# A PROPOSED MODEL OF BUBBLE GROWTH DURING FAST TRANSIENTS IN THE KEWB REACTOR

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ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc

## I. INTRODUCTION

The KEWB Reactor is a prototype 50 kilowatt aqueous homogeneous research reactor equipped with the necessary auxiliary apparatus and instrumentation for the study of transient behavior - see reference (1), (3). The core is a 12¼ inch I.D. stainless steel spherical shell containing 11.5 liters of fuel solution. The fuel solution is 0.75M $UO_2SO_4 \cdot 3H_2O$ dissolved in 0.5M sulphuric acid acidified solution. The core is contained within a cubic graphite reflector 56 inches on a side. During the past three years approximately 900 transients have been performed in the effort to determine the details of the kinetic behavior of this type of reactor. The experimental data with analytical interpretation have been published from time to time and will be found in references (2), (3), (4), (5), and (6).

Analyses of the experimental data have shown that shutdown mechanisms other

*This paper was presented at the Summer Meeting of American Nuclear Society, Gatlinburg, Tenn., as Paper 25-3, 6/15/59.

**The studies reported here are part of the Kinetic Experiment Water Boiler (KEWB) program conducted by Atomics International for the Reactor Safety Branch of the United States Atomic Energy Commission at Santa Susana, Calif. AT-(11-1)-GEN-8.

than temperature become dominant in transients with periods less than 15 milliseconds. For example, thermal expansion of fuel solution accounts for only 20% of the total reactivity compensated at the time of peak power in a power excursion with a two millisecond reactor period.[2] All available evidence supports the contention that the non-thermal component of the compensated reactivity is the result of void growth, and that this void is made up of many very small radiolytic gas bubbles with internal pressures in the range of 10 to 1000 atmospheres. Therefore, in order to understand the dynamic behavior at KEWB, we must be able to account quantitatively for the very rapid growth of small bubbles.

A number of theoretical models have been constructed to account for the observed dynamics of bubble growth in KEWB.. for example, (a) diffusion growth in a homogeneous solution, (b) surface nucleation followed by diffusion, (c) coalescence of small bubbles, (d) interaction of active fission tracks, (e) fission track "explosion" followed by diffusion, and (f) charged bubble growth. These models failed for one or more of the following reasons: (a) they predicted that the void will decrease as time available for growth decreases, contrary to observation; (b) they required lifetimes for transitory physical and chemical reactions far in excess of any plausible value; (c) they required times for bubble growth in excess of that available; (d) they predicted a dynamics of void growth contrary to observation.

The only model studied which is consistent with the experimental data involves the repeated interaction between fission fragments and bubbles. In this model, it is hypothesized that during the interaction a fraction of the radiolytic gas produced in the fission track wake is transferred into the bubble either by surface tension or diffusion which leads to void growth. The initial bubbles are produced within the wake of a preceding fission fragment. They are believed to be limited to sizes of the order of $10^{-7}$ cm.

Experimental Observations

The basic characteristics of the void have been determined from experimental data. They will be reviewed briefly.

1. The appearance of expansion pressure in transients with periods less than 15 milliseconds is proof that the core contains voids.
2. The time available for growth decreases as the rate of void formation and the total amount of void produced increases. This fact is easily demonstrated from a study of void compensated reactivity as a function of period.
3. The void is virtually incompressible for pressure changes of up to 30 atmospheres, which have been produced by the expansion pressure. This fact leads to the conclusion the void must be present in the form of bubbles with high internal pressure. Small bubbles have the high internal pressures required to be consistent with this observation. This point removes from consideration bubbles with radii larger than $10^{-5}$cm, at least until after peak power has been reached.
4. The fact that the solution temperature can be raised to within a few degrees of the boiling point at the start of a transient without affecting dynamic behavior, and the relative pressure insensitivity of the void are both strong evidences that steam is not the shutdown mechanism.
5. The quantity of gas produced during a transient is sufficient to fill small bubbles to a high internal pressure. Radiolytic gas production represents 4% of the energy dissipated in the core.

