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THE FISSION GAS PROBLEM FOR MOBILE FUEL FAST REACTORS

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INTRODUCTION

In reactor systems utilizing fixed, solid fuel elements the dimensions and permeability of the fuel elements are such that only a very small fraction ($< 1.0\%$) of the gaseous fission products reach the surface and these are generally confined either by a clad or a sealed can. In these types of reactors, the significant fission gas problems are the swelling of the fuel elements and the contamination of the coolant or cover gas system due to fuel element failure. Of these two problems, the former is by far the more serious. One of the major motivations for the development of mobile fuel systems has been the avoidance of the problem of radiation damage in solid fuel elements. However, mobile fuel systems for fast power reactors may inherit several fission gas problems which are unknown in the fixed, solid fuel systems. Fast power reactors inherently require much larger critical mass than thermal power reactors. Also, their critical mass increases significantly with increasing size of core (approximately proportional to the 2/3 power of the core volume). Hence, it is necessary that their cores be very compact relative to thermal reactors, and their power densities very high (~1,000 kw/liter of core volume vs. ~100 kw/liter of core volume in thermal reactors). The combination of high power density and the technical limitations on heat transfer and heat transport results in fast power reactor core designs having extremely narrow fuel ligaments and very high wall heat fluxes. In the projected mobile-fuel fast power reactors the presence of fission gas bubbles in the fuel could seriously affect both heat transfer and reactor stability.

The fission product gas problem in fast power reactors has been investigated for two types of mobile fuel systems; namely, a molten alloy fuel system of the type considered by Los Alamos Scientific Laboratory for the Molten Plutonium Reactor Experiment(1), and a paste fuel system of the type being studied by Atomic Power Development Associates for its Paste Reactor Concept.(2) Both systems involve internal cooling and sufficient fuel circulation to satisfy the fuel processing requirements. The core structure in each system is extremely compact with fuel ligaments of 50 to 100 mils hydraulic diameter to accommodate the high power densities. The molten fuel system considered herein is a molten plutonium alloy. The mobile fuel system consists of fuel alloy particles, 100 to 200 microns diameter, that are settled in sodium to form a paste of about 60 volume per cent solids. In both cases the core structure was assumed to be of the shell and tube type with fuel on one side and sodium coolant on the other. At the very high power densities of interest in fast power reactors the amount of gaseous fission products generated could be in the order of one fuel volume per day at the operating temperature and pressure. In the molten alloy fuel system, all of this gas must be accommodated by the fuel system; whereas, in the case of the paste fuel system, it is estimated (on the basis of the fission recoil distance in the fuel particles) that at least ten per cent of the fission gases will be released into the sodium carrier.

The possibility of removing the fission gases from these fuel systems before saturation is reached depends primarily on the solubilities of the gases in the fuel and on the rates at which the fuel can be cycled through a degassing process. To a lesser degree, it also depends on the power density (i.e., the specific power for a given fuel composition). Figure 1 shows the estimated time required to attain saturation of the fuel as a function of the fission gas solubility for the molten fuel system and the paste fuel system at two different values of the specific power. It is seen that even with an extremely low degassing cycle time of 0.1 minute the gas solubility required to prevent supersaturation is of the order of 10^{-8} mol fraction; and at a more practical degassing cycle time of 10.0 minutes the required gas solubility to prevent supersaturation is in the order of 10^{-6} mol fraction.

Estimates of the solubility of the noble gases in liquid metals have been made from a knowledge of certain physical and thermodynamic properties of both the solute and solvent. However, the extremely low solubilities of the noble gases in liquid metals makes their measurement very difficult. The estimated values exhibit wide disagreement, depending on the method that is used for making the estimates. For example, C. R. Mitra and C. F. Bonilla(3) used three different methods to estimate the solubility of xenon in bismuth at 300°C and 1.0 atmosphere, and they obtained three widely divergent values -- namely, 1.4×10^{-5}, 1.2×10^{-12}, and 7.8×10^{-30} mol fraction. W. G. McMillan(4) also estimated the solubility of xenon in bismuth at 300°C and 1.0 atmosphere, and obtained a value of 3×10^{-16} mol fraction. Mitra and Bonilla(3,5) determined the solubility of xenon in bismuth at 500°C by two different experimental methods, and obtained results that varied from about 10^{-5} to 10^{-8} mol fraction per atmosphere. C. J. Raseman et al.(6), at Brookhaven National Laboratory obtained data on the solubilities of the fission gases in bismuth at 500°C, and obtained an average value of about 8×10^{-7} mol

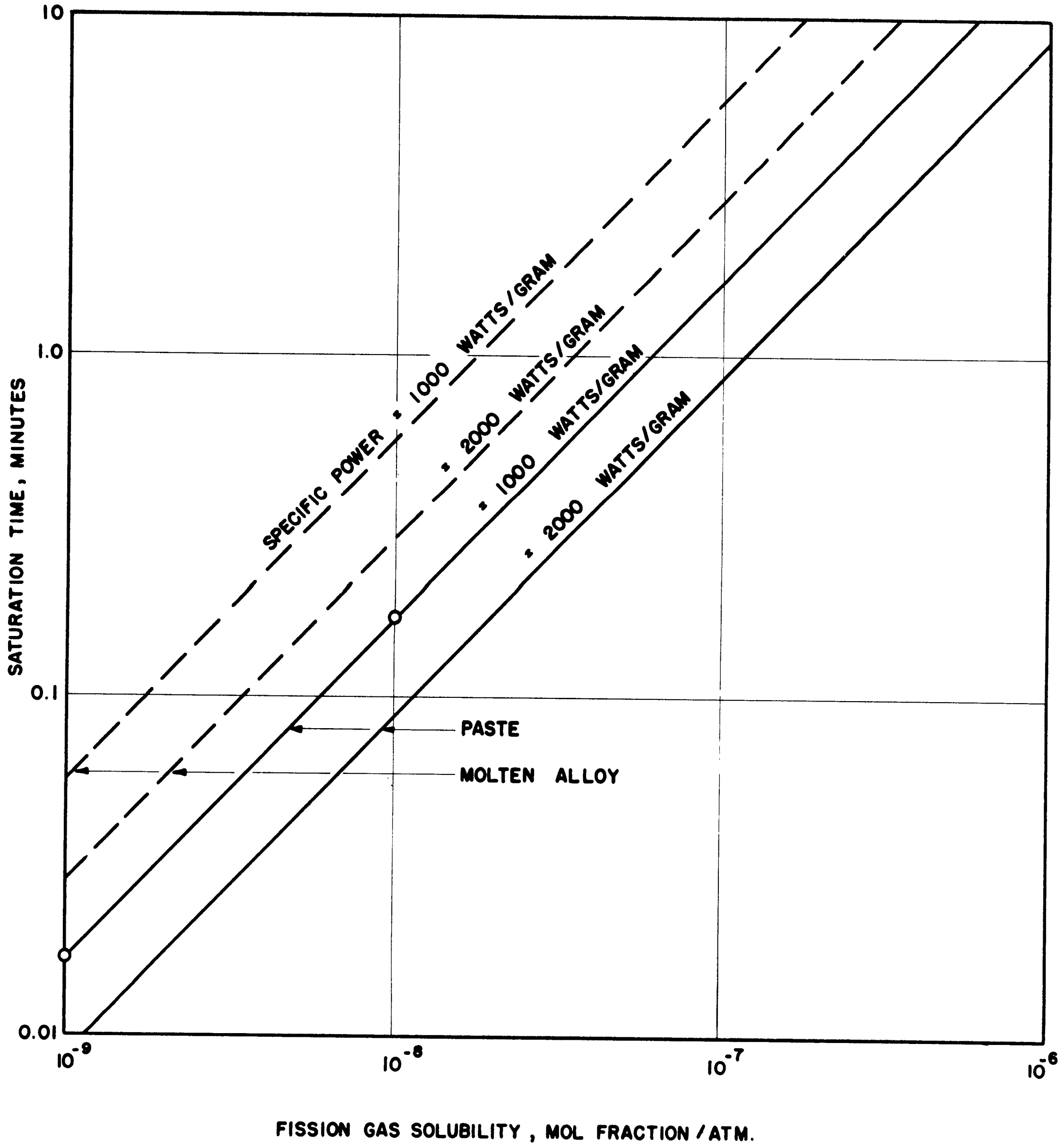

Figure 1. Estimated Time to Saturate Mobile Fuel Systems with Fission Gases at 500°C, 1.0 Atmosphere

fraction per atmosphere for xenon. At Oak Ridge National Laboratory, G. M. Watson determined that the solubility of helium in bismuth at 400°C and 600°C is less than 2 x 10^{-9} mol fraction per atmosphere.(7)

In summary, the noble gas solubilities in liquid bismuth, as determined experimentally, range from 10^{-5} to less than 10^{-9} mol fraction per atmosphere at 500°C. If these results were entirely in a range of solubilities either greater than 10^{-5} mol fraction or less than 10^{-9} mol fraction, there would be a clear cut indication that out-of-core degassing was or was not possible. Unfortunately, the experimental results bracket the range of solubilities that are very critical in this respect. Considering the possibility that the solubilities are so low that out-of-core degassing may not be feasible, the formation of fission gas bubbles and their possible effects within the core have been evaluated in preliminary fashion.

