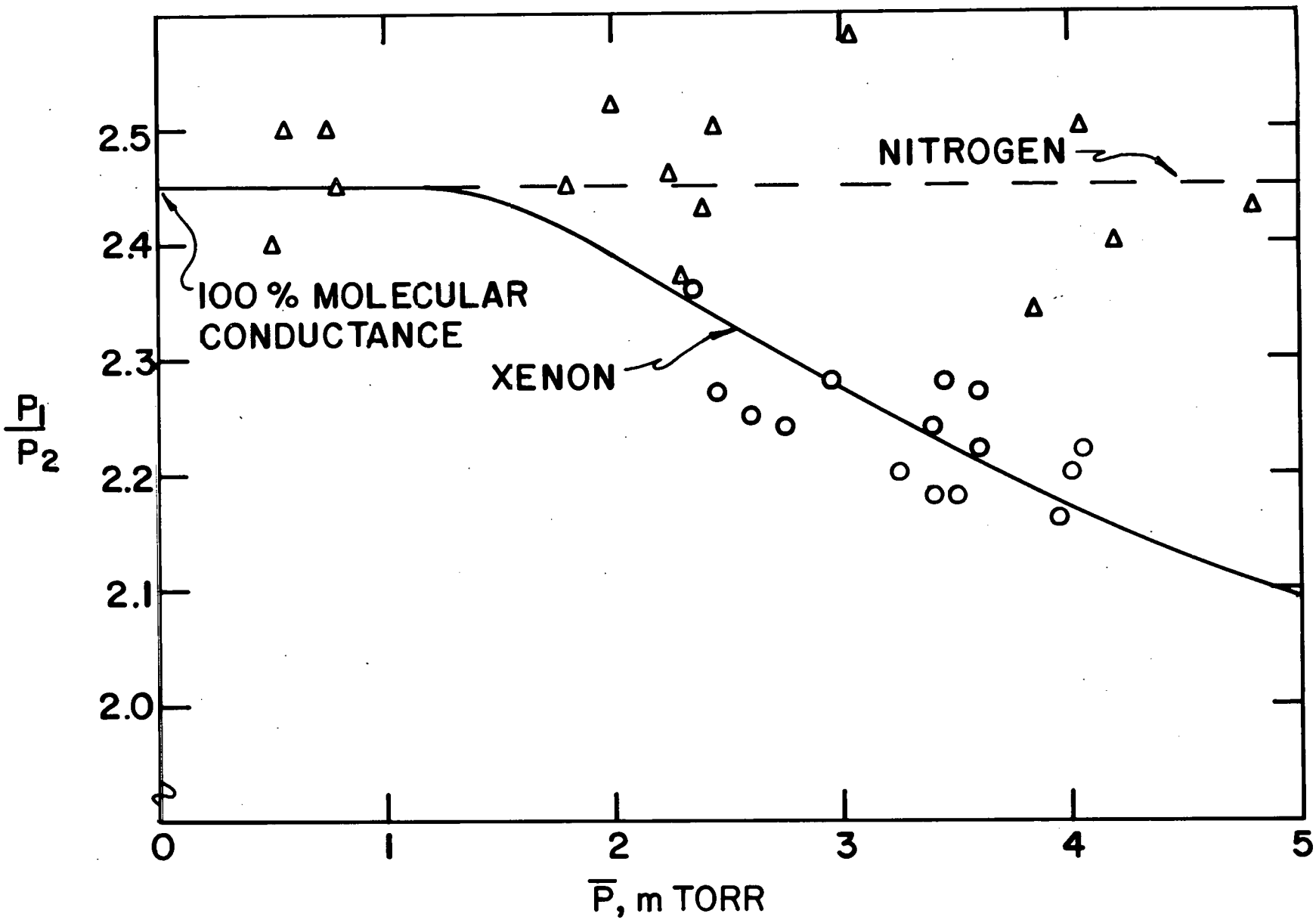
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