Proposed Model of Void Growth

The proposed model is based on the following assumptions:

1. The very small bubbles being considered in this discussion are assumed to have a lifetime greater than 20 milliseconds. These bubbles are sub-

critical in size. Therefore, according to classical thermodynamics, they should collapse in times much shorter than 20 milliseconds. However, there is a considerable body of experimental evidence in the literature, (7), (8), (9), and (11) which can be used to support the contention that small bubbles or nuclei can exist for times substantially longer than this.

2. The space independent reactor kinetic equations can be used with appropriate coefficients to determine system void volume at any time during the transient, see references (2) and (11).

3. The average temperature of the gas in the void is assumed to be the same as that of the solution temperatures at the same time (12).

4. The number of moles of gas available in the system at any time during a transient is the product of the statically determined energy coefficient of gas production (G) and the energy release, or

$$E(t) = \int_0^t P(t')\,dt'$$

Therefore,

$$N(t) = G \cdot E(t)$$

5. It is assumed that the pressure inside the bubble due to surface tension is given by $\frac{2\gamma}{r}$, where $\gamma$ is the surface tension. In other words, the system hydrostatic pressure $P_\infty$ is negligible compared to surface tension pressure.

6. Finally, bubbles grow as the result of gas transfer from a fission track to the bubble in fission track bubble collisions.

<u>Treatment of an Ensemble of Small Bubbles, The Average Bubble Radius</u>

An ensemble of bubbles may be described in terms of several different weighted averages of bubble radius. The most useful average is one weighted by the number of moles of gas in each radius interval.

Consider an ensemble of gas bubbles where the number of bubbles with radius between $r$ and $r + dr$ is $\xi(r)$. Assume for the moment that the bubble size distribution function $\xi(r)$ is known and is a continuous function of bubble radius in the interval from the smallest bubble radius ($r_0$) which contains just one gas molecule per bubble up to a maximum radius bubble ($r_{max}$) for the ensemble.

The number of moles of gas ($n$) contained in any one bubble of radius $r$ is:

$$n = \frac{pV}{R_0 T} = \frac{\frac{2\gamma}{r} \cdot \frac{4}{3}\pi r^3}{R_0 T} = \frac{8\gamma\pi r^2}{3 R_0 T} \qquad (2)$$

where $R_0$ is the gas constant per mole and $T$ the absolute temperature. The total number of moles of gas contained in $\xi(r)$ bubbles of size $r$ in the radius interval $dr$ is:

$$dN = \frac{8\gamma\pi r^2}{3R_0T} \cdot \xi(r)\,dr \qquad (3)$$

and the total moles of gas in the bubble ensemble is:

$$N = \frac{8\gamma\pi}{3R_0T}\int_{r_0}^{r_{max}} r^2\ \xi(r)\,dr \qquad (4)$$

The mole-weighted average bubble radius is

$$\bar{r}_{mole} = \frac{\int r\,dN}{\int dN} = \frac{\frac{2\gamma}{R_0T}\int_{r_0}^{r_{max}} \frac{4\pi r^3}{3}\ \xi(r)\,dr}{\int_{r_0}^{r_{max}} \frac{8\gamma\pi r^2}{3R_0T}\ \xi(r)\,dr} \qquad (5)$$