POSSIBLE GAS EFFECTS FOR EACH FUEL SYSTEM

Molten Alloy Fuel System

The potentially serious problems that can be anticipated due to the formation of gas bubbles in the core of a molten-fuel fast power reactor operating at high power density can be grouped in two categories; namely, those involving heat transfer considerations and those involving nuclear considerations. Of these, the former may manifest itself either as an increase in the maximum fuel temperature due to gas blanketing (either partial or complete) of the heat transfer surfaces or as a non-uniform distribution (in both space and time) of the wall thermal flux due to partial blanketing of the wall by gas bubbles.

Those problems involving nuclear considerations may be considered to manifest themselves in two ways also, one, as a steady-state decrease in reactivity caused by a uniform distribution (in time and space) of gas bubbles in the fuel, and two, as non-steady state changes in reactivity due to sudden displacement of the fuel caused by the movement of large gas bubbles.

The characteristics of the molten alloy fuel under consideration herein are such that only a very extreme rise in the fuel temperature is of any importance; in fact, as far as the fuel itself is concerned the only apparent limitation on the maximum fuel temperature is its boiling point which in this case is beyond all practical power plant considerations. Therefore, the real limitation in the maximum fuel temperature is not the fuel itself, but rather the allowable temperature of the heat transfer surface with which the fuel is in direct contact.

This limit is established by the corrosion resistance of the container material to the fuel, the high temperature creep behavior of the container material, and the maximum permissible thermal flux as determined by thermal stress considerations. If portions of the heat transfer surface were to become covered with gas bubbles or film, the heat transfer loading of the uncovered portions would increase. If the design were not very conservative, failure might be expected. Also, if a bubble were suddenly to uncover a portion of wall and allow overheated fluid, previously shielded by the bubble from good heat transfer with the wall, to contact the wall, conditions of local overheating and thermal shock as well as the inherent thermal cycling might cause tube failure. Bubbles that nucleate on heat transfer surfaces are held to the surfaces by surface tension; and they will grow in size until the sum of buoyant forces and the viscous drag of the fluid exceed the surface tension force. This condition may not be attained until the bubble grows to a diameter sufficient to bridge the narrow fuel passages. Preliminary experiments which the authors have conducted in a static water-glass-carbon dioxide system show that such bubbles will not move upward but remain in place and grow continuously until the critical size is reached. The natural convective flow velocity, present in a high power density fuel might be sufficiently high to carry the bubbles to a liquid interface; if not, forced fluid circulation might be required.

The allowable equilibrium gas volume in a fast reactor is limited by the reactivity change due to fuel dilution that can be safely and practically accommodated. Calculations show that in the type of fuel systems being considered herein, a volume of gas equal to one per cent of the fuel volume is equivalent to about one dollar of reactivity change. If all the bubbles are in the order of 1.0 mil diameter or smaller then even if none cling to the wall the terminal velocities of the bubbles will not be high enough to maintain the gas volume at 1.0 volume per cent or less. On the other hand, the growth of gas bubbles to a diameter of about 50 mils or more could result in bridging of the fuel ligaments with the subsequent slugging action causing serious non-steady state fluctuations in reactivity.

Paste Fuel System

The fission gas emission problem for a paste system is similar in the broad concepts to that previously discussed for a molten fuel system. There are, however, some differences which must be mentioned.

The limit on the maximum temperature attained by the fuel alloys of the molten-fuel system is the vapor pressure of the molten metal. This is not high enough in most cases to be a practical limitation. The situation is different in the paste system in that significant temperature rises in the paste are prohibited by the possibility of sintering of the paste particles. Also boiling of the liquid phase is a factor with sodium which is the liquid metal under consideration. Thus, even ignoring thermal stresses in the tube walls, significant gas-coating of the walls cannot be permitted. Also the formation of gas voids containing several fuel particles would lead to a prohibitive temperature increase of those particles within the void so that sintering would occur.

As with the liquid system, the possibility of separation of the paste column, within the small ligaments contemplated, exists. Similarly, there is the danger of plugging of the small ligaments or orifices, which are necessary for flow control, by gas bubbles or voids which will be held in place by interfacial tension. Since a continuous through-flow of the bed is required for reprocessing and possibly for prevention of stitching by fission fragments, such an occurrence would prevent proper operation.

As mentioned for the molten-fuel system, the effects of a possibly substantial and changeable void fraction on reactor kinetics must be considered.

PRELIMINARY EXPERIMENTATION AND ANALYSIS

Molten Fuel System

In making an analysis of the fission gas problem in the molten fuel system it is necessary to determine, first, if bubble formation is likely and if so where nucleation is likely to occur; and second, what the effect of bubbles will be and what the possible remedies are. Whether or not bubble formation is likely to occur depends on the rate of production of gas, the gas solubility in the molten alloy, the degree of supersaturation that can be accommodated by the fuel system, and the rate at which gas atoms diffuse or are transported from the system.

As previously discussed, the rate of production of fission gas in the molten fuel system could be equal to about one volume per day per volume of fuel alloy. The solubilities of the fission gases in the molten alloy are not known; but it is reasonable to expect that they could not be significantly different from the solubilities of xenon or helium in bismuth. As previously mentioned, data that are available on these latter systems indicate a solubility of 10^{-6} mol fraction at most, and possibly less than 10^{-9} mol fraction. Irrespective of which of these values is more nearly correct, they are both so low that, in the fuel system being considered, saturation of the molten alloy by fission gases would be obtained in a matter of minutes at most and possibly in fractions of a second.

The prospects for the molten alloy fuel system being able to accommodate a significant degree of supersaturation (i.e., in effect, an increase in the gas solubility) have been examined. In the literature there are reported numerous experiments in which degrees of supersaturation equivalent to hundreds of atmospheres pressure have been achieved in aqueous systems.(8-10) These experiments clearly demonstrated that in order to obtain a significant degree of supersaturation the following conditions are prime requisites: (1) the liquid must be free of unwetted particulate matter and/or gas bubble nuclei, (2) the liquid must be quiescent or in non-turbulent motion, (3) the liquid must wet the container and (4) the container must be completely free of surface scratches or pit marks. In the molten alloy fuel system under consideration the fuel will have to be maintained free of particulate matter for reasons unrelated to the fission gas problem; and, in any case, the particulate matter that might be generated within the fuel would, in all probability, be wetted by the molten alloy. Elimination of particulate matter from the fuel would also preclude the inclusion of gas bubble nuclei on the surface of such particles. The likelihood of free gas nuclei being present in the fuel is quite remote inasmuch as the fuel will have been subject to prolonged outgassing during the course of its formulation.

Studies of the mechanisms of cavitation have suggested(11,12) that the negative pressures generated within vortices may be sufficient to overcome the tensile strength of the liquid and thus create nuclei for bubble formation. Experiments using liquids supersaturated with gas have demonstrated the effectiveness of turbulence in the liquid for causing bubble formation. Thus, it appears that if the molten fuel alloy were in turbulent motion supersaturation with fission gases would not occur. However, an analysis of the thermal convective flow in the fuel ligaments and under the conditions previously described indicates that the flow is almost certain to be laminar; therefore, the presence of vortices in the fuel ligaments that might serve as nucleation points for bubble formation seems unlikely.

