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Production of gas bubbles in fluids by tribonucleation

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IKELS, KENNETH G. *Production of gas bubbles in fluids by tribonucleation.* J. Appl. Physiol. 28(4): 524–527. 1970.—This report describes a mechanism, called tribonucleation, for producing gas nuclei by making and breaking contact between solid bodies which are immersed in liquid. A metal ball was rolled inside a glass tube filled with liquid which contains dissolved gas. Formed nuclei may grow to visible bubbles depending on the dissolved gas concentration and pressure applied to the liquid. Unlike other possible mechanisms for forming bubbles, tribonucleation is capable of producing nuclei under relatively mild experimental conditions, such as may be encountered in vivo. The experiments show that viscosity and velocity of separation of surfaces are important determinants of whether or not nuclei will form.

bubble formation: decompression sickness

TO THE AVIATOR, the space traveler, and the diver, whose activities normally require changes in barometric pressure, the avoidance of decompression sickness obviously is of utmost importance. If the symptoms of decompression sickness are the result of bubbles in the body (as is commonly thought), then a logical way to avoid decompression sickness is to minimize or prevent the formation of nuclei from which bubble growth occurs. In spite of this obvious fact, the actual definition and description of the factors responsible for the production of nuclei has received relatively little attention by those interested in decompression sickness. In fact, the important distinctions between gas nuclei and overt bubbles appears to have been largely ignored, due in part, perhaps, to the absence of a completely satisfactory physical description of what constitutes a gas nucleus.

The physical concept of the nucleus which is most consistent with previous experimental results has been summarized by Knapp (11). He concludes that a nucleus is a pocket of undissolved gas in a small crevice in the surface of a solid that is hydrophobic to the liquid and that the effectiveness of the nucleus as a precursor to formation of overt bubbles can be reduced by pressurization or centrifugation (7, 11). For example, if a plentiful supply of nuclei of various sizes are available, bubble growth can occur when the change in pressure is small. On the other hand, if nuclei are absent by virtue of elimination from the liquid by means of centrifugation or by pressurization, the liquid is stable to large changes in pressure which would otherwise result in profuse bubble formation. Theoretical calculations based on various properties of the liquid predict that cavitation of water requires negative pressures equivalent to thousands of atmospheres (6). In practice, with extremely careful technics, pressure changes of about minus 281 atm have been recorded before water cavitated (2).

The difficulty of cavitating liquids solely by pressure changes and the apparent need for nuclei as prerequisites for bubble growth are probably the most relevant observations from which to construct a comprehensive understanding of the biophysical basis of decompression sickness. Inasmuch as the onset of decompression sickness is the result of fairly modest changes in ambient barometric pressure, it is reasonable to suspect the presence of more or less permanent nuclei in the circulating blood. Harvey et al. (7) demonstrated, however, that the blood of cats, rabbits, and dogs (which were at rest and had never been exposed to either low or high pressures) was free of bubble-forming nuclei when tested by decompression to slightly below its vapor pressure. These observations, which have been repeated on human blood, suggest that nuclei are normally not present in circulating blood although they may be trapped on the surface of blood vessels. A special mechanism must be required to form these nuclei from which overt bubbles can form under proper conditions. The experiments reported here have been designed to evaluate one such possible mechanism—tribonucleation, or the production of nuclei by separating surfaces immersed in a liquid.

METHODS AND MATERIALS

According to the literature there are several mechanisms of cavitation and subsequent bubble formation that have been studied. These include cavitation by high-energy particles (13), ultrasonics (14), turbulent flow (5), random nucleation (9), and mechanical stress (7). The mechanical stress theory, which would include the concept of negative tension that may be produced intermittently during muscular contraction and relaxation, has served as a useful model to explain cavitation from which bubble growth occurs in biological systems. The application of this idea. however, apparently fails to explain those cases of decompression sickness where muscular activity is minimized.

Hayward (8) first observed that nucleation can occur in a liquid by a gentle rubbing action between two surfaces, and he called this tribonucleation. Campbell (3) later proposed a theory to explain Hayward's observations. His reasoning was based on the theory of viscous adhesion (4) that utilizes a model in which two adjacent circular plates having equal radii are caused to separate, developing a negative hydrostatic tension in the center of the plates. The equation that illustrates the theory was originally derived by Stefan, cited by Banks (1), and takes the form

$$A - P_t = 3\eta U(R^2 - r^2)h^{-3} \qquad (1)$$

where A = pressure action on the liquid; P_t = hydrostatic tension; η = viscosity; U = velocity of the plate separation; R = radius of the plates; r = distance from the center of the plates; and h = distance of plate separation.

Equation 1 predicts that at sufficiently high values of viscosity (η) and velocity (U) and a proper ratio between the difference in radii (R, r) and the distance of separation (h), negative values of hydrostatic tension (P_t) can occur which theoretically are sufficient to fracture the liquid. The theoretical value can be considerably reduced if there is incomplete wetting of the surfaces or if the surface tension of the liquid is lowered.