The denominator of equation (5) is easily identified as the total number of moles of gas in the bubbles which make up the ensemble, $N$. By rearrangement of the numerator as indicated in equation (5) one is able to recognize the integral as the sum of the volume of each bubble making up the ensemble or the total volume occupied by bubbles $V_B$. Hence, equation (5) becomes

$$\bar{r}_{mole} = \frac{2\gamma V_B}{N R_o T}, \tag{6}$$

and by defining an effective bubble pressure $p_B$ as equal to $\frac{2\gamma}{\bar{r}_{mole}}$, then equation (6) becomes

$$p_B = \frac{N R_o T}{V_B} \tag{7}$$

which has the same form as the perfect gas law. The similarity in form between (7) and the perfect gas law suggests that the behavior of the ensemble under compression may be treated using $p_B$ and $V_B$ in Boyle's law. Detailed examination of this question has shown that the ensemble never exactly follows the Boyle's law relation. However, numerical investigation, assuming different bubble size distributions, has suggested that the errors introduced by using Boyle's law are relatively small ( $< 10\%$ ). Unless bubble size distribution anomalies exist in the fuel solution which renders the deduction based on these numerical computations greatly in error, one can say that $\frac{1}{p_B}$ is a good measure of the compressibility $\left[\frac{1}{V}\left(\frac{\partial V}{\partial p}\right)_t\right]$ of the bubble ensemble.

The value of the mole-weighted average bubble radius is readily computed from the experimental data. The volume occupied by the bubbles is obtained from the void compensated component of the reactivity and the void coefficient of reactivity. The number of moles of gas produced is simply the energy released times the static gas production coefficient. The value of $\bar{r}_{mole}$ can be computed at any time during the transient without knowledge of $\xi(r)$.

In Figure (1) the value of $\bar{r}_{mole}$ at the time of peak power is plotted <u>versus</u> reactor period for a group of KEWB transients. In the period range from 20.0 sec to 0.1 sec, the energy release up to the time of peak power varies less than $\pm 10\%$ of the average value. However, in the same period range the average bubble size at peak power decreases rapidly as the period decreases. Gas filled voids are ineffective as a shutdown mechanism in this region because the gas is contained under high pressure, i.e. small volume. As the period becomes shorter than 0.1 second the bubble size increases from about $10^{-6}$ to $10^{-5}$cm. Hence, the internal pressure of the gas in the bubbles will be between 140 and 14 atmospheres. Fuel solution containing bubbles will have a much larger compressibility than that of the bubble-free solution; however, as required by experiment, the compressibility is still small enough so that the expansion pressure has relatively little effect on total void volume.

The change in mole-average bubble radius as a function of time during the power excursion is shown for a fast transient ($\tau = 3.74$ milliseconds) in Figure (2). The validity of the mole average bubble radius has been checked by comparing the time it takes sound to traverse the core with that computed using the bubble radius of Figure 2 and the volume fraction of gas present in the core (13). The comparison is made in the following way:

It will be shown later in this paper that from both experimental and theoretical considerations that the rate of void production is proportional to the product of power and energy, i.e., proportional to $P \cdot E$ (see equation 16). Void formation is the driving force for the production of expansion pressure. Hence, the time lag between the peak of the $P \cdot E$ driving function and the observed peak in the expansion pressure is a measure of the time required for sound to cross the fuel solution. This time lag from the experimental data is about 1.1 milliseconds corresponding to a speed of sound of 196 m/sec. The value computed from bubble size and volume is 200 m/sec which is in excellent agreement. While this agreement may be somewhat fortuitous, it does indicate that equation (6) is a good description of the void.

Later in the transient (shown in Figure 2) in the time region between 43.8 milliseconds and 52 milliseconds a shock wave, which was formed by the constricting effects at the top of the core, required 8.2 milliseconds to reach the bottom of the core. This corresponds to sound velocities of 38 meters per second. A bubble size of about $5 \times 10^{-5}$ cm would yield this velocity of sound when the original core solution just filled the sphere giving a 15% void volume. The average bubble radius computed from equation (6) in this time interval, is about $4 \times 10^{-5}$ cm, again in excellent agreement.

## Interaction Model of Bubble Growth

In the preceding section we have shown that a meaningful physical description of the void is possible in terms of an average bubble radius computed via equation (6). We therefore are now in a position to investigate quantitatively the concept that collisions between fission tracks and bubbles result in the bubble growth phenomenom observed in the KEWB reactor.