On the basis of the foregoing discussion, it seems almost certain that there will be some supersaturation of the fuel alloy, at least away from the container walls. However, the degree to which it occurs will depend on the rates of diffusion or transport of the gas atoms to a free surface, the availability of nucleation sites within the liquid and on the heat transfer surfaces, and finally on the nucleating properties of the intense ionizing radiation which will be present in the reactor. Judging from the experiments previously noted, it is highly improbable that the liquid itself would sustain supersaturation in excess of 100 atmospheres. The nucleating properties of ionizing radiation in systems such as these are unknown; but there is experimental evidence that the effect would be to reduce the degree of supersaturation.(13) Because of the length of the fuel ligaments (~36 inches) and the very low diffusivities of the fission gases in the molten alloy (estimated to be in the order of 10^{-5} cm^2/sec) the static diffusion of gas atoms out of the fuel will be negligible. Mass transport of gas atoms by thermal convection currents while appearing to be appreciable in some instances (~50 ft/hr.) is also expected to result in negligible removal of gas at the liquid surface due to the short time that the liquid is exposed to the surface, the dependence upon static diffusion at this point, and the extremely small surface area in proportion to the volume that is available at the liquid-gas interface.

In the manufacturing processes the core structural material is marred with a multitude of microscopic crevices. It is impossible to completely fill all these crevices with liquid, particularly those with an acute angle at their base, even though the liquid wets the container. The resulting void spaces fill with gas that diffuses from the liquid, thus, the nuclei are formed from which bubbles may grow. Experiments have been made in a water carbon dioxide system with internal heat generation to create natural convective circulation to determine the site of bubble nucleation in a supersaturation liquid. As was expected, nucleation occurred, entirely as far as could be determined on the solid surfaces and not within the bulk of the liquid.

In the light of the foregoing it seems almost certain that the heat transfer surfaces in the core of a molten fuel reactor will provide the nucleation sites for bubble growth. Also, the number of such sites that are available will be so great that the liquid will not be supersaturated to the extent required for nucleation within the bulk of the liquid.

Bubble Growth After Nucleation

As previously described it seems most likely that bubble nucleation will occur on the tube walls in suitable pores and crevices. Also it seems certain that some degree of supersaturation will exist in the fluid. In this case there will be continuous diffusion of gas from the fluid into the bubbles. If the fluid were stationary, preliminary calculations show that the bubble growth would be very slow. However, substantial velocities will exist in the fluid (order of perhaps 50 feet per hour) because of natural convection due to the internal heat source. Hence, it appears likely that static diffusion will be a significant mechanism only immediately adjacent to the bubble, whereas the macroscopic fluid motion will be of far greater significance at any appreciable distance. The situation appears somewhat similar to that controlling the diffusivities of momentum and heat in flowing fluids wherein the static diffusivities are of importance only in the boundary layer. No attempt has yet been made to estimate the rate of bubble growth under the conditions actually prevailing. It would appear to be much more rapid than a prediction based solely on static properties, would indicate.

Growth of a bubble attached to the wall will apparently continue until a critical size is attained such that the combination of buoyant and fluid-dynamic forces will become sufficient to overcome the interfacial tension forces which prevent dislodgement of the bubble. If estimates are based on the balance between buoyancy and tension only, it is found that very large bubble diameters will be required, much greater than the diameter of the anticipated core ligaments. Figure 2 shows the required bubble diameters to overcome the interfacial tension force as a function of the wetting angle for sodium, mercury, and water. Sodium and mercury were selected as fluids fairly typical of light and heavy liquid metals and of which the physical properties were well known, rather than as fluids particularly applicable to the reactor concept. Water is included only for comparison, and because simplified, confirmatory experiments in water were possible. The calculations are for bubbles formed on a horizontal surface. The substitution of a vertical surface will not make a substantial change in the critical dimension, although it will somewhat alter the bubble shape. It is noted that almost independent of wetting angle or fluid, bubble diameters of the order of at least 1/4-inch are required. A maximum of about 1/2-inch is necessary for sodium. Since ligaments with diameters of the order of 1/16-inch diameter are necessary to provide a sufficient density of heat transfer area for the fast power reactor concept, such bubbles would completely separate the liquid column (as sometimes occurs in a mercury thermometer, for example). If the effects of the natural convection fluid velocity or a superimposed forced

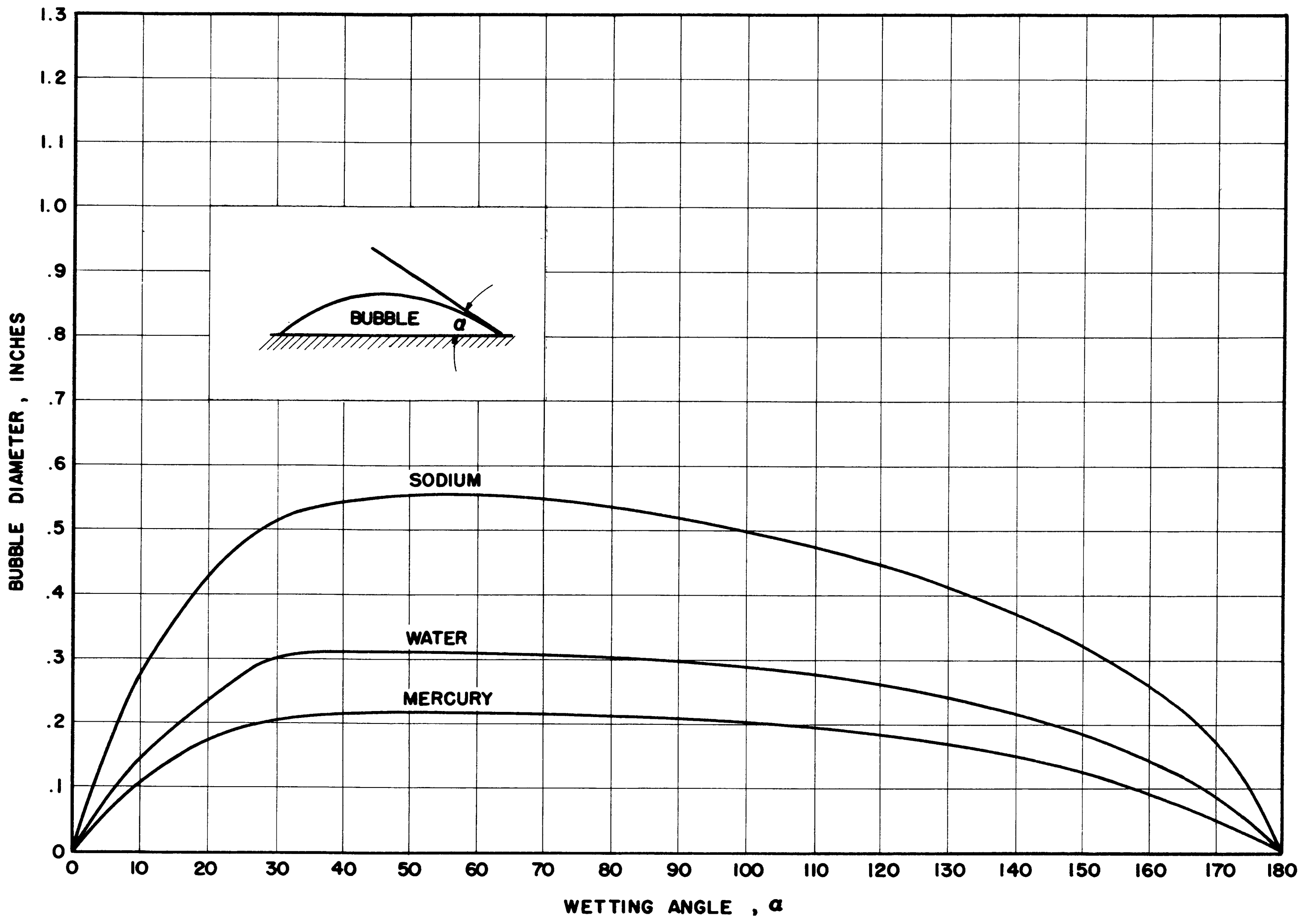

Figure 2. Required Bubble Diameter to Overcome Interfacial Surface Tension Force

convection velocity are included, it may be that sufficient fluid-dynamic drag would exist to sweep bubbles from the wall before they attained such a size. No estimate of this effect has been made. It must await a more detailed specification of the velocity profiles to be expected.

