THE METALLOGRAPHY
OF
DENTAL AMALGAM

A survey of the Influence of Manipulative
Variables on the Physical Properties and
Microscopic Structure of Dental Amalgam.

A thesis submitted to the University of
Sydney in support of my candidature for
the degree of Doctor of Dental Science.

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The original work presented in this thesis has been carried out by me as an individual research project. I have personally prepared, tested and examined all the specimens used in this survey and all photomicrographs were taken by me.

I gratefully acknowledge the metallurgical facilities made available to me by Dr. L.E. Samuels of the Defence Standards Laboratories, N.S.W.

During the course of this investigation I have had much encouragement and I particularly wish to thank Professor J.S. Lyell of the University of Sydney and Mr. A.R. Docking of the Commonwealth Bureau of Dental Standards and his staff with whom I have had many discussions.
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- INTRODUCTION -
"Silver Amalgam", is undoubtedly the material of greatest importance to Dentistry.

The use of amalgam occupies a considerable proportion of the time of both the "average dentist" and the "average dental office patient", and this material has saved and continues to save more teeth than all other restorative materials.

Although amalgam is often called the "bread and butter" of dentistry, it is far from being a despised material, and properly placed restorations using this material will most satisfactorily and economically provide many years of service to the patient.

As befits this material, a wealth of material has been published following voluminous research into aspects of chemistry, metallurgy, manufacture and physical properties of silver amalgam. Research findings have been correlated with and translated into clinical practice to enable our previously mentioned "average dentist" to provide a better service for his patients.

Despite the considerable amount of research already carried out on silver amalgam there is an ever increasing need for further advancement of the knowledge concerning the properties and manipulation of this material.
The need for further research into fundamental properties of amalgam arises for several reasons:

(i) As new facts emerge and these are used by manufacturers to improve their products, the properties of the materials vary and there is again a need for further investigation.

(ii) Much of the present knowledge of amalgam has been handed down for thirty or more years, sometimes having been verified but sometimes merely passing into the realm of "it has been accepted that" until finally it is accepted and oft quoted without its basis, origin, or reliability being known.

(iii) Many workers have carried out investigations on particular aspects of the properties of amalgams. Frequently an investigator wishes to investigate the effect of a particular technique or a major property but includes other aspects as side issues. The result of this type of research is that the knowledge of silver amalgam is a collection of facts compiled by a very large number of workers under different conditions, some simulating clinical conditions, others being merely experimental techniques designed for standardization of experimental results.

The greatest advancement of knowledge in any field is invariably and of necessity made as in (iii) above.

Observing the above facts, I felt that there was a need for an investigation to be carried out by a single individual using techniques, and variables as may be used and introduced clinically but under controlled
laboratory conditions. In this way it is hoped to verify past findings, extend these findings if possible and perhaps make some recommendation for clinical use of amalgam.

So a research project was born.

"To observe the effects of manipulative variables on the physical properties and the microstructure of silver amalgam".

The manipulative variables were to be many. Ward, Peyton and Scott (1937) listed thirteen manipulative variables which they suggested may be of importance in amalgam work. All of these and perhaps more variables were to be covered with the proviso that only variables within the control of the dentist were to be considered and manufacturing variables were not relevant to this survey. Alloys were to be used as purchased on the open market, no co-operation with manufacturers being sought as this may tend to cloud the issue and obscure its value.

The physical properties to be investigated were the major ones considered of importance to clinical practice and for specification of alloys viz. Dimensional Change, Strength, Flow.

While the importance of other physical properties is realised, an accurate assessment of their clinical value is difficult and as limits had to be imposed, detailed consideration of these has been omitted.

I hoped that a study of the microstructure of amalgam may be of value when considered in conjunction with manipulative variables and
physical properties so this was included in the project.

A short review of the literature is included before detailing the experimental work carried out. I feel that a separate review is desirable rather than a bibliography appended to an original investigation.

The literature concerning amalgam is reviewed only in so far as it is pertinent to the investigation carried out and little credit has been given to the large volume of literature which is merely a review of other work, or based on theories or pure conjecture.
CHAPTER I. HISTORY OF RESEARCH ON DENTAL AMALGAM.

That amalgam is with us today is fortunate. To have survived the interpretation placed on its use by the Crawcour brothers and then the Amalgam War 1841 - 1850 with its following "New Departure Creed" to counter the "Accepted Creed" (Sweet 1959) speaks highly for this material.

In view of the empirical nature of the origin of amalgam, there is little wonder that doubt regarding this new material and charlatanism regarding its use existed.

The first experiments of a scientific nature were carried out on amalgam as late as 1861 by John Tomes and although an attempt was made to scientifically investigate the alloys available by men such as Charles Tomes (1871 - 73), Kirby (1871), Hitchcock 1874, Flagg (1882, 1884) and others at this time the first real advance came with the entry of Dr. G.V. Black onto the amalgam scene in 1895.

Black's work in this and ensuing years gave us much fundamental knowledge concerning the composition, manufacturing, and testing of the physical properties of amalgam. Black gave us the balanced alloy and taught us how to manipulate it.

After the commencement in 1919 of investigations on amalgam alloys by the National Bureau of Standards, there followed many profitable years for the profession. The results of this survey were published in 1925 and incorporated in Federal Specification No. 356 for Dental Amalgam Alloys.
Following surveys conducted by the National Bureau of Standards and the American Dental Association in 1928, 1929 and 1933 the American Dental Association Specification No. 1 was published in 1934. This specification, putting a premium on chemical composition, dimensional change, ultimate compressive strength and flow was the first reliable guide to members of the profession to help them in selecting an alloy from which a restoration could be placed and confidently expected to last for many years.

In Great Britain during the 1930's, Dr. K.L.V. Gayler carried out extensive research on problems associated with amalgam, at the request of the Dental Investigation Committee of the Dental Board. (Gayler 1933-37).


In 1959 International Standards for Amalgam Alloys were promulgated. This F.D.I. Standard was adopted by the American Dental Association as its standard in 1960.

The setting up of both the National Bureau and the Commonwealth Bureau of Dental Standards has resulted in the publication of many papers as a result of original research on amalgam.
Running parallel with the research conducted by Bureaus of Dental Standards a good deal of research has been conducted by individuals and Dental School Groups far too numerous to mention. Some particularly noteworthy contributions from this source have been the work of Ward and Scott (1932) on dimensional change, the Dental Materials group led by R.W. Phillips of Indiana who have made particular contributions to the knowledge of mercury content of amalgam restorations and the group led by Ryge at Marquette University, who have made outstanding contributions to the knowledge of the setting mechanism of amalgam.

The knowledge concerning amalgam has advanced very markedly during the last century. Research has thrown much light on the manipulation of amalgam and the properties expected in restorations made from this material.

The setting up of Standards and the adoption of Specifications have led to an improvement in amalgam alloys available and some measure of protection for the dentist and his patients, by aiding the dentist in his selection of a satisfactory alloy.
CHAPTER II. THE CHEMISTRY OF THE SETTING
OF DENTAL AMALGAM.

The process of amalgamation as applied to dentistry consists of
the mixing together of definite quantities of the amalgam alloy, in the
form of filings or shavings, and mercury. A plastic mass is formed which
is then condensed into the cavity where it hardens and becomes dental
amalgam.

This simple undertaking, very much taken for granted is in fact
based on a series of very complex reactions. Without a knowledge of the
changes taking place during the mixing and setting processes an under-
standing of the physical properties of amalgam is not possible.

THE AMALGAM ALLOY.

The modern amalgam alloy is the result of very much research both
by manufacturers and dentists. The need to satisfy certain standards has
cased manufacturers to modify techniques of manufacture in order to provide
a material from which restorations with certain desirable properties may be
made. While the composition of the alloy is important, different manufactu-
ners are able to produce alloys with the same composition but with completely
different properties. The degree of care adopted by the individual
manufacturers is the deciding factor here.

Modern dental amalgam alloys consist of silver, tin, copper and
zinc. While the American Dental Association and Federation Dentaire
Internationale Specifications lay down rigid compositions for amalgam alloys the Australian Specification is not so rigid and the composition of these alloys may be at the discretion of the manufacturer. In practice, however, in order to meet the other requirements of the Australian Specification alloys will have a similar basic composition. The limits imposed by the American and F.D.I. Specifications are:

- Silver: 65% minimum.
- Tin: 29% maximum.
- Copper: 6% maximum.
- Zinc: 2% maximum.
- Mercury: 3% maximum.

The Australian Specification makes suggestions for the benefit of the manufacturer of:

- Silver: 67.0 to 68.5%
- Tin: 26.0 to 27.0%
- Copper: 4.0 to 5.0%
- Zinc: 0.5 to 1.5%

The specifications (A.D.A., F.D.I. and Australian) are principally concerned with the physical properties of amalgam and to meet the requirements of dimensional change, strength and flow, alloys must have compositions close to those recommended.
Before dealing with the manufacture of alloys the contribution of the constituents will be considered.

Silver gives the alloy strength, decreases the flow or creep tendency but causes expansion during setting. Too high a silver content will produce an amalgam which amalgamates unwillingly, sets very rapidly, is very strong but which expands excessively.

Tin while it reduces strength, aids amalgamation, reduces expansion and so it helps to balance the expansion caused by the presence of the silver. Tin also increases the flow of amalgam.

In Europe alloys containing approximately 50% tin are very popular but these contract very markedly, exhibit a great deal of flow and have low compressive strength.

Copper in small quantities replaces some of the silver and imparts the same desirable properties as silver. Too high a percentage of copper would tend to cause a tendency for the amalgam to tarnish.

Zinc is included to aid the manufacturer during the manufacture of the alloy. Zinc acts as a scavenger for oxygen and prevents the oxidation of the other ingredients. While some alloys have been manufactured without zinc these have a tendency to be dirty and less satisfactory to handle, although in general some of them are able to satisfy the various specifications.

Mercury where present in small quantities is provided to facilitate
amalgamation, these alloys being known as pre amalgamated alloys or activated alloys and are popular in Europe for finger stall triturate.

Since dental amalgam consists of approximately 93% of silver and tin it may be regarded for practical purposes as a silver-tin alloy.