If a cavity is created as a result of separation of the plates, dissolved gases in the liquid immediately begin to diffuse into the cavity. As the plates separate further, the negative tension (P_t) produced by viscous adhesion is decreased and will eventually

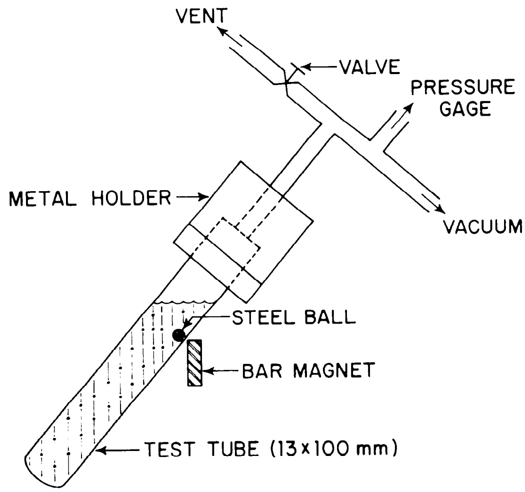

FIG. 1. Test apparatus for observing effects of viscosity and velocity.

approach zero. When this occurs the nuclei thus formed become governed by the familiar relation

$$P_g - P_t - P_a = \frac{2\gamma}{r} \quad (2)$$

where P_g = tension of the dissolved gases; P_t = hydrostatic tension; P_a = pressure acting on the liquid; γ = surface tension; and r = radius of the bubble. It is important to realize that eventual reabsorption, stabilization, or growth of the nuclei to a visible bubble form, as predicted by this equation, are all events which must be preceded by the actual nucleation process and that this process is affected by different physical factors, the most prominent of which is the viscosity of the liquid.

It would be technically difficult to construct the apparatus that Campbell conceived of in his theoretical work, especially the small plates which could separate at various known velocities. The apparatus used in this study, shown in Fig. 1, incorporates a small stainless steel ball (0.3175 cm) that upon removal of a magnet permits the ball to roll down the side of a test tube (a distance of 3.5 cm). The test tube is connected to a vacuum pump through a metal adapter containing an O-ring seal. The whole system can be tilted to any desired angle to change the velocity of the ball. This arrangement permitted observations on the effects of liquid viscosity and ball velocity on nuclei and bubble formation using various liquids and pressures. In this experimental design, therefore, the essential features of Campbell's theoretical model, requiring surfaces to continuously contact and separate at a controlled and repeatable velocity, has been duplicated.

Considerable effort was devoted to perfecting technics of handling aqueous solutions with different gases. The most troublesome problem with such solutions is the container itself. Glass (Pyrex) tubes must be specially cleaned. The technic that has proven satisfactory consists of boiling the tubes in Na_3PO_4 and then thoroughly rinsing them with distilled water. The tubes are then transferred to an initially hot (70 C) chromic acid solution, and left in this solution at room temperature until required. When needed, the tubes are again thoroughly rinsed with distilled water and the sample immediately introduced. At no time during this entire process was the tube permitted to dry. Water, supersaturated with gases and placed in these tubes, can be decompressed to below its vapor pressure without forming bubbles. However, bubbles form much more easily in water in a tube that has been dried. These bubbles are apparently due primarily to gas masses trapped on the surface of the tube or in small cracks or crevices. Organic liquids (ethyl alcohol, benezene, and olive oil) when carefully poured into similar clean but dry tubes, do not produce bubbles when decompressed. The lack of nuclei is thought to depend on the physical relations at the air-liquid-glass interface where contact angles must be small or zero. Under these conditions, it is impossible to sustain nuclei.

Test liquids were placed in cleaned tubes and saturated by bubbling with various gases at ambient barometric pressure (He, Ar, and N_2). The steel ball was included during the saturation process. After 1 hr of saturation the tube was centrifuged at 3,000 rpm to rid the liquid of small bubbles and then placed in the test apparatus (Fig. 1). The pressure above the liquid was lowered to the vapor pressure of the liquid, or as low as possible, to check for the production of bubbles. If none appeared, the pressure was then increased to a predetermined value and the ball permitted to roll down the tube at a rate set by the inclination of the tube and the viscosity of the liquid. The liquid was observed by a stereomicroscope (magnification ×10) for the appearance of bubbles. If none appeared, the procedure was repeated until the pressure at which bubbles finally appeared was established. The criterion used to establish this pressure was the appearance of bubbles with a decrease of 40 torr below the lowest pressure at which no bubbles were observed. All tests were conducted at room temperature (25 ± 1 C).

A liquid of moderate viscosity was first tested. Olive oil was used not only because of its viscosity (71.0 cp) but also because of its high gas solubility and low vapor pressure.