The number of collisions per unit time between a fission fragment and a bubble will be the product of the fission fragment flux times the microscopic bubble cross-section. The average fission fragment flux is proportional to the average reactor power $P(t)$. The constant of proportionality can be readily shown to be the product of the conversion factor between power and number of fission fragments per second, $6.1 \times 10^{16}$/Mw, the average range of the fission fragment $\overline{L}$, and reciprocal core volume $\frac{1}{V_{core}}$, that is:

$$\overline{\varphi}_F(t) = \frac{6.2 \times 10^{16}\,\overline{L}}{V_{core}} \cdot P(t) \qquad (8)$$

The microscopic bubble cross-section is defined as the geometrical projected area of the bubble, $\pi r^2$. The number of collisions per bubble in a second is the fission fragment flux times the microscopic cross section. Hence, it is apparent that the large bubble will have a much higher probability of collision than a small bubble. If each collision leads to bubble growth, then the chance of future interactions increases rapidly with each collision.

The macroscopic bubble cross section ( $\Sigma_B$ ) is the sum of the microscopic cross sections of each bubble present in a cubic centimeter of fuel solution. The number of bubbles per unit volume of the solution is:

$$\text{Number of bubbles} = \frac{1}{V_{core}} \int_{r_0}^{r_{max}} \xi(r)\, dr \,. \tag{9}$$

Therefore,

$$\Sigma_B(t) = \int_{r_0}^{r_{max}} \frac{\pi r^2}{V_{core}} \cdot \xi(r)\, dr \tag{10}$$

And, with recourse to equations (4) and (1), equation (10) becomes:

$$\Sigma_B(t) = \frac{3 R_o T G}{8 \gamma V_{core}} \cdot E(t) \tag{11}$$

The resulting macroscopic cross section is independent of bubble size, or the bubble size distribution function. Since the lower limits of the integration included bubbles with just one molecule of gas per bubble, no distinction need be made between dissolved gas and gas contained within larger bubbles. It is clear that the void volume and gas volume are not identical. In particular, a one molecule bubble will not contribute to the void. However, as a result of the $r^2$ in the averaging processes discussed earlier, this distinction leads to small departures from the description given by equation (6). The number of collisions between bubbles and fission fragments is, then

$$C_{F \underset{\rightarrow}{(t)} B} = \Sigma_B(t) \cdot \overline{\varphi}_F(t) = \frac{\text{collisions}}{\text{cm}^3 - \text{sec}} \tag{12}$$

Substituting equations (8) and (11) into (13) gives

$$C_{F \underset{\rightarrow}{(t)} B} = \frac{3}{8} \cdot \frac{6.2 \times 10^{16} R_o T G \overline{I}}{\gamma V_{core}} \cdot P(t) \cdot E(t) \tag{13}$$

Letting $\epsilon$ be equal to the constant and approximately constant terms, equation (13) becomes

$$C_{F \underset{\rightarrow}{(t)} B} = \epsilon \cdot P(t) \cdot E(t) \tag{14}$$

The value of $\epsilon$ is

$$\epsilon = 3.3 \times 10^{14} \quad \frac{\text{collisions}}{\text{cm}^3 \, (\text{Mw-Sec})^2} \tag{15}$$

It is most interesting to note that the number of collisions per unit time is dependent only on the product of power and energy. The analysis of the experimental data shows that the void volume also depends only on the product of power and energy, (2) and (10), that is

$$\frac{dV}{dt} = \nu \, P(t) \cdot E(t), \tag{16}$$

or (16) a

$$V = \frac{1}{2}\nu\left[E(t)\right]^2$$

The value of $\nu$ determined from the experimental data is 500 $\frac{\text{ml}}{(\text{Mw-sec})^2}$. This value is based on the conventional six delayed neutron group reactor kinetic equations. The discovery of an inhour equation anomaly, which is believed to result from the moderation of fast neutrons in the graphite reflector followed by their slow diffusion back into the core, has complicated the problem. The effect has been dealt with by assuming the neutrons behave like an additional delayed neutron group. The parameters of this seventh group have been tentatively assigned a value which gives a $\nu$ of about 72 $\frac{\text{ml}}{(\text{Mw-sec})^2}$. This latter figure, while not exact, is undoubtedly a more meaningful number than that obtained from the conventional six group reactor kinetics equations.