The equilibrium phase diagram for silver-tin is well established (Murphy 1926) and from this apparently complicated phase diagram the portion of dental interest is the small portion lying between 25% tin to 26.8% tin (see Fig. 1).
When an alloy with this approximate composition is cooled slowly, there is formed at temperatures below 480°C, an intermetallic compound known as the gamma phase (Ag$_3$Sn). Alloys with less than 25% tin contain some of the $\beta$ solid solution while alloys with more than 27% tin contain, in addition to the Ag$_3$Sn, some of the eutectic.

The presence of the $\beta$ solid solution would tend to increase the expansion of an amalgam produced while the eutectic would tend to lower the expansion or even cause a contraction.

Dental amalgam alloys may therefore be considered to be Ag$_3$Sn. Where small amounts of copper are present there is a tendency to form Cu$_3$Sn (Gayler 1937).

The responsibility of the manufacturer does not cease when he has selected his composition within the fairly narrow range which is satisfactory. Care during manufacture cannot be overstressed.

The manufacture of the alloy begins with the selection of pure constituents which are then melted under controlled conditions without oxidation, and cast into ingots which are then comminuted into filings.

Before beginning the comminution process the ingot is subjected to a heat treatment process to render it more homogeneous. In the as cast condition a considerable coring is evident in the ingot (Ryge 1953). Homogenization may be carried out by heating the ingot to approximately 400°C, for varying times. Work on this aspect of the manufacture of alloys
was carried out by Worner et al (1939) and Strader (1949). These workers showed that in general, amalgams produced from ingots which have been homogenized have better working qualities than where this process has not been carried out.

The comminution of the alloy is carried out by means of a lathe, using a suitable tool and feed rate. This results in the production of fine needles of brittle amalgam which may then be reduced in size by ball milling. Fractions of the desired size may then be obtained by screening and air and liquid separating methods.

The filings are then subjected to a process of aging or stress relieving in order to give the amalgam desirable predetermined properties and to render the alloy stable. As long ago as 1895, Black discovered the effect and phenomena of room temperature aging over a period of several months or very short time aging at 100°C. Amalgams made from particles which have been aged, expand less, are stronger and exhibit less flow.

Although it has never been completely explained, it is felt that aging is effective in relieving the stresses induced by strain hardening during the comminution of the ingot into filings.

Amalgam alloys which comply with previously mentioned Specifications are not, as can readily be seen, the result of good fortune but are the result of careful manufacture. Following the selection of satisfactory alloys by the dentist the responsibility for successful restorations
passes to him. Because of faulty manipulation he can produce unsatisfactory amalgams by improper use of a basically good alloy (Phillips 1957).

THE AMALGAMATION AND SETTING PROCESS.

The actual process taking place during the mixing or trituration of the amalgam alloy and mercury has posed a problem for many years.

In order to determine the exact chemical reaction taking place, basic scientists have attacked the problem from the angles of testing for physical properties, metallurgy, x-ray diffraction, micro-structure, crystallography and thermal analysis.

Despite a good deal of work by many people there is still some doubt as to the actual mechanism of the setting reaction. In a recent paper Ryge et al (1961) evaluated the present knowledge available.

There is general agreement that the main reactions during setting, take place between the silver, tin and mercury (Knight and Joyner 1915, Gray 1923, Gayler 1933-1937, Troiano 1938). It has also been established beyond doubt that Black's alloy was essentially the intermetallic compound Ag$_3$Sn (Knight and Joyner, Gray, Murphy 1926, Gayler 1933-1937).

Controversy presents when we begin to consider the reactions taking place when mercury is added to the Ag$_3$Sn.

The first equation of amalgamation was that proposed by
McBain and Joyner (1912).

\[ \text{Ag}_3\text{Sn} + 4 \text{Hg} \rightarrow \text{Ag}_2\text{Hg}_4 + \text{Sn}. \]

The next suggested equations for the setting reactions were those proposed by Gayler (1937). She based her theory of setting on work carried out on dimensional change during setting, thermal and chemical determinations, and metallurgical and x-ray diffraction studies. The results of this work were published between 1933 and 1936 in a series of six articles.

Gayler's equations are:

\[ \text{Ag}_3\text{Sn} + \text{Hg} \rightarrow \beta, + \gamma_2 \quad \cdots \quad (i) \]
\[ \beta, + \gamma_4 \rightarrow \beta, + \gamma, + \gamma_2 \quad \cdots \quad (ii) \]

\( \gamma, \) and \( \beta, \) are phases of the silver mercury system while \( \gamma_2 \) is a phase of the tin mercury system.

Gayler thought that silver rich alloys expanded due to the predominance of the gamma phase of the silver mercury system while tin rich alloys which had long been considered to contain \( \text{Ag}_3\text{Sn} \) and Sn cause contraction due to the formation of large quantities of the gamma phase of the tin-mercury system. The explanation of contraction being caused by the formation of the \( \gamma_2 \) phase is a contradiction particularly in view of her acceptance of a theory put forward by Gray (1923) that expansion is caused by crystal growth and contraction by solution.
Troiano (1938) proposed the following equation for the setting reaction.

\[ \text{Ag}_3\text{Sn} + \text{Hg} \rightarrow \gamma_1, + \delta_2 + \text{Ag}_3\text{Sn} \text{ (unattacked)} \]

\[ \gamma_1, + \delta_2 + \text{Ag}_3\text{Sn} \rightarrow \gamma_1, + \gamma_2 \]

\[ \rightarrow \gamma_1, + \gamma_2 + \beta_1 \]

the same end result as Gayler but proceeding in the opposite direction. Troiano defined \( \gamma_1 \) as \( \text{Ag}_3\text{Hg}_4 \).

Although at times various workers have affixed the formulas \( \text{Ag}_3\text{Hg}_4 \), \( \text{Ag}_5\text{Hg}_8 \), \( \text{Ag}_4\text{Hg}_5 \) for the \( \gamma_1 \) phase more recent work by Frankel and Fankuchen (1952) and Ryge, Moffett and Barkow (1953) using x-ray diffraction methods have shown this phase to be the naturally occurring Moschellandsbergite \( \text{Ag}_2\text{Hg}_3 \).

This was first described by Berman and Harcourt (1938).

As a result of work carried out by Moffett, Ryge and Barkow (1952), Ryge, Moffett and Barkow (1953) and Fairhurst and Ryge (1956) using a recording x-ray spectrometer this group of workers concluded that mercury reacts with \( \text{Ag}_3\text{Sn} \) to form a matrix consisting of dodecahedral \( \text{Ag}_2\text{Hg}_3 \) crystals and the hexagonal crystal gamma phase of the tin mercury system. Ryge and his co-workers grew single crystals on silver tin alloys in excess of mercury. The \( \gamma_1 \) and \( \gamma_2 \) phases were found to be present as early as ten minutes after the start of trituration. Winterhager (1955) demonstrated that the formation of the \( \gamma_2 \) phase precedes the formation of the \( \gamma_1 \) phase for
low silver alloys and the $\gamma_1$ phase is formed first in high silver alloys.

Schmitt (1960) was able to demonstrate the presence of three phases in the set amalgam by means of a selective etching technique using microscopic technique. His three phases were the original alloy ($\delta$), the gamma phase of the silver-mercury system ($\gamma_1$) and the gamma phase of the tin-mercury system ($\gamma_2$).

Although much light has now been thrown on the $\gamma_1$ and $\gamma_2$ phases, Frankel and Fankuchen have expressed doubt as to the existence of the $\delta$ phase and claim that it certainly does not exist in the early stages of setting as claimed by Gayler. They also express doubt as to the existence of unattacked original particles. Schmitt however, showed that the presence of original particles is dependent on manipulative variables.

In the light of previous work, the present knowledge of the mechanism of the setting of dental amalgam has been summed up by Ryge et al (1961) as follows:

\[
\text{Ag}_2\text{Sn} + \text{Sn} + \text{Hg} \rightarrow \text{Ag}_2\text{Hg}_3 + (\text{Sn},\text{Hg}) + \text{Ag}_2\text{Sn}
\]

The constituents other than silver and tin do not contribute to the setting reaction but modify the properties of the set mass. Frankel and Fankuchen did however, find small quantities of $\text{Cu}_2\text{Hg}_4$.

With this present knowledge of the setting reactions of amalgam a better understanding of the physical properties of amalgam is possible.
CHAPTER III. DIMENSIONAL CHANGE.

The expansion or contraction of amalgam which takes place during its setting is one of its most characteristic properties and one to which a great deal of importance is attached. Although the true clinical significance of dimensional change is not known, it seems reasonable that a slight expansion is preferable to a slight contraction. The expanding mass of amalgam will tend to deform the somewhat elastic tooth structure and so ensure intimate contact between the amalgam and tooth when changes of dimension of both tooth and restoration occur during thermal assaults on the oral tissues.

Although a slight expansion is undeniably desirable, surveys conducted with amalgams which were known to exhibit a final contraction, failed to produce any clinical evidence of this shrinkage (Phillips et al 1945, McDonald et al 1950).

These workers are, however, by no means indicating a change in the specifications to provide a contracting amalgam but are rather suggesting that a margin of safety exists with regard to amalgam unintentionally or unavoidably abused during manipulation.

Both the American Dental Association Specification for Amalgam Alloy which is adopted from the Federation Dentaire Internationale Specification and the Australian Specification provide for a slight expansion at 24 hours these being 0-20 microns/cm. and 2-15 microns/cm.
respectively while the Australian Specification sets limits for the initial contraction taking place. This initial contraction is not to exceed 4 microns/cm.

The measurement of such small changes must be made with an instrument capable of accurate measurement to at least 0.5 microns.

The instrument of first choice is undoubtedly the light interferometer (Sauer and Paffenberger 1942) which is used by the National Bureau of Standards, Washington, and the Commonwealth Bureau of Dental Standards, Australia, as well as many other institutions. The degree of accuracy afforded by this type of instrument is regarded as essential for specification work. Use has been made however, of other types of instruments and among these are optical micrometers (Ward and Scott 1932, Worner 1940, Gayler 1934) and accurate dial gauges (Worner 1937, Van Gunst and Hertog 1957, Rantenen 1960, Langeland 1961).