Bubble Motion in Ligament and Effects

Some predictions are possible regarding the motion of a finite bubble released from the wall into the stream, or of bubbles generated in the bulk of the liquid. In the absence of fluid motion a bubble will, of course, rise to the surface at a rate which is a direct function of its size. Bubbles of a microscopic diameter will have a low terminal velocity and the equilibrium gas content of the ligament based upon such bubbles may be large. An estimate of the equilibrium gas content in the free bubbles as a function of bubble diameter has been made based on the Stokes' velocity for unhindered settling. The results are given in Figure 3. It is seen that for bubbles in the order of 10 microns diameter the fuel dilution would be in the order of 1.0 to 10 per cent excluding the volume of bubbles held on the heat transfer surfaces, an excessive dilution from a nuclear standpoint. If significant fluid velocities exist due to natural convection, as seems likely, then the equilibrium gas content of the core as a function of mean bubble size may be drastically altered.

On the other hand, the sudden displacement of larger bubbles may involve important fluctuations in the steady-state reactivity. In the fuel systems being considered, 1.0 cc of fuel is approximately equivalent to one dollar of reactivity change. The control systems of fast reactors can be designed to accommodate increases in reactivity equal

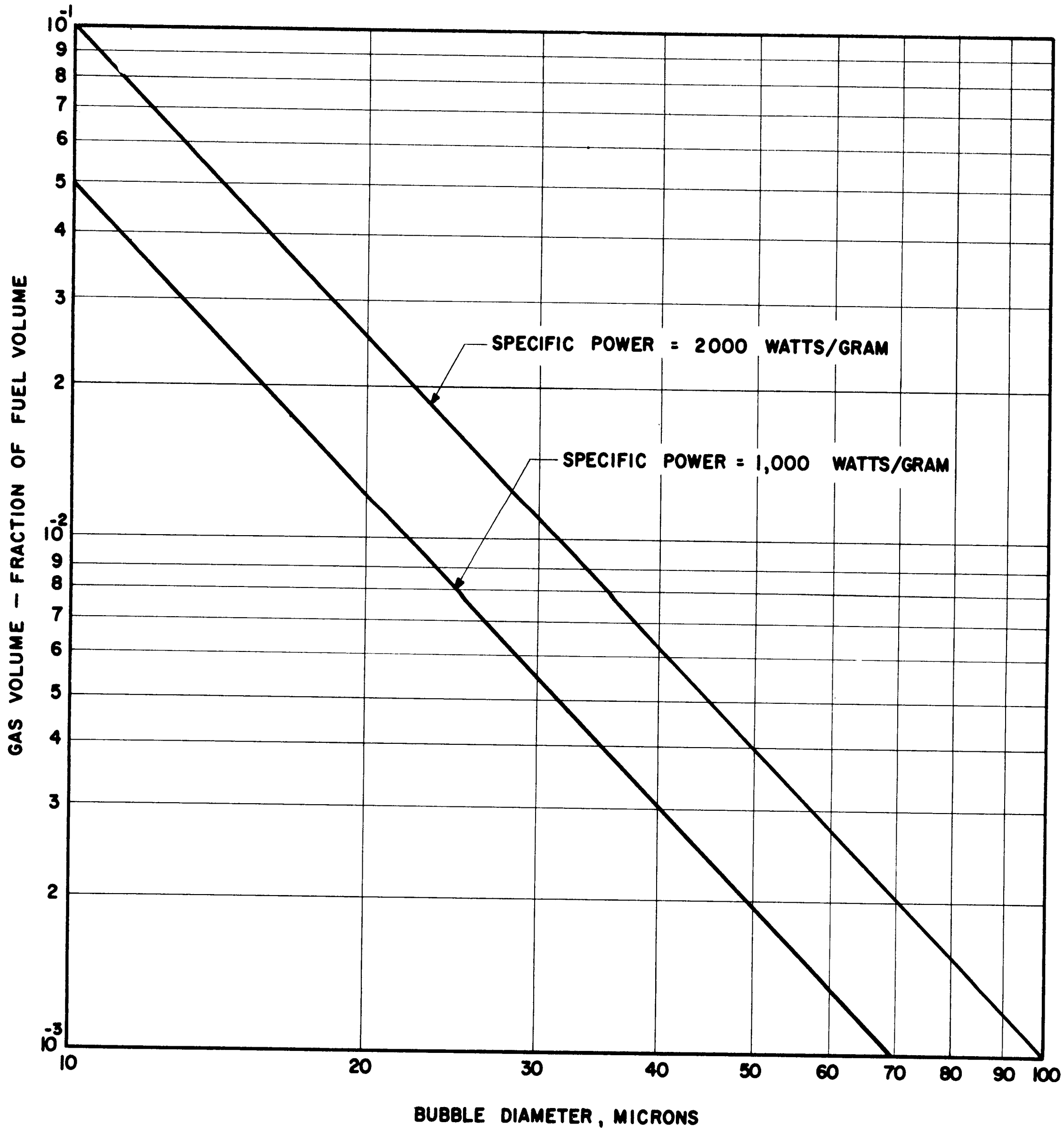

Figure 3. Equilibrium Content of Unhindered Gas Bubbles in a Molten Alloy Fuel System

to one per cent/second which in this case could result from a net fuel displacement of 0.01 cc per second.

Preliminary experiments in a water-air system in vertical glass tubes have been conducted to confirm the maximum bubble diameter estimates previously mentioned. As predicted, it was found that gas emitted through the lower end of a 1/4-inch glass tube would indeed form a complete block, and would not rise to the surface. However, such a result could not be attained in a 1/2-inch tube. If bubbles considerably smaller than the tube diameter were injected at the lower end of the 1/4-inch tube, it was found that they would rise. Their motion, however, could be controlled by imposition of a downward velocity. Depending upon the magnitude of this velocity the relative force balance between buoyancy and drag would be affected, and hence the rate of bubble rise. Of course, there is substantial increase in bubble diameter as the bubble rises toward the surface due primarily to the diminution of static pressure. In the reactor ligament there is a second mechanism to be considered in that the fluid is supersaturated, so that there will be a continual influx of gas into the bubble. No estimate of the magnitude of this effect has as yet been made.

The nuclear effects of significant and changing voids within the reactor have been previously mentioned. Also to be considered are the heat transfer and stress effects. If a substantial portion of the tube wall were to be blanketed by gas in the form of small bubbles, or as a continuous film, or due to large-scale liquid column separations as previously described, the wall heat flux imposed upon the unblanketed portions would increase in inverse proportion to the unblanketed area, assuming the power output were maintained. One of the limiting design factors is tube-wall thermal stresses. These are directly proportional to wall heat flux. If an economically feasible design were postulated for a gas-free core, failure could be caused by such a gas accumulation.

Figure 4 shows the fluid temperature rise to be expected from such an occurrence in a sodium system (as the paste system), and it is noted that this rise is substantial. A similar result could be expected for the liquid-only system. However, for the low vapor pressure fluids under consideration, such a temperature rise in the fluid does not seem important, provided the high temperature portions of the fluid do not contact the wall.

Similar to the overall effect mentioned above there is a local increase of wall heat flux to be expected in the immediate vicinity of a finite bubble since the fluid directly behind the bubble will become overheated. No quantitative estimates of this effect have been made as yet.

An additional perturbation of wall heat flux as calculated from static diffusivity conditions is to be expected at the extremes of the ligament (increase at top, decrease at bottom) due to natural convection effects. This has been investigated in a generalized way by the authors.(14,15) However, no concrete, quantitative estimates are yet possible for the liquid-metal ligament systems here described.

Possible Remedies

The most obvious remedy for the fission gas emission problem in the molten fuel system would be continuous or periodic external degassing, so that supersaturation within the fluid would not be attained.