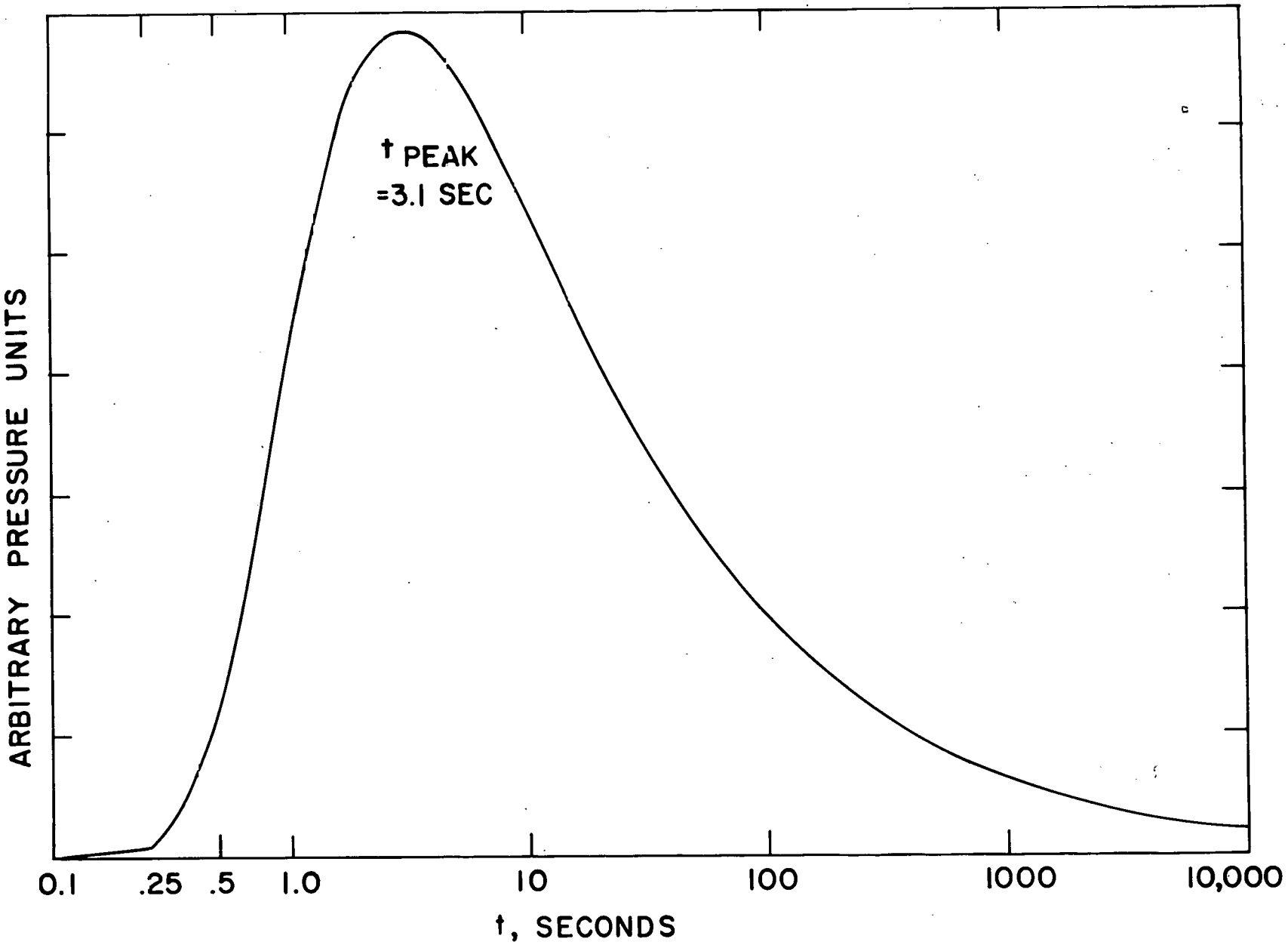
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