While the supremacy of the interferometer can not be denied both because of its accuracy and the ease of adjustment, much valuable work has been done using other types of instruments. Greater care must be taken and the limitations of these instruments realised.

Measurements are usually made on cylindrical specimens 10 millimeters in length for convenience of calculation of change per centimeter.

The classical picture of the dimensional change of amalgam during setting is one of an initial small contraction, followed by an expansion
with a gradual levelling off and perhaps very slight contraction and stabilization at 24 hours and thereafter (Skinner and Phillips 1960).

![Graph of dimensional change vs. time (hours)](image)

**Fig. 2.**

It is generally thought that the initial contraction is in most cases rapidly overcome by the expansion and the net result is a slight expansion.

Rantanen (1960) commented on the requirements of the specifications for measurements to commence 15 minutes after the commencement of trituration and showed that for three alloys, normally satisfying the requirements of both the American Dental Association and F.D.I. Specifications...
when measurements are begun five minutes after the commencement of trituration an exceedingly strong and swift contraction took place which was never completely eliminated by the subsequent expansion, and the net result was a contraction.

This finding which is in conflict with generally accepted theory may be due to a settling of the anvil of the mikrokator, although the measuring pressure was only 10 grammes. These measurements were made while the specimen was within the mould cavity.

THE THEORY OF DIMENSIONAL CHANGE.

The dimensional changes taking place during the setting of amalgam have been explained by the changes of state and the formation of new phases occurring during the hardening process.

In the light of the present knowledge of the mechanism of the setting of amalgam it seems that the initial contraction takes place during the solution stage of amalgamation as suggested by Gray (1923). The expansion is thought to be due to the formation of the $\gamma$ and $\gamma_2$ phases which growing dendritically cause an increase in volume of the amalgam (Troiano 1938). Secondary contraction is thought to be caused by the formation of the $\beta_1$ phase which having a greater specific gravity than the other phase causes a contraction (Troiano 1938).

If we are to deny the presence of the $\beta_1$ phase, an explanation of the small secondary contraction is difficult unless we consider that
further solution takes place at this stage. If this is the case - what is the reaction and the end product of this reaction?

Peyton (1960) offers the opinion that this slight shrinkage is not a predominant characteristic of most amalgam alloys that are currently available.

In summary, it may be said that anything causing or facilitating solution of the mercury causes contraction or at least decreased expansion, while the formation of the $\gamma_1$ and $\gamma_2$ phases causes expansion (Gray 1923).

While the solution of the mercury in the Ag$_2$Sn takes place mainly during trituruation, further solution is thought to take place during condensation and even after the completion of packing.

The degree of initial contraction will depend upon the degree of solution taking place, the degree and rapidity of formation of $\gamma_1$ and $\gamma_2$ crystals tending to nullify and negate this contraction and as suggested by Gray (1923), unreacted particles may, by touching, form a framework to prevent contraction.

THE FACTORS AFFECTING THE DIMENSIONAL CHANGE.

While it is recognised that composition and manufacture play an important part in determining dimensional change, modern alloys which are regarded as satisfactory, have compositions falling within fairly narrow ranges and in general have been manufactured carefully, to reach a standard
which is acceptable to the profession. As a result of this, there is a
good deal of similarity between many of the amalgam alloys marketed and
it is the effect of manipulative variables introduced by the dentist which
is responsible for variations of the physical properties of the amalgam.
The most important difference between alloys as supplied to the dental
profession is the particle size.

The dimensional change of an amalgam is affected at every stage
in the manipulation.

The factors affecting dimensional change which will be considered
are trituration, particle size, mercury:alloy ratio, condensation and
moisture contamination.

TRITURATION AND PARTICLE SIZE.

The effects of particle size and trituration on dimensional
change must be considered together since it is thought that these factors
operate in a direct relationship.

From purely theoretical consideration, a decrease in particle size
and an increase in trituration will increase the amount of solution and hence
decrease the degree of expansion of the amalgam. These theoretical
considerations have been shown to be correct by many workers.

Ward and Scott (1932) in their monumental work on dimensional
change showed that increased trituration resulted in an increased initial
contraction followed by a normal or reduced expansion and resulting in either a net contraction or a net reduced expansion. This has been verified by many workers and mention must be made of the work of Ware and Docking (1954) who combined the various factors of trituration in the concept of "trituration work", being a product of the load on the pestle and the number of revolutions during mixing. They showed that an increase in trituration work whether by increased load or an increased number of revolutions resulted in a decreased expansion. This is in keeping with the findings of other workers (Jarabak 1942, Phillips 1957, Jendresen and Ryge 1960).

Running parallel to the effects of trituration on dimensional change are the effects of altered particle size. Jarabak (1942) showed that decreasing particle size caused an increased initial contraction and decreased net expansion. Crowell and Phillips (1951) verified this work, putting much more stress on the influence of the specific surface area of the particles since it is at the surface that solution takes place. They placed more importance on the thickness of the particles than the original size of the particles. Jendresen and Ryge (1960) were able to verify the importance of thickness and they reported a decreased expansion at 24 hours with thinner particles.

It is generally agreed that increase in trituration results in a breaking up of alloy particles which in turn results in greater ease of solution of the mercury. If this is the case, it is fairly obvious why the thickness of the particles is the important factor since thin particles will tend to fracture more readily
than thicker particles. Although this will be the case where long slender particles are concerned, it has been theorised that it will not be so where the particles are small and approximately equi-axed since these will not readily break up. Claims have been made, by Jarabak (1942), Smith (1949), Mosteller (1953) as well as others, on the basis of the above theory for the lessened influence of trituration on dimensional change with fine grained alloys. The work of Jendreyson and Ryge (1960) did not support this claim. They showed a comparable increase in contraction for both fine and coarse grained alloys with the same degree of additional mixing.

While trituration is most frequently carried out using a mortar and pestle, in more recent times more use has been made of some form of mechanical amalgamator in research undertakings as well as clinical practice in order to more closely standardise this operation. The claimed advantages for this method of mixing amalgam are the uniformity of mixes, the short time required (usually only a few seconds), and the elimination of human variables.

The chief disadvantage of this type of amalgamator is the apparently inherent tendency of amalgams mixed mechanically to exhibit a contraction or at least very markedly reduced expansion as compared with amalgams mixed by hand and having comparable properties of strength and flow.

The reasons for this contraction have been postulated. Skinner (1947) and Romnes (1953) suggest that this may be due to overtrituration because of the proportionate importance of an inaccuracy of even a few
seconds. Taylor et al. (1949) suggested that there may be some advantage to longer contact between the alloy and mercury, as provided in mortar and pestle trituration.

That this tendency towards contraction exists is evident from the work of Phillips (1944) who found only two out of twelve alloys tested which produced an expansion when mixed in a high energy mechanical amalgamator. All of these showed an expansion when mixed by mortar and pestle. Other evidence for this tendency has been seen in the work of Skinner and Mizera (1958), Jendresen and Ryge (1960). By comparison, the production of a contracting amalgam following hand trituration results from definite attempts at overtrituration.

Another method of trituration, which attempts to minimise initial contraction and control expansion, is that advocated by Gayler (1934), viz: The Rubber Finger Stall Method. This method aims at preventing the breaking up of alloy particles and giving more uniform mixes. Little work has been done on this method of trituration but it would seem that since one of our aims is in fact to reduce the size of the alloy particles, at first sight this method would seem to be contra-indicated.

Before leaving the effects of trituration on dimensional change, mention must be made of an interesting observation made by Worner (1940) regarding the influence of trituration temperature on dimensional change. Worner observed that in general when trituration was carried out at elevated temperatures there was a decrease in the expansion at 24 hours and when
trituration was carried out at lowered temperatures there was an increase in the 24 hour expansion but more importantly the dimensional change continued to increase. It is now known that the continuing expansion was due to moisture condensation when the temperature of trituratation fell below the dew point of the room but an explanation for the variation in 24 hour dimensional change presents more of a problem. At the time of carrying out this work, the dangers of moisture contamination were not realised and standard technique was to mull the amalgam in the palm of the hand following mixing in a mortar and pestle. This contributed towards clouding the results obtained by many of the early workers.

**MERCURY : ALLOY RATIO.**

While it is now generally considered that it is the percentage of mercury in the final restoration which is important with regard to the physical properties of an amalgam, it is also realised that this is dependent on the initial amounts of mercury and alloy used. Phillips and Boyd (1947) found that for every additional 15% of mercury used initially an additional 1% is retained in the finished restoration despite all attempts to remove it.

The greater the amount of mercury mixed with the alloy initially the greater the amount retained in the restoration and the greater the expansion (Skinner and Phillips 1960, Ware and Docking 1954). Ware and Docking showed that over a wide range of mercury:alloy ratios (13:10 to 17:10) there was an increased expansion of less than 1 micron/cm, where there has been efficient mercury expression during manipulation. They also drew a
parallel between initial ratio and dimensional change and degree of residual mercury in the restoration.

The rationale for the increased expansion with an increased initial mercury:alloy ratio as given by Skinner and Phillips (1960) is that the more free mercury retained in the restoration the greater the amount of the $\gamma_1$ and $\gamma_2$ phases formed and hence the greater the expansion. Added to this is the fact that the additional mercury weakens the amalgam thereby allowing the additional crystallization to be effective in producing expansion.

Little data is available with regard to the effect of very low mercury:alloy ratios as suggested by Eames (1959). Skinner and Mizera (1960) in their evaluation of "The Eames Technique", found that the greatest expansion or least contraction occurred at a mercury:alloy ratio of 5:6 and the expansion did not increase at ratios above 1:1 as expected. They thought that this may be due to wedging of the particles.

CONSENSATION.

When all other factors are held constant the effect of increasing the condensation pressure is to decrease the expansion (Ward and Scott 1932). They found that even with abnormally high packing pressures (256,000 p.s.i.) although expansion was reduced, a contraction was not produced.

Gray (1923) considered that condensation was a continuation of trituration and with additional condensation it may be considered that the $\gamma_1$ and $\gamma_2$ phases formed are rubbed off the particles allowing further
solution to take place. Initial contraction may be hampered by the wedging of particles but the net expansion will be mainly governed by a smaller amount of \( \gamma_1 \) and \( \gamma_2 \) phases produced.