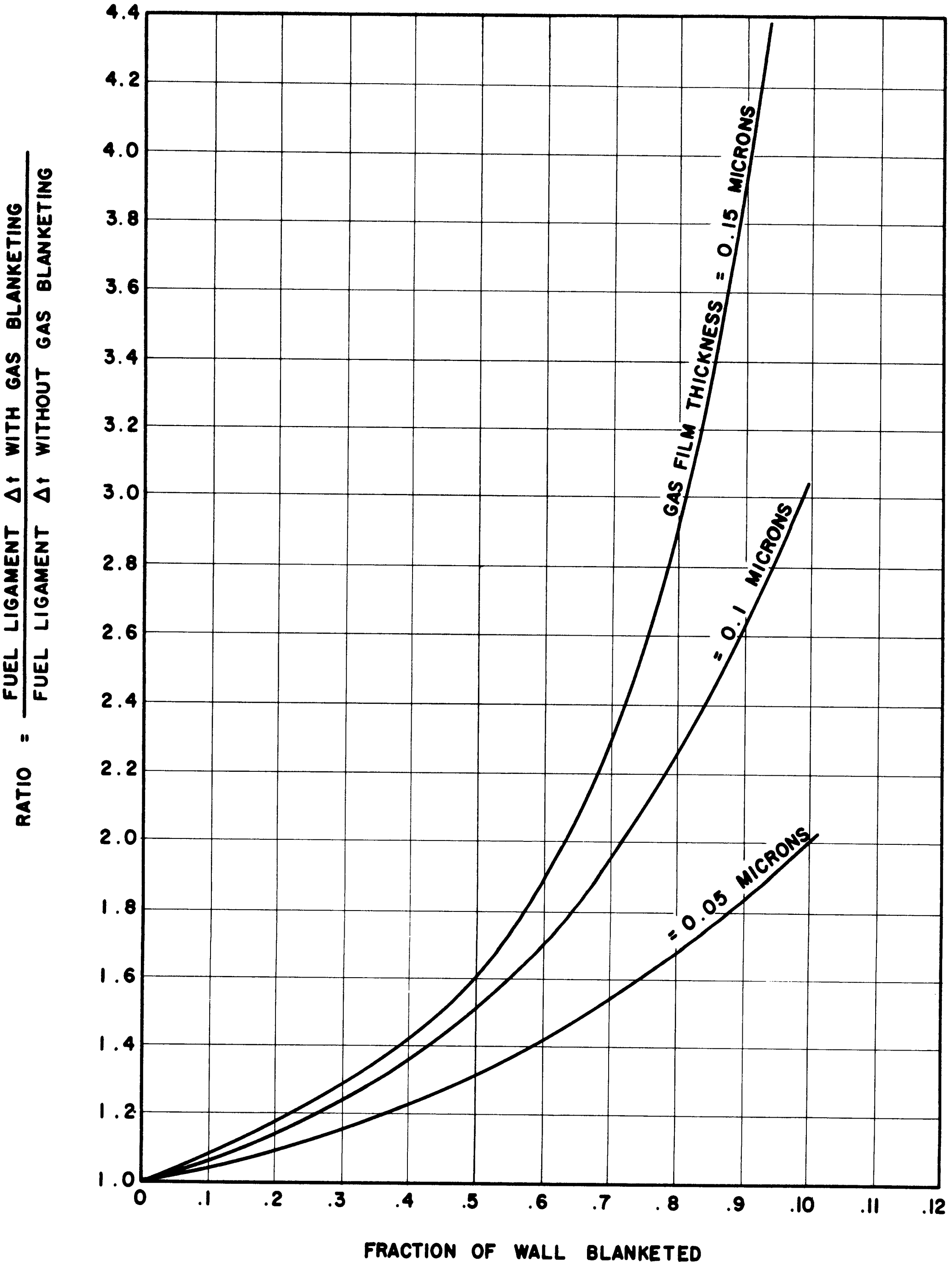

Figure 4. Effect of Gas Blanketing of Heat Transfer Surface in a .050 Inch Diameter Paste Fuel Ligament

As previously discussed, the fact that the somewhat sketchy information presently available indicates very low solubilities for gases in the applicable liquid metals appears to be the overriding difficulty. It appears that supersaturation will be attained very quickly so that a continuous, rapid circulation of the entire core would be required. This would involve a much more complicated system with respect to pumps, seals, valves, piping, etc., than desired; and would also increase the out-of-core inventory of fissile material.

Another possibility is the prevention of significant supersaturation (so that bubble growth by infusion from the surrounding fluid is not possible) by artificially induced nucleation. Thus, there would be a continuous supply of a very great number of microscopic bubbles in an approximately saturated fluid. These would rise to the surface or be carried there by the natural convection circulation. Possible methods of inducing nucleation include magnetostriction devices, mechanical vibration of the core (probably prohibitive for reasons of structural integrity), ultrasonic techniques, and perhaps the application of suitable oscillating electromagnetic or electrostatic fields. It is possible that the high radiation flux within the reactor will cause significant nucleation before any appreciable degree of supersaturation is attained. A preliminary experiment was conducted in water, supersaturated with carbon-dioxide, to determine whether nucleation on a large scale could be induced by gamma radiation. A cobalt-60 source provided approximately 200 mr/hour within the test section. There was no effect visually noticeable. However, it is known that under suitable conditions radiation does cause bubble nucleation. Such an effect is used in the "bubble-chamber" for study of radiation.(13) The negative result from the preliminary experiment described above in no way indicates that significant nucleation will not be triggered by the very intense radiation within the core. A positive result would have indicated the certainty.

If the remedies described above prove ineffective, the controlled injection of microscopic bubbles has been suggested.(16) These would continuously sweep the liquid and prevent the attainment of significant supersaturation. They would provide a statistical equilibrium with regard to void fraction which would be considerably more favorable from the nuclear viewpoint than the rapid fluctuations which might occur with the sudden release of bubbles of significant size. Also they would prevent the appearance of bubbles sufficiently large to block the ligaments. If good wetting between fluid and container could be obtained (perhaps by suitable trace additives), no blanketing of the walls should occur. However, even with such injection, it might be desirable that some vibratory method as described above be included to assure the prompt removal of small bubbles from the wall, if they did in some manner collect. The design of a sufficiently simplified mechanism for the bubble injection must yet be considered. It is safe to say that considerable additional research is required before a decision regarding the necessity or suitability of any of the provisions described above can be made.

Paste System

The general nature of the paste fuel system under consideration has been discussed previously. Basically, it consists of a multiplicity of vertical passages with a hydraulic diameter of the order of 50 mils filled with a fuel-particle (order of 5 mil diameter), liquid-metal paste.

Since the density difference between particles and liquid metal (sodium in a typical case) is substantial and the velocities low, it is to be expected that the particles will be supported by direct contact with each other rather than hydrodynamically as in a slurry. Thus, the packing will be governed by the geometry of the particles (order of 40 per cent void fraction).

It is necessary that a continuous through-flow be maintained in order that the bed may be reprocessed as required to maintain homogeneity of particle size and remove fission products (particularly, fission gases) and probably to prevent stitching of the particles by fission spikes. While a finite through-flow is necessary, only a low flow rate is desirable, since, to attain reasonable particle life it probably is necessary to minimize the number of temperature cycles to which a particle may be submitted. Also increased bed motion entails an increased amount of fuel handling.

Based upon considerations regarding stitching and reprocessing requirements, and the practicalities of paste flow control, it appears that a particle downward velocity consistent with an in-core residence of about one hour is desirable. Since increased liquid phase velocity through the bed increases the forces on the particles impelling them in the direction of flow, the particle and liquid flow velocities are related. The liquid velocity profiles may be considerably affected by the radial show that the velocities due to this effect are substantial compared with the small through-flow velocities.

Assuming a one-hour core residence time for the particles, a specific power of 2,000 watts/gram and 8 per cent release of fission gases from the particles, preliminary calculations indicate the generation within the core of inert fission gases in the amount of approximately one per cent by volume of the liquid volume per pass. In other words, sufficient gas may be released per pass through a 36-inch ligament to form a void 3/8-inch long, fully blocking the ligament.

The problems of analyzing the effects of the fission gas release phenomenon in the context of the paste system are two-fold:

1. Under the particular conditions of the gas source as it exists in the system -- i.e., formed atom by atom in a volumetric distribution similar to that of the neutron flux -- it is necessary to establish the likelihood of the formation of voids of significant size; and

2. assuming that significant voids can be formed, it is necessary to estimate their probable effects on heat transfer, particle and liquid velocity, and nuclear reactivity and stability.

Possible plausible mechanisms for the formation of substantial voids can be imagined and these will be discussed later. Certain preliminary experimentation and analysis have been performed regarding the effects of the assumed voids. These will be described in the subsequent paragraphs.

Motion of Gas Voids in Bed

Paste-bed degassing through the removal of gas from a space above the bed depends upon the existence of an upward relative motion of the gas voids with respect to the bed. In the light of preliminary experiments in a glass-bead, water system, this does not appear likely.

If the force balance on a bubble is considered, it is apparent that interfacial tension tends to prevent relative motion between particles and bubble, while buoyancy tends to cause an upward motion of the bubble through the bed. Since the interfacial tension forces depend upon surface area, and buoyancy upon volume, it is obvious that the buoyancy will become more important for large bubbles. In addition, there are fluid-dynamic drag forces of the moving liquid on particles and bubbles, since a relative motion between liquid and particles will exist in a vertical ligament with restricting orifice at the bottom if a pressure gradient (hydrostatic gradient due to ligament height, for example) is imposed on the liquid.