If a volume of gas transferred into the bubble per collision is a constant amount $K$ per collision, then the rate of void growth will be

$$\frac{dV}{dt} = K \cdot V_{core}\, C_{F \underrightarrow{(t)} B} \tag{17}$$

The number of collisions per second per unit volume must be multiplied by the core volume $V_{core}$ to obtain the total rate of void growth. Returning to equation (14) for the number of collisions, equation (17) becomes

$$\frac{dV}{dt} = K\,\epsilon\,V_{core}\,P(t)\cdot E(t) \tag{18}$$

where $K$, $\epsilon$ and $V_{core}$ are constant. The form of equation (18) is now identical to the experimentally determined empirical equation (16). Therefore, the constant terms can be equated so that

$$\nu = K \epsilon V_{core} \tag{19}$$

and the value of $K$ can be determined as $1.9 \times 10^{-16}$ ml / collision.

An alternate assumption to the constant volume increment per collision is the concept that a constant number of moles are transferred on the average per collision. Consider one bubble, then let a constant average number of moles of gas $(\bar{n})$ be deposited in the bubble per collision. Then, the radius $r_i$ of the bubble after the $i^{\underline{th}}$ collision will be

$$r_i = \left[\frac{3R_oT}{8\gamma\pi}(\bar{n}\cdot i + n_o)\right]^{-1/2} \tag{20}$$

Where $n_o$ is the number of moles of gas in the original bubble. The initial bubbles produced without interaction in the fission track wake are believed to have a radius comparable to the radius of the intensely ionized region of the track which is probably about 50 Å. A bubble 50 Å in radius contains $3.4\times10^{2}$ molecules of gas. From the static gas production coefficient, one can compute that $1.7\times10^{6}$ gas molecules are produced in each fission track wake. If only 1.0% of the gas in the track were transferred to a bubble per collision, then it is clear that the number of moles of gas in the bubble would be negligible. Therefore, $n_o$ in equation (20) is negligible and the volume of the bubble $V_i$ after the $i^{\underline{th}}$ collision is

$$V_i = \frac{4\pi}{3} r_i^3 = \frac{4\pi}{3}\left(\frac{3R_oT}{8\gamma\pi}\right)^{3/2}(\bar{n}\cdot i)^{3/2} \tag{21}$$

The next step is to compute the microscopic cross section for the non-collided, 1st, 2nd, ....collided bubble groups. This computation requires a knowledge of the bubble size distribution function $\xi(r)$ as a function of time during a transient. No attempt has been made to solve this problem. However, progress can be made in a more restricted case where it is assumed that only one collision per bubble can occur. Under this restriction $i = 1$ and equation (21) becomes

$$V_{i=1} = \frac{4\pi}{3}\left(\frac{3R_oT}{8\gamma\pi}\right)^{3/2}(\bar{n})^{3/2} \tag{22}$$

All of the terms in the right hand side of equation (22) are constant, therefore $V_{i=1}$ is constant and is equal to the volume of gas transferred per collision, but this is equal to $K$ in equation (12). Therefore, the assumption of only one hit per bubble converts the constant-mass-transferred-per-collision model into the constant-volume-transferred-per-collision model. Setting equation (22) equal to the value of $K$ ($1.9\times10^{-16}$ ml/collision), and solving for $\bar{n}$, we obtain $3.0 \times 10^{-19}$ moles, or $1.8 \times 10^{5}$ molecules per collision. The value of $\bar{n}$ is, therefore, a factor of 1000 larger than $n_0$. This shows the assumption that $n_0$ can be neglected is valid.