Ware and Docking (1954) basically agreed with the work of Ward and Scott and showed that a regular but now linear relationship exists between condensation and dimensional change and that the effect of a variation is greater for lower condensation loads than for higher loads.

The effects on the physical properties of using some form of mechanical condensation have been studied and findings vary with regard to dimensional change.

Ryge et al (1952) found that for both the pneumatic condenser and two types of vibrating type condensers the expansion was slightly reduced when compared with hand condensation techniques.

On the other hand Skinner and Mizera (1958) found comparable dimensional change for specimens packed using ultrasonic vibration (The Cavitron) and specimens packed by hand.

Other workers who have shown comparable or slightly reduced dimensional change using various forms of mechanical condensers are Swartz et al (1954) and Wilson et al (1957).

MOISTURE CONTAMINATION.

The effect of moisture contamination on amalgams prepared from
alloys containing zinc is now well known. When these amalgams are contaminated with moisture, during triturating or condensation, a delayed or secondary expansion takes place beginning 3 - 5 days after condensation and continuing for some months. This secondary expansion may reach values as high as 400 microns/cm. and occurs irrespective of the source of moisture, whether saliva, perspiration from palming and mulling the amalgam or water or sodium chloride introduced experimentally during manipulation of the amalgam (Schoonover et al 1942, Phillips et al 1954).

Although many theories had been put forward to attempt to explain the mechanism of the delayed expansion, none of these theories satisfactorily explained all cases of delayed expansion recorded.

Blodk (1896) thought that this expansion was due to the presence of zinc in the amalgam. Romnes and Skinner (1938) considered that the use of very fine alloys and a mercury rich layer of amalgam on the floor of the cavity was responsible. Worner (1940), as mentioned earlier considered that the most important factor was the triturating temperature. A preliminary report from The Research Commission of the American Dental Association on Palming Amalgam (1941) considered that the important factor was palming the amalgam.

Although the first real clue to the cause of delayed expansion was given by Crowell and Angell of the S.S. White Company in 1935 - 36, one of whom was introducing moisture into the amalgam on his hands, the full explanation was put forward by Schoonover and his co-workers.
Water, reacting with the zinc in the amalgam, in the presence of mercury, causes the formation of hydrogen, which after a few days builds up considerable pressure. This pressure is sufficient to cause the amalgam to flow or expand and continue to do so.

As well as measuring the excessive delayed expansion, Schoonover et al were able to collect the gas formed, measure its volume, and analyse it and calculate the pressure exerted by this gas. They also demonstrated the presence of blisters formed on the surface of an amalgam specimen. These workers demonstrated the need for zinc to be present in the amalgam and they were able to show that non zinc alloys did not exhibit delayed expansion. This had already been shown by Sweeney (1941).

Claims have been made by Liebig (1942) that if the zinc content of the amalgam is low enough it will not exhibit delayed expansion even when contaminated by moisture. Although this has not been substantiated by Phillips (1957), who stated that moisture can not be tolerated in any zinc containing amalgam, work by Van Gunst and Hertog (1957) showed that delayed expansion can be avoided provided the zinc content of the alloy is less than 0.01 per cent. These workers suggested that copper may influence this phenomenon and the higher the copper content, the less is the delayed expansion. This relationship has not, however, been verified.

The problem of delayed expansion has led to interest in the use of non zinc alloys. While these alloys are less pleasant to handle than
zinc containing alloys it has been generally accepted that when correctly manipulated the results obtained by these two types of alloys is comparable. Jendresen and Ryge (1960) found that for two specially manufactured alloys, one containing zinc the other zinc free the dimensional change for these alloys was comparable.
CHAPTER IV. STRENGTH.

Strength is an important property in amalgam restorations. One of the requirements of a satisfactory restorative material is that it is able to withstand the forces of mastication. In order to do this a certain degree of strength is necessary.

The measure of strength usually made is that of compressive strength and it is this property at which much of the research on amalgam has been directed. Although the importance of tensile strength has not been underestimated, this aspect of amalgam has received little attention. Having a close relationship with the property of strength in amalgam restorations is the mercury content of the restoration. This aspect and its relationship to other properties has received a good deal of attention, particularly in the last ten years.

At present neither the American Dental Association, F.D.I. nor Australian Specification provides for a measurement of the compressive strength of a fully set amalgam. The Australian Specification has a provision for testing the one hour compressive strength, this being used as a test for the setting rate.

At present the only measure of strength in the American (F.D.I.) Specification is the property of Plastic Deformation or Flow. It has been felt that alloys meeting the requirements for flow, possess sufficient strength.
In testing for compressive strength, small cylinders are used. Various sizes have been used the most usual being 4 mm. diameter and 8 mm. height, although cylinders 6 mm. diameter and 12 mm. height have been used. It appears to be important to maintain the relationship between the height and diameter of 2:1 and it was shown by Taylor et al (1949) that the smaller specimens give slightly higher values for compressive strength than the larger ones. These workers also demonstrated the importance of stating the rate of application of the load during crushing and the need for maintaining a constant and slow loading rate. The importance of this had been demonstrated much earlier by Ward (1924).

The most important factor concerning the compressive strength of amalgam is probably the rate at which the amalgam develops its strength. Taylor et al (1949) and Phillips (1949) studied the rate of development of compressive strength by amalgam. Their results proved to be similar and showed that the amalgam reaches approximately 80% of its maximum strength at approximately 8 - 10 hours after condensation while at the end of 24 hours in most cases the maximum strength has been attained. Some slower setting alloys do not reach their maximum strength for approximately 7 days. The development of high one hour strength is claimed to be desirable by many workers, but even the most rapidly setting amalgam may not set quickly enough to prevent damage if subjected to the forces of mastication early and patients will perhaps always require to be warned of the need to avoid biting on newly placed restorations for 6 - 8 hours.
Amalgam is weak in tension and its tensile strength may be only one sixth to one tenth of its compressive strength (Ward 1924, Taylor 1930). It appears that little can be done to produce higher tensile strengths in amalgam, although increasing the percentage of tin in the alloy has the effect of reducing the degree of brittleness and increasing the tensile strength. This has a tendency to decrease the compressive strength and increase the flow as well as to produce probable contractions (Souder and Paffenbarger 1942). In general it would appear to be better to maintain the desirable properties of low flow, slight expansion and high compressive strength and to prepare cavities, so designed as to minimise failures due to low tensile strength.

THE FACTORS AFFECTING COMPRESSIVE STRENGTH.

Compressive strength will be affected by all of the manipulative variables within the control of the dentist and these include the particle size of the alloy selected, the mercury:alloy ratio, the trituration and the condensation technique. Compressive strength is also affected by moisture contamination.

PARTICLE SIZE.

In general, where other factors are maintained constant, the compressive strength of fine grained alloys is greater than that of the coarser grained alloys (Crowell and Phillips 1951). The main claimed advantage, from a strength point of view, of the fine grained alloys is that they not only have a slightly higher 24 hour compressive strength, but there is a more rapid development of early strength, the rate of set of
these alloys being greater. It is to be realised that when speaking of fine grained alloys we mean alloys having small particle size as supplied by the manufacturer, or which readily break up into fine grains.

Although there has been fairly general agreement that fine cut alloys do exhibit greater compressive strength than coarser cut alloys (Taylor et al 1949, Mosteller 1953, Jendresen and Ryge 1960), Peyton (1960) warns of the need to display caution in accepting the belief that all fine cut alloys are superior to all coarser alloys.

**MERCURY:ALLOY RATIO.**

One of our aims during the manipulation of amalgam is to produce a restoration having as low a residual mercury content as possible. Much work has been done on the mercury content of amalgam restorations the principal workers being Crawford and Larson (1953, 1954) and Phillips and Swartz (1949, 1954, 1956, 1961 with Nadal).

Although it is the residual mercury content of the amalgam which is most important Phillips and Boyd (1947) showed that this is related to the initial ratio of mercury:alloy. Swartz and Phillips (1956) found that the compressive strength of fully set amalgam decreased very markedly where the amount of mercury in the restoration was greater than 55%. Where the mercury content is between 45 - 50% the effect on the physical properties seems to be at a minimum (Ryge et al 1952, Crawford and Larson 1954, Swartz and Phillips 1954).
Nadal et al (1961) investigated amalgams placed by techniques, standardised to provide residual mercury percentages of 48%, 58% and 62%. Although previous investigations have shown mercury contents as high as 70% in the marginal areas, Nadal had to adopt a technique of adding mercury to the mixed amalgam to produce percentages of 58 and 62%.

The effect of the initial mercury to alloy ratio varies from amalgam to amalgam, the amount required depending largely on manufacturing variables. Phillips and Boyd 1947 showed that an increase in the ratio of mercury to alloy resulted in a decrease in the 1 hour and 24 hour compressive strengths. As opposed to this Ware and Dooking (1955) showed that a decrease in the amount of mercury used initially resulted in a lowered 1 hour compressive strength which they attributed to an undertrituration due to the smaller amount of mercury available. They claimed that above a certain mercury:alloy ratio there was no further effect on the 1 hour compressive strength.

Ryge et al (1958) showed that there was an optimum ratio (approximately that recommended by the manufacturer) and above and below this the compressive strength was decreased.

In 1959, Eames introduced his technique of using a very low initial mercury:alloy ratio and claimed increased strength and rate of set of amalgam. Eames was working on a clinical basis but an evaluation of this technique has been made by Skinner and Mizera (1960) who verified the findings of Eames and investigated mercury:alloy ratios from 1:3 to 11:5.
They found that for most alloys tested, the compressive strength at 1 hour was at a maximum at 5 mercury to 6 alloy. There was little difference in the strengths for a particular alloy at 1 week, irrespective of the original ratio of mercury to alloy. These workers queried the importance of the final mercury content in relationship to the compressive strength.

Taylor (1930) stressed the importance of not interfering with the mercury:alloy ratio once trituration has commenced. He claimed that the additions of small amounts of mercury to keep the mass of amalgam plastic during and prior to condensation could produce an amalgam with a compressive strength as low as one tenth of the normally expected compressive strength.

Little work has been done on this aspect of amalgam, although this fact is well accepted.

TRITURATION.