In the absence of high liquid velocities through the bed, it seems reasonable that motion of gas bubbles relative to the particles is only possible if the bubbles are quite large (relative interfacial tension forces small). This general hypothesis has been confirmed by two sets of preliminary experiments.

In the first case microscopic bubbles were admitted from below into a static bed of glass beads and water (about 4 mil beads). It was noted that no gas penetrated through the bed (about 3 in. in height) until the microscopic bubbles had formed large voids of the order of 1/4-inch diameter. These escaped from the bed surface.

In the second case, gas bubbles which appeared to completely fill the tube were injected into a vertical glass "ligament" about 7/32-inch in diameter and filled with the same glass-bead and water mixture. Downward flow from the ligament was restricted by an 0.042-inch orifice at the bottom. With the orifice blocked there was no tendency for the bubbles to rise. With flow under gravity through the orifice allowed, it was noted (by measuring the effluent) that there was relative motion between water and beads (downward flow of water greater than beads) and also between beads and gas void (bead downward velocity greater than that of the voids). The basic mechanisms in a three-phase flow of this sort are certainly not clearly understood and could be the worthwhile subject of further basic research. However, from these preliminary experiments it seems unlikely that the fission gas can escape from the top of the ligaments. Instead it is likely to be carried downward with the bed.

Possibility of Ligament Blockage

As indicated above, it is likely that the gas voids (if they exist) will be carried downward through the ligaments to the restricting orifice at the bottom. In general such a restriction is required to maintain the low flow rates desired. If gas pockets exist of sufficient magnitude to bridge the orifice, it seems likely that a blockage will occur. A pressure great enough to dislodge such a bubble considering the interfacial tension between gas, liquid, and container material will be necessary to re-establish the flow. Preliminary experiments have shown that the required pressure cannot be assumed to be based upon the orifice diameter, but rather must be based upon the effective opening between particles. Since the force balance relation between surface tension and pressure is of the form:

$\Delta p \alpha \frac{4\sigma}{D}$ where Δp is pressure differential across the interface

σ is surface tension

D is bubble diameter

it is apparent that considerably higher pressures may be required than would be at first assumed. It appears at least possible that if a suitable configuration of gas voids and particles arrive at the orifice together, an effective block can be formed by the voids bridging the interstices between particles and between particles and container walls. In this case the effective bubble diameter will be on the order of the interstice dimensions. Such results were observed in preliminary experiments of the type previously described wherein a 7/32-inch vertical glass ligament was used with glass beads in water with a 0.042-inch restricting orifice at the bottom. Figure 5 shows the effective diameter (as a fraction of the particle diameter) upon which the surface tension calculation must be based to explain the pressure drop necessary to re-establish the blocked flow. Experimental points were taken for 70 and 100 micron beads. It is noted that the diameter ratio is about 0.3 in either case. This is approximately the ratio of interstice to bead diameter if square packing is assumed, and more than twice the ratio for triangular packing (which seems physically more likely). Thus, it appears that the full pressure potential expected on a basis of uniform beads is not realized, possibly because of the lack of uniformity in bead size. If it is assumed that the ratio measured is typical, a similar orifice and bead size in a sodium-uranium particle system would require an applied pressure of 30 to 40 feet of sodium to remove a block of this type, due to the high surface tension of sodium. For such pressures, it must be assumed that the over-all force on the bed is balanced by shear between particles and container, whereas surface tension plugs the interstices between particles.

The mechanism described seems independent of orifice size. However, the experiments covered only one orifice diameter although the ratio between bead and orifice diameter was varied over a narrow range. It appears likely that the mechanism will break down if the orifice

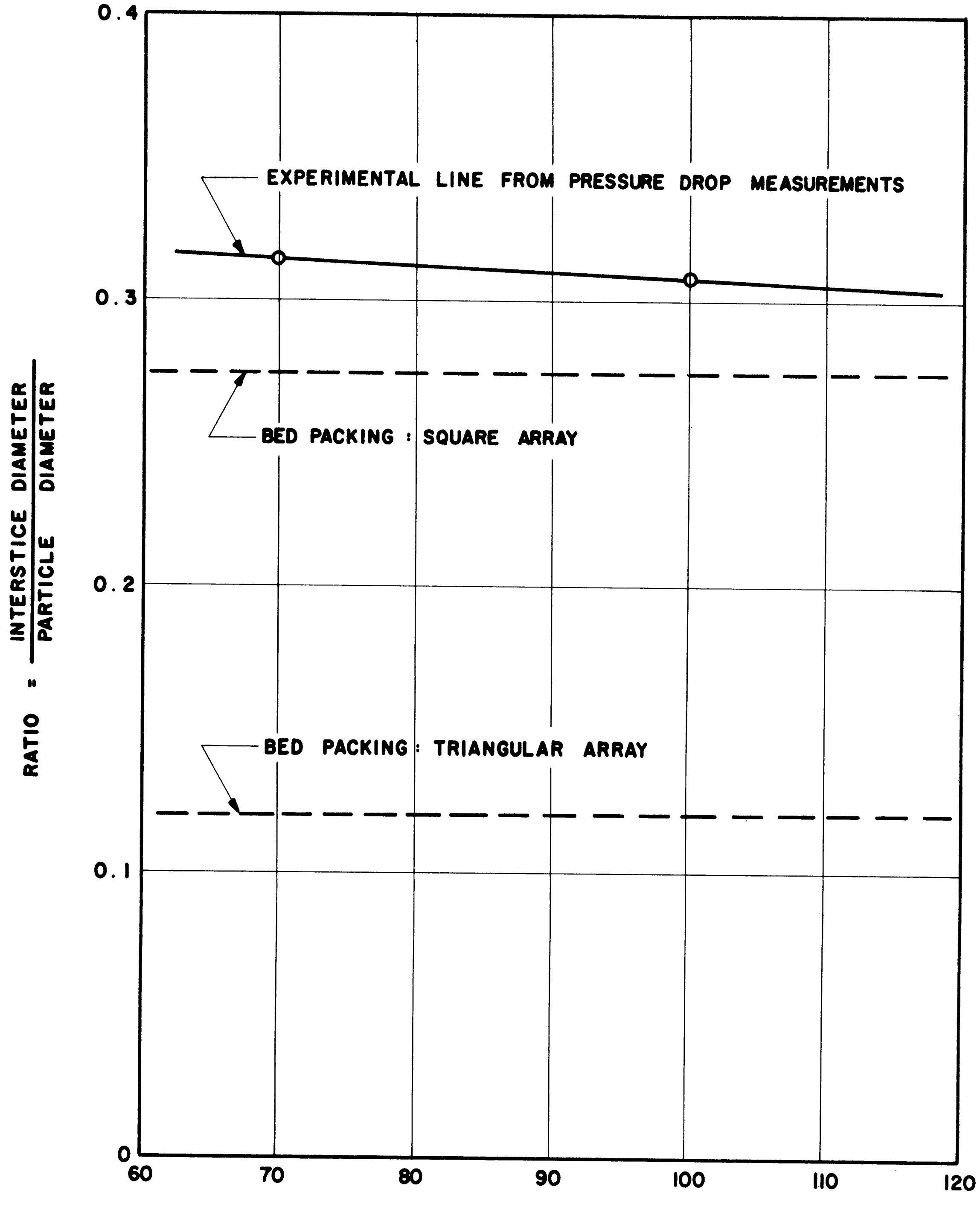

Figure 5. Interstice to Bead Diameter Ratio Vs. Bead Diameter from Surface Tension Calculations

diameter is a large factor greater than the bead diameter, since the formation of a bridge of this type is a statistical phenomenon which would become increasingly unlikely as the required number of participating beads increased. Further simplified experimentation to establish such a limit should be fruitful.

As verified by the preliminary experiments, the minimum usable orifice-to-bead diameter ratio is limited by the occurence of bridging by particles alone without the assistance of a gas phase. This apparently occurs when the ratio is about 7:1. In such a case, no application of pressure to the liquid phase will cause a removal of the block.

Relative Motion Between Particles and Liquid

An understanding of the complex phenomena involved in the prediction of the disposition and effects of fission gas within a paste system involves an understanding of the basic fluid-dynamics of the system with and without a gas phase. For example, a knowledge of the velocity profiles with and without natural convection is necessary to the estimation of the bubble growth rates and the effects of static diffusivity. As a first approximation it appears that a force balance considering viscous shear between particles and liquid and pressure gradient imposed on the liquid, and based on some characteristic diameter such as the hydraulic diameter of the interstice between particles, might be useful.