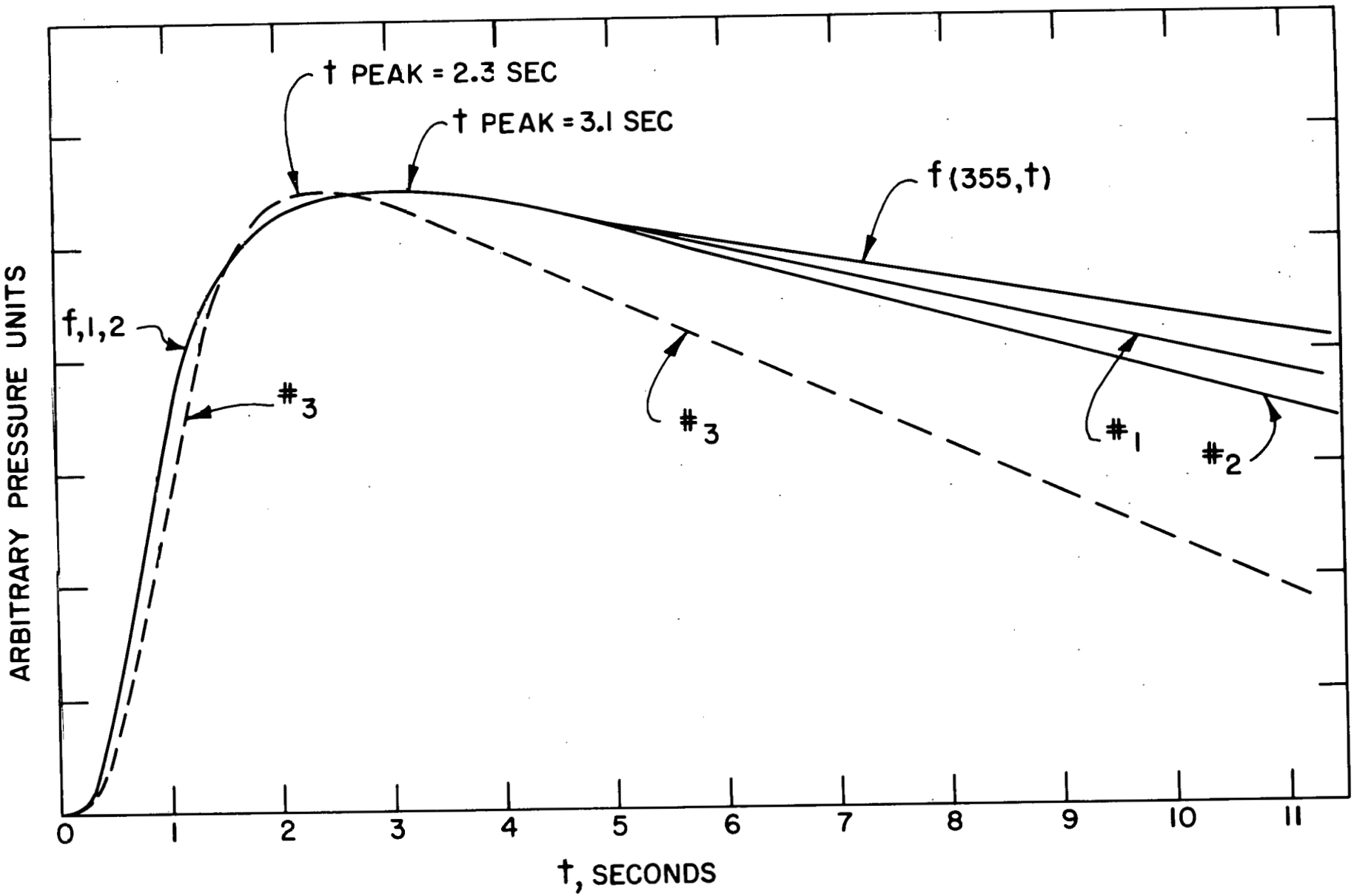
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