The effect of varying the amount of trituration tends to vary from alloy to alloy. Ward and Scott (1932) found that 3 of the 5 alloys tested by him showed increased strength when overtriturated, while the other 2 alloys showed a decrease in their compressive strength.

More recent studies tend to point to an increase in compressive strength with increased trituration. Taylor et al (1949) showed a general
increase in the compressive strength at 24 hours with additional trituration. Ware and Docking (1955) showed that an increase in trituration work produced an increase in the 1 hour compressive strength but with higher levels of trituration work a limiting value was reached. Phillips (1957) considers that trituration is the most important variable in amalgam manipulation. He stresses the need to avoid undertrituration. Phillips claims and has shown that adequate trituration is one of the most important factors in ensuring maximal removal of mercury thereby producing optimal restorations.

Increased trituration will tend to produce higher compressive strengths because of the tendency for alloy particles to break up giving increased strength and also because of the greater ease of removal of mercury, as already pointed out.

While the advantages of mechanical amalgamators are obvious from the point of view of standardization, no increase in strength is apparent when the resultant amalgam is compared with a properly hand amalgamated amalgam (Phillips 1944).

CONDENSATION.

The importance of condensation for the development of adequate strength cannot be overstressed and yet a great deal of confusion exists with regard to the various factors involved in the condensation process.
The dual purposes of condensation are:-

(i) to condense the mercury coated alloy particles so that a considerable degree of propinquity exists between them,

(ii) to remove excess mercury.

Ward and Scott (1932) showed that an increase in condensation pressure resulted in an increase in compressive strength and a decrease in the mercury content of the restoration. As a result of this work an optimum figure of approximately 8,000 p.s.i. has been quoted as desirable for the packing of amalgam.

Ware and Docking (1955) showed a linear relationship between the condensation load and the one hour compressive strength.

Confusion arises with regard to condensation when a consideration is made of forces applied, size of condensers used as well as the method of condensation into the cavity. Various techniques have been suggested, from clinical practice and some of these have been evaluated.

A great deal of importance has been attached to the significance of pressure, as opposed to condensation load and a consideration of the pressures resulting from various forces on different sized packers has been made by many workers.

The effect of mechanical condensation on compressive strength has been studied by Phillips (1944) Ryge et al (1952) and their findings tend to
indicate that although there is a slight tendency towards increased compressive strength with mechanical condensation, the difference between hand packed and mechanically packed specimens is not marked. The findings of Skinner and Mizera (1958) using a "Cavitron" were in agreement with these earlier findings.

The need for speed during condensation has been stressed. Once the mix is made, amalgam must not be permitted to stand for too long before condensation (Miller 1947). Amalgam that is over 3 or 3½ minutes old should be discarded and a new mix made (Phillips 1957). Phillips showed that there was a marked decrease in the compressive strength of amalgam that had been allowed to stand beyond this time. It was thought by Phillips that the decrease in strength is due to the breaking up of the $\gamma_1$ and $\gamma_2$ phases which have formed during standing. In conjunction with this there is a retention of additional mercury in the restoration producing a decrease in strength.

CONTAMINATION.

Another factor influencing compressive strength is moisture contamination during the trituration or condensation of amalgam. Moisture contamination is thought to primarily influence the dimensional change but it has been shown by Schoonover et al (1942) and Phillips et al (1954) that moisture contaminated specimens, due to the bubbles of gas throughout, have a compressive strength approximately 25% less than a comparable,
non-contaminated amalgam. This loss in strength does not manifest itself for some months after condensation. Phillips and his co-
workers observed that this loss of strength does not occur with alloys which do not contain zinc.
CHAPTER V. FLOW.

Silver amalgam exhibits the property of flow. When an amalgam specimen is placed under a static load, well below its proportional limit it flows or creeps and a shortening takes place.

Skinner (1938) considered that clinically flow is important and manifests itself by the disappearance of contact points, the appearance of gingival overhangs and the fracturing of occlusal margins. These manifestations appear when the amalgam has been in use for some time. It has been claimed by many workers that flow is not a clinical problem and rarely if ever manifests itself. Mosteller (1953) makes the claim that since we have intermittent dynamic loads in the mouth and never static loads the clinical implications of flow are doubtful. Phillips et al (1943) showed that even when amalgam was manipulated in such a way as to produce a flow value as high as 10% it was difficult to clinically demonstrate flow. In addition to this, Healey and Phillips in 1949 in a clinical study of amalgam failures failed to find one failure attributable to flow.

While the clinical significance of flow is not clear, it has maintained an important place in the testing of amalgam and as pointed out earlier, flow is the only means of evaluating the strength properties of an amalgam, in the American Dental Association Specification for amalgam.

The first research on flow of amalgams was carried out by Black (1896) who compared the flow of pure tin, pure silver and amalgam. He found that the silver completely strain hardened while neither the tin nor the amalgam did so.
Amalgam did, however, partially strain harden while tin did not exhibit the property at all, at room temperature. Ward and Scott (1935) verified the fact that amalgam does partially strain harden when subjected to flow testing.

THE MEASUREMENT OF FLOW.

In both the American Dental Association (F.D.I.) Specification for amalgam alloys and the Australian Specification, flow is measured by applying a load of approximately 1500 pounds per square inch to a cylindrical specimen 4 mm. diameter and 8 mm. high. This load is to be applied 3 hours after the preparation of the specimen and be maintained constantly for 24 hours. During the test the temperature is to be maintained at 37°C. The flow test for the American Specification was previously conducted at room temperature using a load of 3550 pounds/sq. in.

Worner in 1937 showed the relationship between temperature and flow and the Australian Specification has always required testing at 37°C. Worner showed that at higher temperatures less load was required to produce a comparable degree of flow. For practical purposes the load of 1500 p.s.i. does give flow values comparable with the load of 3500 at the lower temperatures. Recently Jendresen and Ryge (1960) showed that the flow values obtained by this means are lower than those obtained by the old American Dental Association Specification.

In order to satisfy both sets of specifications the specimen is not to shorten by more than 4%.
The time of application of the load to the specimen, after the completion of packing is important. Worner (1937) showed the relationship between the flow and the time of application of the load after the formation of the specimen. When the load is applied to less mature specimens, the flow is greater, as would be expected. The additional flow may be due to the influence of an initial contraction, if started sufficiently early, or incompleteness of set and would more properly be called flow-set.

While the flow determination of the Specifications is the total flow for 24 hours, Skinner (1938) has introduced the concept of "constant flow" which is a value related to the flow when its graph becomes a straight line after several hours. The constant flow is probably the more important clinically as it may give an indication of the actual flow likely to take place when the patient begins to use the restoration. Because flow is adopted as the only measure of strength in the International Specification (American Dental Association) it is the total flow which is really more important.

THE EFFECTS OF MANIPULATIVE VARIABLES.

Ward and Scott (1932) showed that an increase in trituration resulted in an increase in flow for three alloys and a decrease for two others. It is of interest to note that the three alloys showing an increase in flow for overmixing, showed an increase in crushing strength at 24 hours, while the other two alloys showed a decrease in crushing strength with overtiruration. Skinner (1938) showed that trituration had little or no
effect on flow irrespective of the way in which the trituration was
effected. Ware and Docking (1955) also showed that flow was independent
of trituration work as well as being independent of the initial alloy
mercury ratio.

Both Skinner and Ware and Docking showed that an increase in
condensation pressure decreased the flow, but despite condensation pressures
of 41,000 p.s.i. used by Skinner the flow was never completely eliminated.

Jendresen and Ryge (1960) showed that particle size has an influence
on flow. Fine grained alloys have a tendency to exhibit less flow than
coarser alloys. These same workers showed that amalgams prepared from alloys
containing zinc tend to exhibit less flow than zinc free alloys under the
same conditions.

The findings of some workers with regard to flow are a little
surprising. The reason why the initial ratio of mercury and alloy and the
trituration do not affect the flow while condensation pressure does is
difficult to explain.

While there is opposition to the flow test on clinical grounds, in
summary the words of Skinner and Phillips (1960) may be applicable.

"Thus the clinical significance of this particular property remains
unknown. It would seem that, until proven otherwise, the main merit of the
flow test is that it provides a satisfactory and convenient way to evaluate
the general strength properties of the amalgam restoration".
CHAPTER VI. MICROSCOPIC STRUCTURE.

Several workers have observed the microscopic structure of set amalgam but little attempt has been made to correlate the microscopic structure with the physical properties. The most valuable contribution to the literature in this field of investigation of amalgam appears to be that of Schmitt (1960) whose work was concerned essentially with the low silver content alloys but who did investigate some higher silver content alloys as well.

Rosenhain (1926) showed pictures of the microscopic structure of hardened amalgam, but a good deal was open to conjecture as a result of this work and correct interpretation was difficult.

Gayler (1937) showed some typical microstructures. She commented on the difficulty of polishing specimens due to tarnishing and the problem of differential hardnnesses in the amalgam. She investigated a series of amalgams containing vastly different percentages of silver and tin.

Ryge, Moffett and Barkow (1952) investigated the original alloy (Ag<sub>3</sub>Sn) and followed the growth of crystals, of the \( \gamma_1 \) and \( \gamma_2 \) phases, microscopically in amalgams packed against glass but unetched.

In 1952, Ryge, Dickson, Smith and Schoonover produced photomicrographs of pieces of amalgam specimens of known properties. The pieces were obtained from compression test specimens after fracture. These specimens had previously been used as dimensional change specimens. The small pieces
of amalgam were embedded in acrylic resin, polished and etched. These workers were not able to draw any conclusions concerning any relationship between the microstructure and the physical properties of amalgam. The photomicrographs showed the presence of original alloy particles surrounded by a matrix of amalgam.

Smith, Ferguson and Schoonover (1953) investigated the microstructure of both original alloy and set amalgam prepared by various techniques. They showed the presence of a Widmanstätten structure in the original particles when etched by means of chromic acid and sulphuric acid. Smith and his co-workers investigated various etching techniques for set amalgam and showed that similar results could be obtained using an electrolytic etching technique or a 30% nitric acid etch. Their photomicrographs showed the presence of the original alloy particles in an amalgam matrix which they showed to be softer using a scratch hardness method. They concluded that the percentage of mercury in the final restoration affected the appearance of the structure appreciably but the amount of trituration did not.