Order of magnitude calculations based on the applicable dimensions and fluid show that the flow is almost surely laminar. Thus, something of the type of Poisseule's equation for pipe flow might be approximately applicable with an empirical constant. A similar approach has been followed in the literature in a study involving static beds with throughflow.(17)

Preliminary experiments were conducted in the vertical glass ligament previously described to determine whether such a correlation might be used. Imposed pressure gradients were varied and the relative flow rates of water and particles observed (no gas in bed; however, it is not believed that a scattering of gas voids would materially affect the results unless tube blockage as previously described occurred). The velocity of the water relative to particles was inferred from the measured overall particle and water flow rates, the water velocity being based upon the interstice area. This area was taken as the average between square and triangular packing. Once the relative velocity between liquid and particles had been computed, it was referred to the full tube area by using the ratio between total interstice area and tube area. On this basis, a constant for the Poisseule's type equation was computed. The resulting equation and constant are as given below:

$$v = K \frac{\Delta p}{\Delta x} \frac{D^2 p}{\mu} \; ; \; K = 0.0030 \text{ (non-dimensional)}$$

Δp = pressure imposed

Δx = length of tube

Dp = particle diameter

μ = absolute viscosity

v = fluid velocity based on tube area

Any consistent set of units is suitable. It is noted that the relative flow rates so computed are a factor of about 5 greater than these given for a static bed.(17) It is felt that this matter might be usefully pursued with more detailed experimental and analytical methods.

According to the assumed mechanism the velocity of water relative to the bed is not a function of the tube or orifice diameter. This is obvious in the case of the orifice since it is found that the portion of the overall pressure drop which may be ascribed to the orifice is negligible. It seems intuitively obvious in the case of the tube diameter provided it is many times the particle diameter. This is substantiated by Reference 17 where a correction for the wall effect is included. It was noted that the effect is substantially negligible unless the ratio between tube and particle diameter is small.

A plot of the experimental data upon which the constant for Poisseule's equation was based is included as Figure 6. Although the constant was found to be not entirely independent of bead size, an average value was used. The precision of the experiments was not sufficient to make this discrepancy necessarily meaningful. Figure 7 shows the ratio of solid to liquid flow which was measured as a function of pressure gradient. If there were no relative motion between particles and liquid, the solid/liquid ratio would be expressed as

(1 - Void Fraction) / (Void Fraction).

In this case, the ratio would be 1.4 since the measured void fraction is 0.418. If there were no imposed pressure drop on the liquid (positive pressure imposed from below to counter the static head), and the particles fell under the action of gravity, it might be supposed that the fluid in the bed interstices would be entrained and move with the bed so that such a ratio would be attained. It is noted from the figure that the experimental points tend to approach this limit as the pressure drop is reduced. If a negative pressure were imposed upon the liquid sufficient to prevent its motion in spite of entrainment by the particle motion, the ratio would, of course, be infinity. As the imposed pressure is increased, the water velocity between the interstices, of course, increases

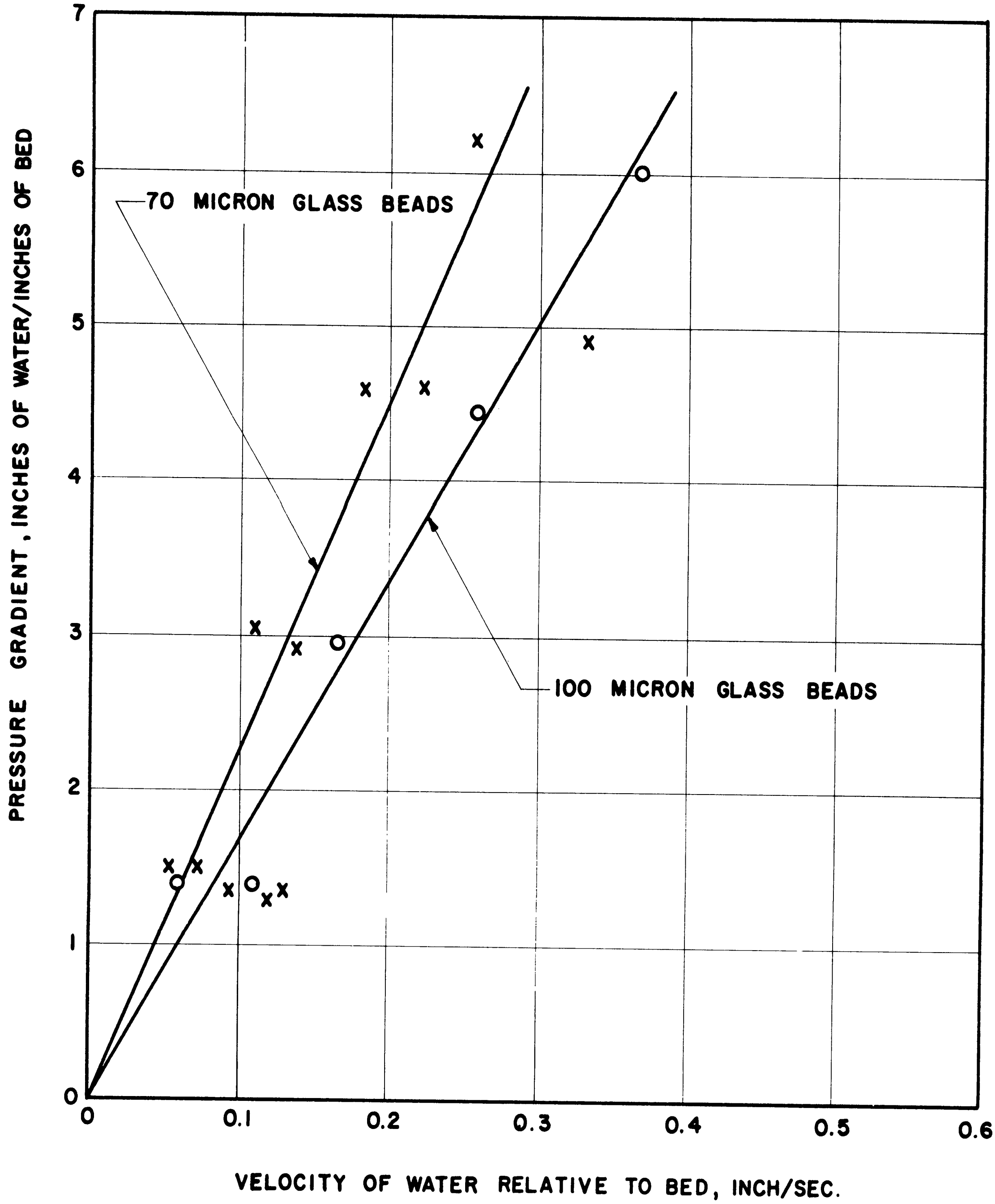

Figure 6. Pressure Gradient Vs. Relative Liquid Velocity for Paste Flow Through a 0.225 Inch Tube with a 0.042 Inch Orifice