In the case of olive oil, once the pressure for bubble appearance had been established, 1 ml of the sample was removed and analyzed for gas content by a chromatographic technic (10).

To establish the decompression required for the olive oil with less than 1 atm saturation, a gas-saturated sample was exposed to a low pressure (approximately 60 torr). At this pressure the ball was rolled over the inner surface of the tube by means of a small magnet. This caused the formation of numerous bubbles thereby reducing the concentration of the dissolved gas. The period of producing bubbles was varied in accordance with the approximate concentration of dissolved gas desired. The sample was then centrifuged and the decompression for bubble formation determined. One milliliter of the sample was then analyzed for gas content and the Ostwald coefficient (L) determined, the Ostwald coefficient being defined as

$$L = \frac{Vg}{Vs}$$

where Vg = volume of gas dissolved and Vs = volume of solvent. Having previously determined the Ostwald coefficient at 1 atm saturation, the relative gas content can readily be calculated from the following expression

$$\text{relative gas content} = \frac{L}{L\ (1\ \text{atm saturation})}$$

RESULTS

A preliminary effort was to determine the effectiveness of the cleaning procedure of the test tubes and to test for the presence of nuclei in circulating human blood. Twenty-nine blood samples from the antecubital vein on 17 human subjects were collected. A portion of each heparinized sample was transferred from the syringe and then immediately decompressed in specially clean, wet tubes to approximately 15 torr and retained at this pressure for 10 min. Of the 29 samples, 1 formed bubbles at about 60 torr. After 10 min at 15 torr, the pressure was further reduced to approximately 5 torr. At this low pressure, 11 samples formed bubbles at times ranging from 10 to 40 min, and 17 samples showed no visible evidence of bubbles after 45 min. These observations support and confirm the findings of Harvey et al. and demonstrate that under the conditions employed a liquid as complex as blood requires extremely severe conditions for bubbles to form as a result of decreased pressure. These observations, as previously noted, suggest that significant numbers of nuclei are rarely present in blood and also that a special mechanism must be required to form bubble precursors.

The use of separating surfaces as the mechanisms for producing overt bubbles in liquids has provided the following experimental results.

Gas solubility effect on ΔP at constant viscosity and velocity. Figure 2 shows the results of using olive oil as the test liquid under conditions that included various concentrations of He, N_2, and Ar and a 1-sec roll of the steel ball. The amount of decompression required for bubble formation increased as the relative gas concentration decreased. There appeared to be a tendency for Ar to require a lesser decompression and He a slightly larger decompression than N_2; however, insufficient data preclude an analysis of the significance of this difference. The amount of gas supersaturation required for bubble formation is of the order of 100 torr as shown by the upward displacement of the data with reference to the line of gas saturation.

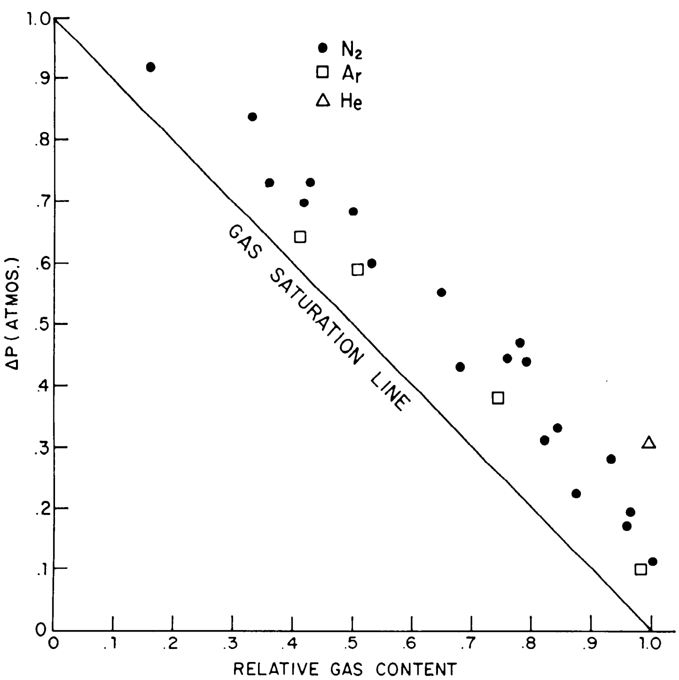

FIG. 2. Decompression required for bubble formation and growth in olive oil at various gas concentrations. Each point represents the midpoint of 40-torr pressure excursion between bubbles and no bubbles.