The number of molecules transferred is about 10% of the gas available along the fission fragment track. Therefore, it is apparent that more than sufficient radiolytic gas is available in the track, and that the number of collisions are adequate to explain the KEWB reactor bubble growth. The restriction of one collision per bubble is not very realistic; therefore, 10% of the gas available is an upper limit for transfer from a fission track to a bubble. The more reasonable constant mass transferred per collision theory which permits multiple collision will have the effect of decreasing the value of $\bar{n}$.

The more general computation of void growth assuming multiple collisions with constant mass transfer will resemble the constant volume computation in the early stages of the transient. In this time region of the transient relatively few single collided bubbles will be present compared to the non-collided bubbles. As the transient proceeds, ever, the inventory of once hit bubbles will increase so that multiple collisions will become important. When this happens, the void growth will be faster than that given by the power and energy product model. The difference between the two models could be expressed in terms of some appropriately averaged $\bar{l}^{3/2}$. The value of $l$ may be relatively unimportant until after peak power in the very fast transients.

Preliminary analysis of the experimental data using the 7th reflector delayed neutron group suggests that the gas volume is proportional to a power of energy release higher than the square given by the PE model, especially in the post peak power region. A more detailed study of the void growth as a function of time will be possible as soon as the parameters characterizing the 7th group have been more accurately determined.

## Conclusions

1. The model is consistent with the experimentally observed reactor behavior.
2. The model quantitatively predicts the production of a void consisting of very small bubbles.
3. The model leads to a rate of void production which increases as the reactor period decreases, which is consistent with experimental data.
4. The interactions have a sufficiently high probability of occurrence to explain void growth.
5. The gas transferred from a fission fragment track to a bubble during the collision is but 10% of that produced in the track.
6. The appearance and character of the expansion pressure is consistent with the model.

## Summary

The void is considered to consist of an assemblage of radiolytic gas bubbles. It is shown that the void obeys the same equation of state as the individual bubble, providing the correct average bubble radius is used. This average radius leads to a prediction of the velocity of sound in agreement with experimental data.

The rate at which bubbles are hit by fission tracks is shown to be proportional to the product of power and energy. The rate of void growth is also (approximately) proportional to the product of power and energy. This has lead to the conclusion that collisions between fission fragments and bubbles are responsible for void growth.

The interaction model is still under development. So far no major objections to the theory have developed, but the matter is being explored further with a capsule type of experiment where void production will be measured directly as a function of time. It is felt that some of the details of the theory may change with time, but the broad picture of void consisting of small bubbles and growth as a result of interaction will remain.

## Acknowledgments

The author is indebted to members of the KEWB staff who obtained the extensive and excellent experimental information upon which this paper is based and performed the preliminary data analysis. J. W. Flora and Amos Norman encouraged the work and contributed many helpful suggestions.