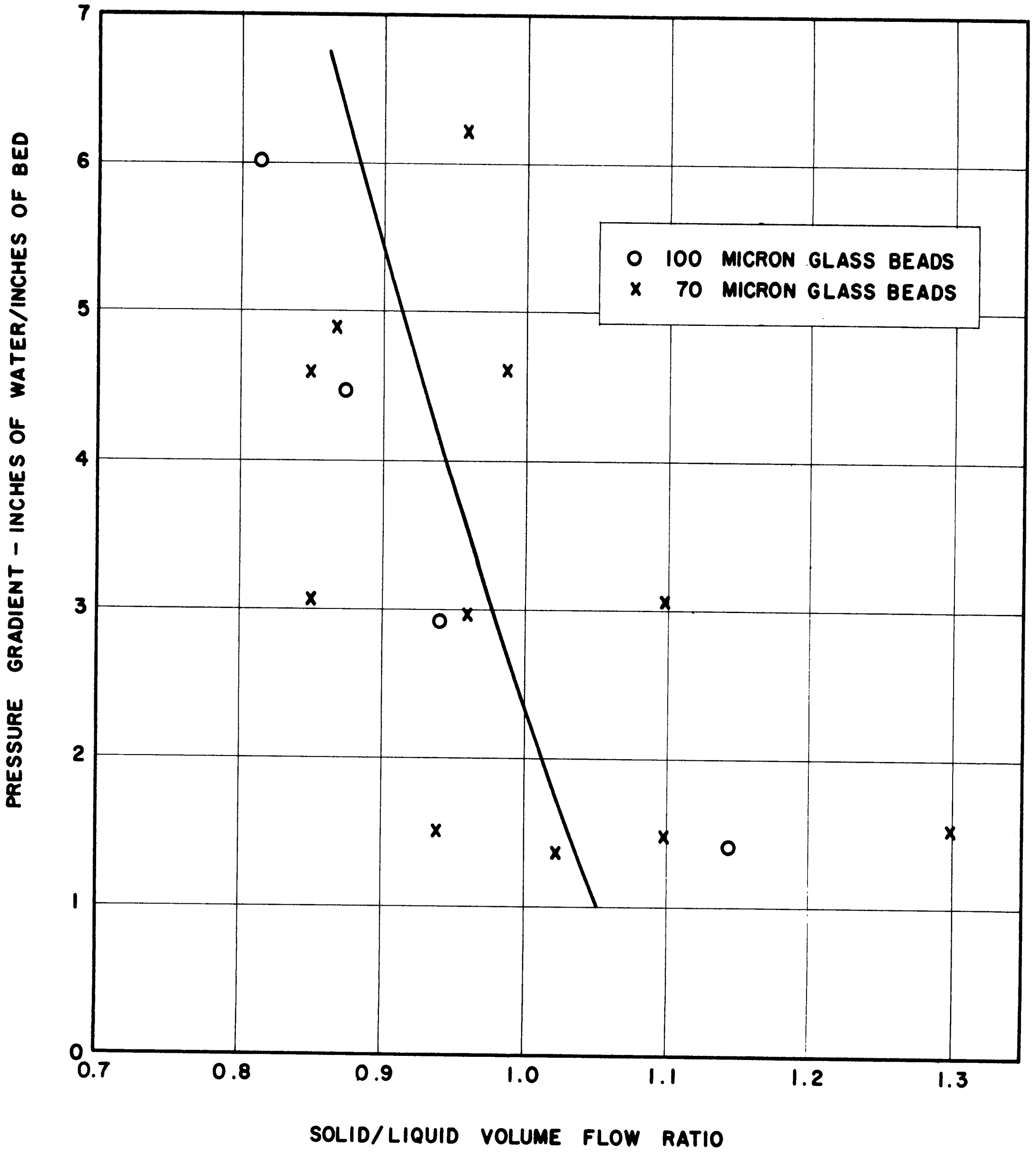

Figure 7. Pressure Gradient Vs. Solid/Liquid Volume Flow Ratio for Paste Flow Through a 0.225 Inch Tube with a 0.042 Inch Orifice

and causes a downward hydrodynamic drag on the particles. Thus, it is to be expected that the particle motion also will be increased, although not in proportion to the fluid velocity. Therefore, the solid/liquid ratio will decrease as the imposed pressure gradient is increased. This is noted from the experimental results.

The overall problem of predicting the motion of fluid and bed, with and without entrained gas is not to be solved in so simple a manner, although these preliminary experiments do shed some light on the mechanisms involved. It is probable that the significant parameters include at least particle size and tube size, particle shape and uniformity, fluid viscosity, relative densities of fluid and particles, coefficients of friction between particles and container, as well as the natural convection parameters in cases where a strong heat source is involved. The restraining forces on the particles must include a strong shear with the container walls, as confirmed by the following experiment. If an inverted U-tube is filled on one side with a static bed, it is obvious the shear is capable of supporting any height of column since the particles will not rise on the empty side. Presumably the force is the result of static friction with the wall in conjunction with a normal force due to the wedging action between the particles. It will not exist if the central portion of the column (i.e., along the centerline) is allowed to escape as in a verticle tube with open ends.

Heat Transfer Effects

Figure 4 shows the effects upon the fluid temperature differential between centerline and wall of a gas film of various thicknesses covering various portions of the wall of a sodium-filled ligament of an 0.050-in. diameter. It is noted that the increase is substantial (about 50%) if a film of 0.1 microns covers 50 per cent of the wall. Such an increase would probably be prohibitive from the viewpoint of sintering of particles. In addition, the unblanketed portion of the wall would suffer a 2:1 increase in heat flux raising the inside wall temperature and increasing thermal stresses by a factor of two. If the design were limiting in this respect, such an increase could cause tube failure. The likelihood of the formation of such a film is not known; however, it must involve the relative surface tension of the liquid and container.

Another effect regarding heat transfer which is of importance in the paste system, is the formation of gas pockets which include fuel particles. Preliminary calculations summarized in Table I show that a particle-filled gas pocket of only 0.020-inch in diameter would cause a temperature increase within the pocket of 560°F under typical conditions. These calculations are based on published data for the thermal conductivity of powders in gases.(18) From the viewpoint of sintering, this temperature rise is, of course, completely prohibitive with metal alloy fuel particles.

A consideration of the forces upon a fuel particle on the surface of a gas pocket will include interfacial tension and gravity force on the particle. If quantitative estimates are made, it is obvious that the interfacial tension is far too great to allow a penetration of the particle into the gas pocket. Preliminary experimental observations using the apparatus previously described confirm this expectation where the particles are free to move. However, it is necessary that reliable

TABLE I

TEMPERATURE DIFFERENTIAL FROM CENTER TO WALL OF FISSION GAS BUBBLES FILLED WITH 125 MICRON FUEL PARTICLES

Bubble Diameter	Temperature Differential	Number of Particles per Bubble
0.002 inch	6 °F	1
0.010 inch	140 °F	9
0.020 inch	560 °F	75
0.040 inch	2200 °F	600
0.100 inch	14000 °F	9400

wetting of the particles by the fluid be obtained and also that the bed be free to expand. The likelihood of this condition being fulfilled in the gas-generating paste is not known, and would bear further experimental and analytical examination.

Likelihood of Void Formation

All the previous discussion has been based on the assumed existence of macroscopic gas pockets within the paste. It was previously mentioned that in a typical case sufficient gas is generated per pass to form a gas void 3/8-inch in length and of a diameter equal to the ligament diameter. However, the gas is generated "in situ", distributed throughout the fluid volume, atom by atom, as the fissioning occurs. Order of magnitude calculations considering the solubility of the gas in the liquid metal indicate that this solubility will be exceeded very quickly. The particles provide a multitude of suitable bubble nucleation sites so that it is not likely that large degrees of supersaturation will be attained. Thus, it appears certain that microscopic bubbles will be formed throughout the volume, probably within suitable crevices of the particles or walls. It is at least possible that these will remain as a very large number of microscopic bubbles, clinging to the particles, and will pass from the core in this fashion. Suitable degassing of the reprocess stream would then suffice to prevent difficulty.

However, a detailed examination of the situation reveals various possible mechanisms whereby, pockets of magnitude sufficient to cause plugging and deleterious heat transfer effects may be formed. One which seems obvious is the following. Suppose a microscopic gas bubble clinging to a suitable crevice in the wall material. In view of the rates of gas production and the mechanisms for bubble formation, it must be assumed that some degree of supersaturation exists within the liquid. Then there will be continuous gas diffusion from the passing liquid stream into the stationary bubble. The bubble will grow until it attains a size such that the combination of buoyancy and fluid-dynamic forces are capable of overcoming the interfacial tension forces holding it in place. Estimates previously described (Figure 2) indicate that the diameter will be considerable in this case, and may well cause a separation of the liquid column.

An estimate of the time necessary for the attainment of such a diameter depends upon knowledge of the degree of supersaturation existing within the fluid, the velocity profiles as influenced both by the imposed pressure gradient and the natural convective forces, and the diffusion coefficient for the gas in the liquid metal. An estimate sufficiently precise to be meaningful has not yet been made.

Conclusions

Problems involving the effects upon reactor operation of the fission gas generated within the core of a mobile-fuel fast power reactor may exist both for the molten-fuel and for the paste system. In both cases they could be formidable and very complex. The present paper is an attempt to point out the nature of these problems and the results of some very preliminary analyses and experiments which attempt to illustrate the basic mechanisms involved and some of the overall trends. It seems likely that methods can be found to obviate the gas effects to a sufficient extent that suitable reactor operation can be attained. However, it is apparent that considerable additional basic research is required before the necessary understanding of the phenomena can be achieved.

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