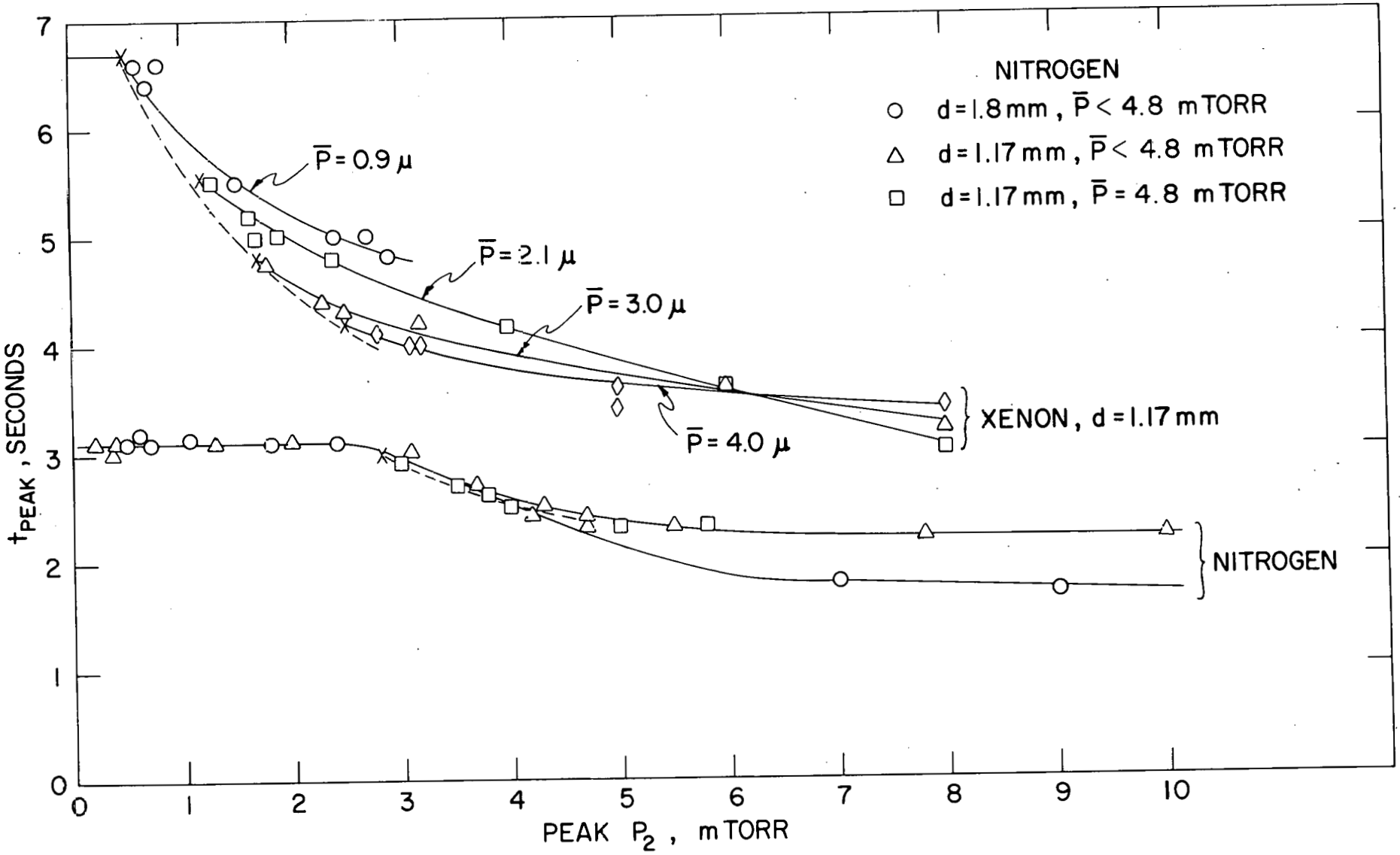
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