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# AVERAGE BUBBLE RADIUS AT TIME OF PEAK POWER-vs-REACTOR PERIOD

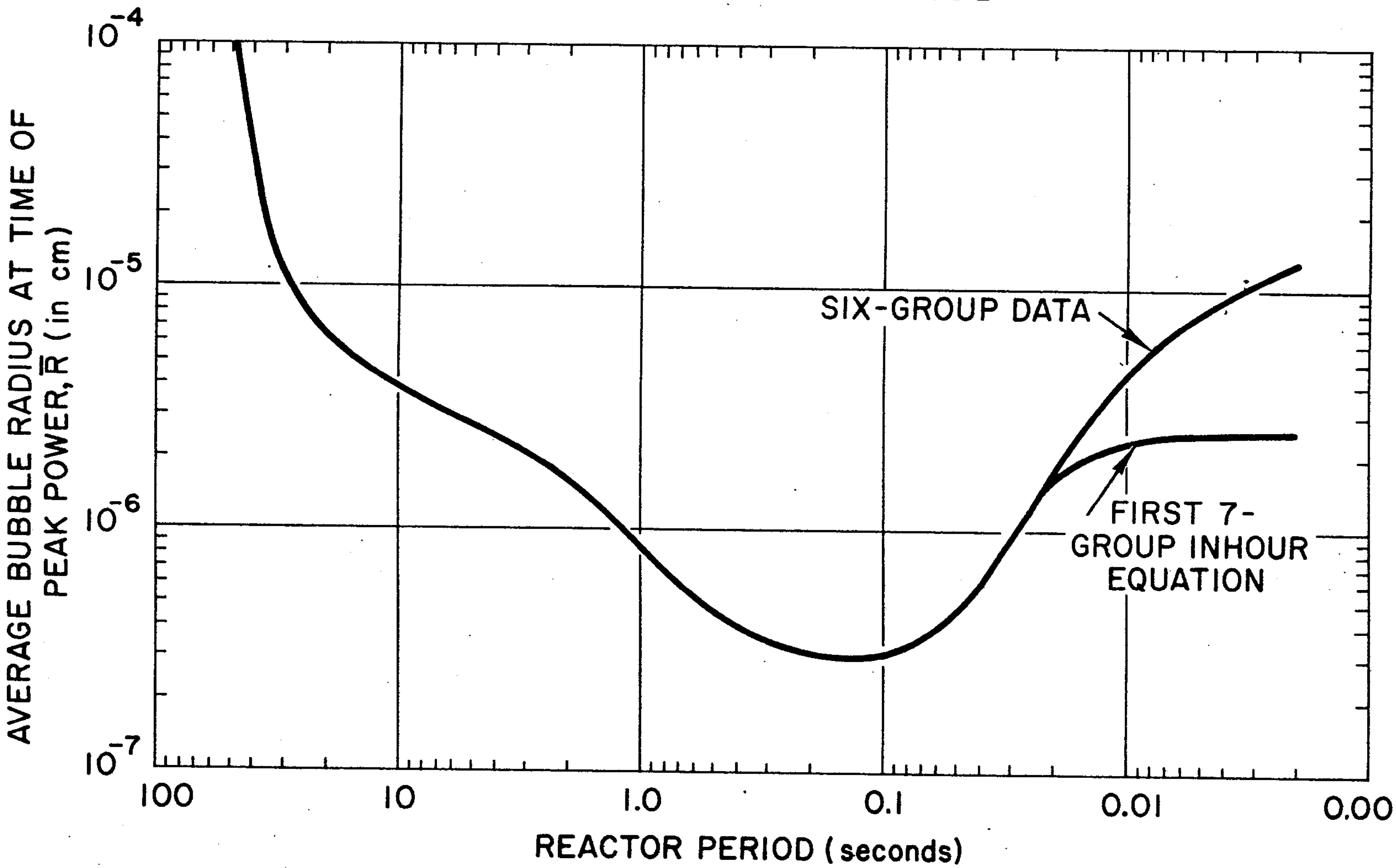


FIGURE 1

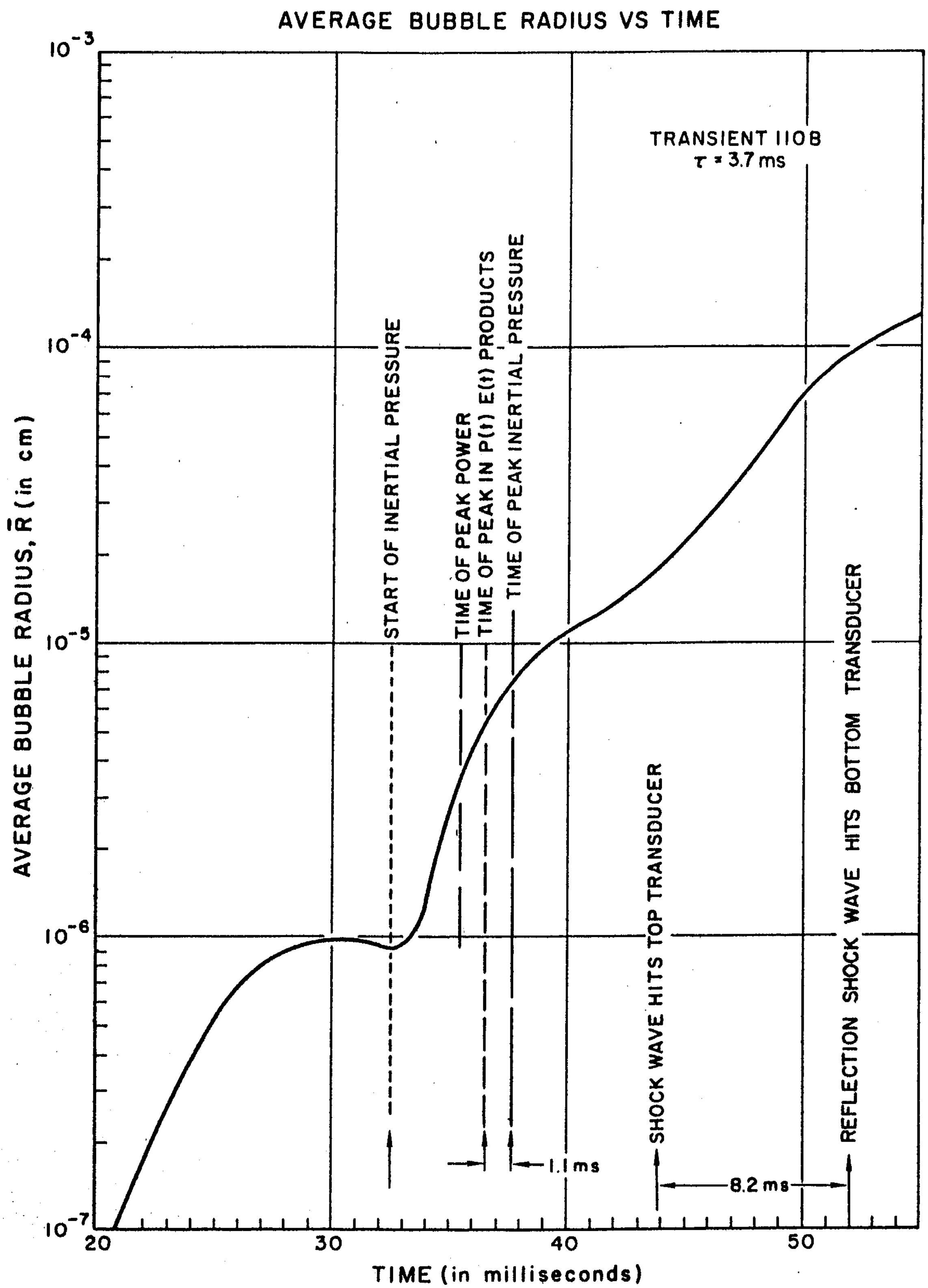
AVERAGE BUBBLE RADIUS VS TIME
TRANSIENT 110B
τ = 3.7 ms
START OF INERTIAL PRESSURE
TIME OF PEAK POWER
TIME OF PEAK IN P(t) E(t) PRODUCTS
TIME OF PEAK INERTIAL PRESSURE
1.1 ms
SHOCK WAVE HITS TOP TRANSDUCER
REFLECTION SHOCK WAVE HITS BOTTOM TRANSDUCER
8.2 ms
AVERAGE BUBBLE RADIUS, R̄ (in cm)
TIME (in milliseconds)
10⁻³
10⁻⁴
10⁻⁵
10⁻⁶
10⁻⁷
20
30
40
50

FIGURE 2