In 1958 Schnuck demonstrated the presence of three phases by a technique of relief polishing. Until this time photomicrographs had shown amalgam to consist essentially of original particles and an amalgam matrix.

Schmitt (1960) investigated the influence of various manipulative variables (mercury:alloy ratio, methods of mixing, methods of condensation, plastic deformation, and temperature of storage) on the structure of amalgam,
Schmitt used a microtome to cut sections of his amalgam specimens and claimed that this method produced a less disturbed surface layer on the specimen. Ten methods of etching, including both chemical and electrolytic were investigated and finally 10\% hydrochloric acid followed by a 10\% solution of FeCl₃ to which is added some concentrated hydrochloric acid was used.

Schmitt described three phases, being the original particles, into which have grown a second phase, and a third phase consisting of a matrix of amalgam. On the basis of the relative degree of the preciousness of the phases and assuming that the $\gamma_2$ phase is the least precious, Schmitt concluded:

(i) the matrix is essentially $\gamma_2$ (tin-mercury) because it is most heavily attacked by the acid etchant.

(ii) the unattacked original particles are the $\gamma$ (Ag₃Sn) phase.

(iii) the phase growing into the original particles is the $\gamma_1$ (silver mercury) phase.

In Schmitt's photomicrographs no degree of resolution was obtained in the $\gamma_2$ phase. The form of this phase was not readily discernible.

Schmitt in his investigation of the effects of variables on the structure of amalgam come to the following conclusions.

(i) Where an insufficient amount of mercury is used, all tin is transformed into the $\gamma_2$ phase and as a result of this, little mercury is available for reaction with the original alloy so only small areas of $\gamma_1$ can be seen
in the photomicrograph. Where an excess of mercury is used it is not possible to distinguish the three phases.

(ii) In the 50% silver alloys no original particles remain when the amalgam is mixed in a mechanical amalgamator. Such is not the case for alloys containing higher percentages of silver.

(iii) The shape of the packing instruments has no influence on the structure of the amalgam, nor does deformation of the set amalgam by external forces.

(iv) When heated to 80°C, the set amalgam changes form. Temperatures of this order have no influence on amalgams in clinical practice.

Although Schmitt has made a very valuable contribution to the examination of the microstructure of amalgam, he did not attempt to correlate his findings with known properties of amalgam specimens prepared under controlled conditions.
- PART B. THE ORIGINAL INVESTIGATION -
CHAPTER I. THE SCOPE OF THE INVESTIGATION.

Basically the aim of this original investigation may be stated as "to observe the effects of manipulative variables on the physical properties and microstructure of dental amalgam".

Broadly speaking the manipulative variables to be investigated are:

1. The selection of alloys.
2. The mercury:alloy ratio.
3. Trituration.
4. Condensation.
5. Contamination.
6. Reinforcement.
7. Temperature.

The effect of these variables on compressive strength, dimensional change and flow are to be determined.

1. The selection of Alloys.

The investigation was carried out on a series of seven amalgam alloys, all readily available on the Australian market and all complying with the Australian Specification for amalgam alloys. Specification of dental materials is now so much a part of dentistry that I decided not to investigate any alloys not complying with these specifications. A wide
selection of alloys is available which do meet the requirements of the specifications.

The alloys selected for investigation were:

Alloy 1. G. & G. Quick Set *  Medium grained.
Alloy 2. G. & G. Superfine *  Fine grained.
Alloy 3. True Dentalloy **  Medium grained.
Alloy 7. Zinc-Free True Dentalloy **  Medium - coarse grained.

As can be readily seen these alloys represent a fine grain and medium grain alloy of each of two manufacturers and a zinc free alloy of one of these manufacturers. As well as these alloys a medium and a fine grained alloy by two different manufacturers have been selected. From this selection of alloys, a representative picture of the effects of manipulative variables on the properties of amalgam is likely to be obtained.

All seven alloys are representative of the high silver content alloys, containing percentages of silver in excess of 65%. Alloy 6 represents a preamalgamated alloy, while alloy 7 represents a zinc free alloy.

* Englehard Industries.  ** S.S. White Co.
No attempt was made to investigate methods of manufacture, heat treatments, method of cutting the particles and the care taken in manufacture since this is an investigation of the effects of the dentists' variables of manipulation.


The initial ratios of mercury to alloy ranging from 4:5 to 10:5 were investigated. In this aspect of the investigation, a determination of the importance of the removal of mercury after trituration and before condensation is to be considered as well as the influence on the properties of amalgam of the addition of mercury after the commencement of mixing.

3. Trituration.

Various methods of trituration are to be considered, including the mortar and pestle, mechanical and finger stall methods of trituration. Since the mortar and pestle method is by far the most popular method of trituration of amalgam, most consideration is to be given to this method and the variables within this method of mixing amalgam. Time and speed of trituration as well as the type and condition of mortars occupy a place here.

The use of mechanical amalgamators as well as the finger stall method of amalgamation are to be investigated.

Since the temperature of trituration may vary and is under the
control of the operator, and since this may have an effect on the physical properties of amalgam, an investigation of this aspect of trituration has been carried out.

4. Condensation.

Many variables are included in the investigation of condensation. The importance of force, the size of the condenser, the pressure, the number of pieces of amalgam, the number of thrusts on the condenser point, and the time of condensation have been investigated. Hand as well as mechanical condensation methods have been considered.

5. Moisture Contamination.

Although the effects of moisture contamination are now well known this variable is included for completeness and because little is known of the effect of this variable on the microstructure of amalgam.

6. Reinforcement.

The use of stainless steel wire to increase retention in large amalgam restorations is assuming more importance, following the work of Markley, so an investigation of the use of threaded stainless steel wire for retention and as a possible reinforcement for amalgam has been included in this survey.
7. Temperature.

Although we are not able to vary the temperature at which amalgam sets under clinical conditions, for which reason all specimens will be allowed to set and where possible be examined at mouth temperature, the effects of allowing amalgam to set at other temperatures has been investigated and is included for, at least, academic interest.
CHAPTER 2. EXPERIMENTAL PROCEDURE.

THE PREPARATION OF SPECIMENS.

Specimens for compressive strength, dimensional change, flow and microscopic examination were all prepared in the same mould cavity. The mould cavity used is similar to that used by Ware and Docking (1954) and produces cylindrical specimens with a diameter of 4 mm. These specimens may be 8 mm. or 10 mm. long depending on the choice of spacer made.

The die consists essentially of an accurately ground hole in a cylindrical piece of steel, and a base with a post which fits to a sliding fit inside the hole. (See Fig. 3).

--- Cavity 9 or 11 mm. by 4 mm.

--- 1 mm. spacer.

--- 8 or 10 mm. spacer.

Fig. 3.
The end of the post and the body of the die are ground level and at right angles to the hole. A mould cavity of the desired length is obtained by using a suitable spacer. An 8 mm. spacer is available to form compressive strength and flow specimens, while dimensional change specimens are 10 mm. long.

In preparing specimens an attempt is made to render the specimen more homogeneous by using an additional spacer (1 mm. high) to provide a cavity which is initially 9 mm. (or 11 mm.) long. The excess 1 mm. which corresponds to the mercury rich excess layer in a clinical restoration, may be removed by taking out the 1 mm. spacer, partially ejecting the specimen and removing the excess amalgam now protruding. This is most satisfactorily carried out using a razor blade which ensures that the top of the specimen is flush with the top of the ground surface of the die. The amalgam specimen may then be simply ejected from the mould by removing the remaining spacer and pushing the base to eject the specimen. The amalgam specimen may easily be ejected immediately after the completion of packing of the specimen or a few minutes later, when tests are due to begin.

The main advantages of this type of die for the preparation of specimens are:—

(i) Because of the fact that the cylindrical hole is in a substantial single piece of metal, greater accuracy can be expected than where we have a split mould with a join.
(ii) The main advantage comes from the fact that we are packing into a solid "four walled" cavity, with no problem of closing off the lower end, and can then easily eject our specimen by hand without the use of a punch. The fact that we are able to provide an excess of amalgam, which was packed under compression into the cavity, which can then easily be removed to provide a parallel ended specimen is an advantage.

Where the mould cavity is exactly the correct final length, the packing of excess beyond this does not give an assurance of the quality of the uppermost layer of amalgam in the specimen.

For compressive strength and flow determination, specimens 8 mm. long and 4 mm. diameter were used as provided in the Australian Specification. Dimensional change values were originally obtained on cylindrical specimens 10 mm. long and 4 mm. diameter as provided in the Australian Specification. Later, to ensure a greater degree of standardization in specimens, dimensional change measurements were made on specimens 8 mm. long and 4 mm. diameter and the dimensional change per cm. calculated. The specimens used for the study of the microstructure of amalgam were either specially prepared in the 8 mm. mould or were the dimensional change specimens after these had completely set.

In the preparation of the specimens, a standardized technique was developed to attempt to ensure uniformity and reproducibility of results. Before outlining the actual technique of packing the specimens, consideration will be given to some basic factors in the control of specimen production.
(a) Proportioning of Mercury and Alloy.

Mercury and alloy were proportioned using a conventional type of Ash Alloy balance. Although the greater accuracy of some volumetric dispensers is realized, this method of weighing the components of the amalgam was used because of its more universal application to all types of alloys. Following a previous survey (Iyell and Wing 1960), it is known that with this balance, proportions of mercury and alloy could be obtained to within 0.75% provided that these were both weighed separately using the same slide and provided that care was taken to place the alloy particles in the centre of the scale pan.

The amount of alloy required to provide sufficient amalgam to fill the mould under almost all conditions for all alloys was determined and used throughout this experimental work. 20 grains of alloy, mixed with the appropriate amount of mercury, provided sufficient amalgam with very few exceptions. These will be detailed later.

(b) Trituration.