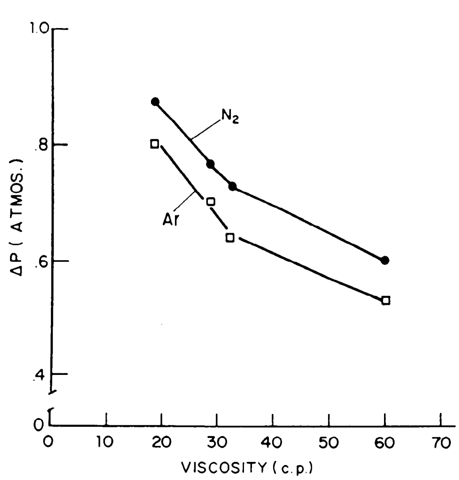

FIG. 3. Decompression required for bubble formation and growth in gas saturated glycerol-water solutions of various viscosities. Each point represents the midpoint of 40-torr pressure excursion between bubbles and no bubbles.

TABLE 1. *Results of cavitation and bubble formation in various liquids by tribonucleation (temp, 25 C)*

Liquid or Solution	Viscosity, cp	Gas, 1 atm Saturation	Time Steel Ball Travels 3.5 cm, sec	Decompression, ΔP, No Bubbles to Bubbles, torr
Olive oil	71.9	Ar	1	70–110
		N_2	1	85–125
		He	1	225–265
		Ar	4	270–310
		N_2	4	285–325
		He	4	410–450
Glycerol-water	39.5	Ar	1	405–445
		N_2	1	470–510
		He	1	450–490
		Ar	5	510–560
Glycerol-water	60.1	Ar	1	380–420
		Ar	5	450–490

Viscosity effect on ΔP at constant velocity. The results of glycerol-water solutions of various viscosities saturated with N_2 or Ar are shown in Fig. 3. The values clearly demonstrate that as the viscosity of the solution increased, the ΔP or decompression required for bubble formation decreased. In this case also, the ΔP required to produce visible bubbles appeared related to the solubility of the gases, the more soluble gas Ar required less of a ΔP. It is equally important to realize that the solubility of N_2 and Ar in the glycerol-water solutions is greatly decreased with the increased viscosity obtained with high glycerol concentration. This fact emphasizes the importance of viscosity in bubble formation inasmuch as ΔP would normally increase with decreasing gas content, as seen in Fig. 2. Instead of increasing with decreasing solubility, the ΔP decreases in accordance with the increasing viscosity of the solution. At comparatively high viscosities (beyond 40 cp) the solubility of gases in glycerol-water is greatly diminished. The fact undoubtedly accounts for the apparent independence of the ΔP between viscosity of 40–60 cp.

Velocity effect on ΔP at constant viscosity. Table 1 shows that when velocity of the steel ball is high, bubble formation is facilitated. In each of the three liquids tested, as the velocity decreased from a 1- to 5-sec roll over the prescribed 3.5 cm, the ΔP required for bubble formation increased. These values illustrate the importance of the rate of separation of the surfaces as well as the viscosity of the liquid and gas solubility.

When water alone was the liquid (η = 0.89 cp) and N_2 was the saturating gas, no bubbles formed using a 1-sec roll, even when the pressure above the water was lowered to below its vapor pressure.

DISCUSSION

The experimental evidence presented in this report emphasizes the importance of the viscosity-velocity product in bubble formation by tribonucleation. When considering cavitation and subsequent bubble formation by this means, the tension (P_t) must remain sufficiently high during the time of separation of the surfaces to maintain the stability of the cavity or bubble, or both.

Should the tension (P_t) return to zero before the conditions required for *equation 2* are satisfied, the cavity or bubble may collapse. On the other hand, if the conditions set forth in *equation 2* are achieved, so that bubble growth is favored, then the bubble will mature, i.e., become visible and rise to the surface of the liquid.

These observations have shown that undisturbed gas-supersaturated solutions of water, glycerol-water, and olive oil do not form bubbles when care is exercised as to the cleanliness and wettability of the container. On the other hand, the phenomenon of tribonucleation produces cavitation and leads to subsequent bubble formation under conditions that require relatively moderate decreases in barometric pressure and which are rationally related to basic physical properties of the liquid.

Tribonucleation may conceivably manifest itself by various means in the biologic system for there are numerous sites where surfaces may contact and separate or rub across one another. Most obvious, perhaps, would be the articulating surfaces of joints where synovial fluid may display a high viscosity similar to the solutions tested here (12). Other possible sites include small circulatory vessels that may collapse and expand in response to various physiologic requirements or muscle tendon inserts to bone joints. Many other examples are undoubtedly possible.

The study of tribonucleation is continuing with the hope that the conditions controlling cavitation and bubble formation can be reduced to a set of strictly definable physical variables, the measurement and integration of which will be meaningful to human decompression sickness.

Research reported in this paper was conducted by personnel of the Biophysics Branch, Physiology Division, US Air Force School of Aerospace Medicine, Aerospace Medical Division (AFSC), Brooks Air Force Base, Texas. Further reproduction is authorized to satisfy the needs of the US Government.

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