Trituration was basically carried out using a raised centre mortar and pestle (S.S.White No. 8). To maintain a constant surface, this was conditioned by grinding with Fcarborundum and water after every tenth amalgam was mixed or more frequently if the mortar required it. Although mechanical amalgamation was considered in the survey and other degrees of hand trituration were investigated, the standard method of trituration was
one using a pestle load of $1\frac{1}{2} - 2$ pounds as measured on a spring balance and where trituration was continued till the amalgam could easily be shaken into a single ball in the mortar, after an homogeneous, shiny appearance of the amalgam in the mortar had been obtained. For each alloy investigated, the time required to achieve this was determined and standard mixes were then made at this time and pestle load. A visual check served to provide a high degree of uniformity of trituration and eliminate possible variations in the speed of rotation of the pestle.

Trituration was also carried out under varying conditions using an S.S. White Amalgamator, a Wig-l-bug Amalgamator and a rubber finger stell.

(c) Condensation.

Condensation was carried out by hand using a circular, smooth faced packer of 1.5 mm. diameter, except where the condenser was the variable factor. The force of condensation was controlled by placing the die into which the amalgam was being packed, on a spring balance. At low condensation loads (to 6 pounds) a constant force could be maintained to an accuracy within 0.5 pounds but at higher loads (10 - 15 pounds), accuracy could be maintained only to 1 pound. This degree of accuracy was sufficient to give consistently reproducible results.

Where the condensation was not the variable factor, specimens were prepared using a condensation load of 10 pounds.
(d) Standard Technique for the Preparation of Specimens.

Specimens were packed basically in accordance with the methods used at the Commonwealth Bureau of Dental Standards for specification work.

(i) Amalgam alloy and mercury are proportioned, using an alloy balance as outlined previously, and transferred to the mortar.

(ii) Trituration is carried out until the amalgam could be shaken into a single ball.

(iii) The full mass of amalgam is rolled into a rope which is cut into eight pieces.

(iv) Excess mercury is squeezed from each piece using a linen squeeze cloth and heavy finger pressure and the whole piece is introduced to the mould cavity. Where an 8 mm. specimen is being prepared, the mould cavity is 9 mm. long at this stage.

(v) Each piece of amalgam is condensed using the 1.5 mm. non serrated plunger. Twenty five thrusts of 10 pounds are used for each piece of amalgam moving the plunger over the whole surface of the specimen.

(vi) Excess mercury, and plushy amalgam brought to the surface are removed, using a le cron carver to ensure a reasonable degree of mercury removal before the introduction and condensation of the next piece of amalgam.
(vii) All 8 pieces of amalgam are treated likewise and the mould cavity completely filled. The amalgam has therefore been subjected to 200 thrusts in 8 layers to fill the cavity.

(viii) The 1 mm. spacer is removed immediately, the specimen mould moved up 1 mm. in the mould and the excess amalgam trimmed off level with the top of the die, using a sharp razor blade.

(ix) The specimen is ejected from the die immediately after the completion of packing and carefully stored ready for testing.

(x) The time from the beginning of trituration till the completion of packing the specimen is six minutes.

The procedure outlined in this section for the production of specimens has been used to produce what I have chosen to think of as a standard specimen. Where a particular variable is being considered all other factors are held at the level of the standard although at times the effect of cumulative variables has been considered. An easy comparison is then possible with the standard specimen.

The methods of altering the manipulative variables will be considered in the relevant sections.

THE TESTING OF PHYSICAL PROPERTIES.

(a) Compressive Strength.

Using cylindrical specimens 8 mm. high and 4 mm. diameter,
compressive strength was determined on a hydraulic compressive testing machine.

As pointed out by Taylor et al (1949) the speed of application of the load on the specimen is most important. The rate of application of the load was held constant at 300 pounds per minute. This represents a rate of approximately 15,000 pounds per square inch per minute. It was observed that where the specimen was loaded at a faster rate, the load at which fracture under compression occurred, was higher. It is of interest to note that where specimens had a length which was greater than twice the diameter, the compressive strength in pounds per square inch was less than for a specimen where the ratio of 2:1 was maintained. The compressive strength was calculated from the force at which the specimen crushed, by multiplying this force by 51.4, the reciprocal of the surface area. This gave the compressive strength in pounds per square inch.

(b) Flow.

The test for flow or plastic deformation was carried out as laid down for the Australian Specification for Dental Amalgam Alloys. The property measured was that of total flow, no attempt being made to determine the constant flow for the specimen.

After preparation, as outlined earlier, the 8 mm. specimen for flow is stored at 37°C. The flow test is begun three hours after the completion of packing of the specimen. Just before this time the specimen
is removed from the incubator and the ends are made smooth and at right angles to the long axis of the specimen by grinding on 2/0 emery paper using a "Vee Block". Before beginning the test the length of the specimen is measured using a micrometer. A load of 1,500 pounds per square inch is applied to the specimen, using a lever type flow apparatus (Ware and Docking 1955). The specimen remains under this load for 24 hours, at which time it is removed and the length measured. The difference in length, before and after the flow test, expressed as a percentage of the original length is the value for flow. It is essential that the flow apparatus and specimen be maintained at 37°C throughout this test, since flow is very dependent on temperature.

(c) Dimensional Change.

As no interferometer was available, dimensional change determinations were made using a Mercer Dial Gauge No. 110. This instrument is fitted with a ground, flat platen and a flat anvil, 3/16 inches in diameter, which is parallel to the platen. The instrument is capable of accurate measurement to 0.5 microns.

Measurements were originally made on specimens 10 mm. in length with a diameter of 4 mm., but as this size specimen offered no advantages over specimens 8 mm. long except the elimination of a calculation, specimens after the preliminary phase of the investigation were 8 mm. long. This offered the advantage of greater standardization of specimens for compressive
strength, flow and dimensional change.

Because of the nature of the measuring instrument used, a technique had to be devised for the measurement of dimensional change, which would not introduce variables not likely to be present in measurements made using an interferometer.

The method of placing the anvil of the gauge on the specimen, during setting proved to be unsatisfactory as the instrument exerted too great a force. The force exerted by the instrument is approximately 95 grams.

The technique which proved to be most successful was one where the weight of the spindle and the force exerted by the return spring were counterbalanced. Counterbalancing was carried out by variable weights attached to the lifting lever of the gauge or by using a knife edge pulley and a weight to directly lift the spindle. The gauge could be counterbalanced so that it exerted a load as low as 5 gm. Counterbalancing beyond this proved to be unsuccessful.

Immediately after the completion of condensation of the specimen it was ejected from the mould and placed on the platen under the anvil of the gauge. A firm positive pressure was applied to the spindle to assure intimate contact between the anvil and the specimen, and an initial reading was taken. Specimens were then allowed to remain on the gauge during the period of measurement of dimensional change. Readings were taken at 15 minute
intervals for the first \(1\frac{1}{2}\) hours, then at 30 minute intervals till 4 hours, after which hourly readings were made till 24 hours after preparation of the specimen. During the actual reading period, a load of 25 grams was momentarily applied to the spindle to ensure intimate contact between anvil and specimen.

Using the above outlined method of measuring, very constant results could be obtained for dimensional change.

Although the requirements of the Specification (A.D.A., F.D.I. and Australian) require readings to commence 15 minutes after the start of trituration, a truer picture of the dimensional change is given by beginning readings as soon as possible after the completion of the packing of the specimen.
CHAPTER 3. RESULTS OF INVESTIGATIONS
OF PHYSICAL PROPERTIES.

THE SELECTION OF ALLOYS.

The alloys investigated were all obtained on the open market, sufficient of each alloy being obtained to enable all tests to be conducted on the same batch of alloy. It is only to be expected that there may be a slight variation between different batches of the same alloy.

Using the mercury:alloy ratio as recommended by the manufacturer, and the standard methods of triturating and condensation as outlined in Chapter 2, a series of standard specimens was set up for the seven alloys being investigated.

These standard specimens were used to determine the one hour compressive strength, the rate of set of the amalgam as determined by a series of specimens crushed at varying times and the compressive strength of fully set specimens (7 days). On similar standard specimens the flow and a dimensional change curve were determined.

All values obtained were the mean of determinations on five specimens. Any result not falling within 5% of the mean was discarded.

Table 1 shows the values obtained for the standard specimens prepared for each of the seven alloys under investigation.
### TABLE 1.

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Mercury: Alloy Ratio</th>
<th>Mixing Time (Seconds)</th>
<th>Compressive Strength (p.s.i.) 1 hour</th>
<th>Compressive Strength (p.s.i.) 7 day</th>
<th>Flow</th>
<th>Dimensional Change at 24 hours. (\Delta L/\Delta W) *</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.*</td>
<td>7:5</td>
<td>40</td>
<td>15,400</td>
<td>55,000</td>
<td>0.8</td>
<td>+ 8</td>
</tr>
<tr>
<td>2.*</td>
<td>7:5</td>
<td>35</td>
<td>17,500</td>
<td>60,000</td>
<td>0.5</td>
<td>+ 8</td>
</tr>
<tr>
<td>3.</td>
<td>7:5</td>
<td>45</td>
<td>14,500</td>
<td>54,000</td>
<td>1.2</td>
<td>+ 11</td>
</tr>
<tr>
<td>4.*</td>
<td>7:5</td>
<td>40</td>
<td>16,500</td>
<td>58,000</td>
<td>1.2</td>
<td>+ 11</td>
</tr>
<tr>
<td>5.</td>
<td>8:5</td>
<td>45</td>
<td>14,200</td>
<td>52,000</td>
<td>1.2</td>
<td>+ 3</td>
</tr>
<tr>
<td>6.*</td>
<td>6:5</td>
<td>30</td>
<td>16,000</td>
<td>54,000</td>
<td>2.2</td>
<td>+ 10</td>
</tr>
<tr>
<td>7.</td>
<td>7:5</td>
<td>45</td>
<td>14,700</td>
<td>51,500</td>
<td>1.3</td>
<td>+ 9</td>
</tr>
</tbody>
</table>

* Fine grained alloys.

An analysis of the above table leads to some interesting observations.

Although it is generally accepted that fine grained alloys amalgamate very much more readily than their coarser counterparts, corresponding alloys (fine and medium) made by the same manufacturer require the same proportions of mercury and alloy and require approximately the same amount of triturating to produce a standard mix. The time to triturate the pre-amalgamated alloy (No. 6) was less than for all other alloys both fine and medium grained.

The strength at one hour tended to be higher for the fine grained alloys than for the coarser alloys and this slight increase in
strength was maintained for fully set amalgams. An exception to this was alloy No. 6 which showed a comparatively lowered strength at 7 days although it maintained its early rate of development of strength for some time (see Fig. 4) before the flattening of its setting rate curve occurred.

Every alloy tested had a setting rate such that beyond 24 hours the increase in compressive strength was quite insignificant. The setting rate is affected by the inherent properties of the alloy but also by the technique of manipulation.

The values obtained for flow are interesting when considered in conjunction with the strengths at 1 hour and at 24 hours. There appears to be no relationship between the values of flow and compressive strength. Alloy No. 1 which has a lower strength at both 1 hour and 24 hours than alloy No. 4, has a significantly lower flow than alloy 4. Much more marked however is alloy 6 which has strength comparable with all other alloys, but has a very much higher flow than any other alloy under investigation. A study of the setting rate curve for these alloys (Fig. 4) throws no light on this problem as at 3 hours when the flow test is begun the compressive strengths of all alloys bears a relationship to the 1 hour strength rather than the flow.

The dimensional change values obtained over a 24 hour period were interesting if a little anomalous. The lowest expansion of all amalgams was recorded for alloy 5 which was the coarsest of all alloys. The highest
figure for expansion was recorded for alloys 4 and 3 which are products of the same manufacturer and one of which is fine grained. Alloys 1 and 2 gave slightly lower but comparable dimensional change values while alloy 6 (fine) gave a comparatively high value. These findings are in keeping with the findings of Ware (1960).

The other interesting feature of the dimensional change determinations is the fact that for the method of preparation of the standard specimens, only one alloy (No. 5) exhibited an initial contraction and a subsequent secondary contraction (see Fig. 4).

The rate of development of the maximum degree of expansion for amalgams prepared from all alloys by this technique was quite rapid and occurred between 3 and 4 hours. To some extent this rate of dimensional change parallels the rate of development of compressive strength (Fig. 4).

The behaviour of alloy 7 (the zinc free alloy) as far as the physical properties are concerned is similar to other alloys. The handling characteristics for this alloy are however different. The alloy is slightly coarser than its counterpart and has a definite tendency to soil while being manipulated. The mortar and pestle and linen squeeze cloths used were blackened. The amalgam obtained after trituration had a tendency to be granular (this granularity was never really completely eliminated) and the amalgam packed apparently very poorly. It was a surprise to find that the compressive strength values obtained with this alloy were comparable
with the zinc containing alloys.

Of the alloys investigated, alloy 6 gave the impression of being most easily condensed while as a general rule the fine grained alloys resulted in amalgams which appeared to be smoother and more easily packed than the coarser grained alloys.

**MERCURY:ALLOY RATIO.**

In studying the effect of the initial amounts of mercury and alloy used to prepare an amalgam restoration, all other factors were held constant except the mercury:alloy ratio, and specimens were prepared in the standard manner.

Trituration was carried out using a mortar and pestle and although for low mercury:alloy ratios this produced a mix which was apparently undertriturated and for high mercury:alloy ratios, one which was overtriturated the trituration time was held constant at the value determined in the preparation of the standard specimens. The effect of cumulative variables will be considered later.

Initial ratios of mercury:alloy ranging from 4:5 to 10:5 were investigated. Less mercury than that giving a ratio of 4:5 was not considered since the mixed amalgam bears no resemblance to normal amalgam unless the trituration time is very markedly increased.
The effect of varying the initial mercury:alloy ratio on the 1 hour compressive strength for the various alloys is shown in Table 2.

**TABLE 2.**

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Mercury:Alloy</th>
<th>4:5</th>
<th>5:5</th>
<th>6:5</th>
<th>7:5</th>
<th>8:5</th>
<th>9:5</th>
<th>10:5</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>18,000</td>
<td>18,000</td>
<td>16,000</td>
<td>15,400</td>
<td>14,000</td>
<td>12,000</td>
<td>9,000</td>
<td></td>
</tr>
<tr>
<td>2.</td>
<td>22,000</td>
<td>21,500</td>
<td>19,000</td>
<td>17,500</td>
<td>15,000</td>
<td>12,500</td>
<td>10,500</td>
<td></td>
</tr>
<tr>
<td>3.</td>
<td>15,000</td>
<td>16,000</td>
<td>15,400</td>
<td>14,500</td>
<td>14,000</td>
<td>12,500</td>
<td>9,500</td>
<td></td>
</tr>
<tr>
<td>4.</td>
<td>20,500</td>
<td>19,500</td>
<td>17,000</td>
<td>16,500</td>
<td>15,800</td>
<td>14,000</td>
<td>11,300</td>
<td></td>
</tr>
<tr>
<td>5.</td>
<td>15,400</td>
<td>15,500</td>
<td>15,200</td>
<td>14,500</td>
<td>14,200</td>
<td>13,000</td>
<td>9,400</td>
<td></td>
</tr>
<tr>
<td>6.</td>
<td>15,500</td>
<td>16,000</td>
<td>16,000</td>
<td>15,500</td>
<td>12,800</td>
<td>11,000</td>
<td>6,000</td>
<td></td>
</tr>
<tr>
<td>7.</td>
<td>11,000</td>
<td>16,000</td>
<td>15,000</td>
<td>14,700</td>
<td>13,500</td>
<td>13,000</td>
<td>9,000</td>
<td></td>
</tr>
</tbody>
</table>

* Standard Mix.

In general the effect of decreasing the original mercury:alloy ratio has the effect of increasing the 1 hour compressive strength. While the fine grained alloys in general respond more satisfactorily to a decrease in the amount of mercury available during trituration, alloy 6 appears to have its optimum one hour compressive strength at approximately the ratio recommended by the manufacturer.
The effect on the 7 day compressive strength is less marked. It appears that there is some levelling off of the influence of the original mercury:alloy ratio as the amalgam sets. While the 1 hour compressive strength may be increased by approximately 20 - 25% by reducing the ratio from 7:5 to 5:5, the 24 hour compressive strength is increased by only 5 - 8% for the same change in ratio.

Where the amount of mercury available is increased above the recommended level there is in all cases a reduction in the 1 hour and 7 day compressive strengths.

The effect of altering the original ratio of mercury and alloy, on the setting rate is seen in Fig. 5.

The effect on the dimensional change was investigated for mercury:alloy ratios of 5:5, 7:5, 9:5.

Where the low ratio of 5:5 was used it was found that for all alloys there was a reduction in the amount of expansion at 24 hours. In the case of alloy 5 an actual contraction of 3 microns/cm. was observed.

The shape of the dimensional change curve is altered (see Fig. 5) for all alloys although an initial contraction was not observed with alloys 1, 2, 3, 4, 6 or 7. There was however a general flattening of the curve and except in the case of alloy 6, a typical secondary contraction took place.
Fig. 5.

Compressive Strength (N/mm²)

Time (Hrs.)

Time (Hrs.)

Alloy 1, Replasticised

Dimensional Change

Alloy 1 9:5
Alloy 1 7:5
Alloy 1 5:5
Alloy 5 5:5

4 8 12 16 20 24

4 8 12 16 20 24

6 min.
15 min.
Where the initial ratio was 9:5, the 24 hour expansion was increased for all alloys. Alloy 5 still exhibited a slight initial contraction but this was rapidly overcome. No secondary contraction took place.

Table 3 shows the influence of mercury:alloy ratio on the dimensional change of all alloys.

**TABLE 3.**

<table>
<thead>
<tr>
<th>Alloy</th>
<th>24 hour Dimensional Change</th>
<th>Mercury:Alloy</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>5:5</td>
<td>7:5</td>
</tr>
<tr>
<td>1.</td>
<td>+ 2</td>
<td>+ 8</td>
<td>+ 10</td>
</tr>
<tr>
<td>2.</td>
<td>+ 2</td>
<td>+ 8</td>
<td>+ 11</td>
</tr>
<tr>
<td>3.</td>
<td>+ 5</td>
<td>+ 11</td>
<td>+ 13</td>
</tr>
<tr>
<td>4.</td>
<td>+ 7</td>
<td>+ 11</td>
<td>+ 13</td>
</tr>
<tr>
<td>5.</td>
<td>- 3</td>
<td>+ 2</td>
<td>+ 5</td>
</tr>
<tr>
<td>6.</td>
<td>+ 9</td>
<td>+ 11</td>
<td>+ 18</td>
</tr>
<tr>
<td>7.</td>
<td>+ 5</td>
<td>+ 10</td>
<td>+ 14</td>
</tr>
</tbody>
</table>

The effect of an alteration of the initial ratio of mercury to alloy, was to vary directly the flow of specimens produced from all alloys.

All alloys showed only a slight decrease in flow when the initial
ratio was reduced from 7:5 to 5:5. Where the ratio of mercury to alloy was increased to 9:5 there was a slight increase in the flow for all alloys except alloy No. 6 which showed a marked increase in the flow.

Table 4 shows the influence of a change in mercury:alloy ratio on flow.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>0.7</td>
<td>0.8</td>
<td>1.0</td>
</tr>
<tr>
<td>2.</td>
<td>0.5</td>
<td>0.5</td>
<td>0.6</td>
</tr>
<tr>
<td>3.</td>
<td>0.9</td>
<td>1.2</td>
<td>1.5</td>
</tr>
<tr>
<td>4.</td>
<td>1.0</td>
<td>1.2</td>
<td>1.4</td>
</tr>
<tr>
<td>5.</td>
<td>0.9</td>
<td>1.1</td>
<td>1.3</td>
</tr>
<tr>
<td>6.</td>
<td>2.2</td>
<td>2.5</td>
<td>3.2</td>
</tr>
<tr>
<td>7.</td>
<td>1.0</td>
<td>1.3</td>
<td>1.5</td>
</tr>
</tbody>
</table>

As well as the initial ratio of mercury:alloy, consideration must be given to the influence of mercury removal, after trituration and before condensation. This factor was studied for alloys 1, 2, 3, 5 and 6.

Amalgam mixes were made using the standard initial ratio and
trituration time and specimens were packed following the normal method of removing excess mercury by hand from each of eight increments, immediately before introducing the increment to the cavity. During the packing of a typical specimen, each increment was weighed before removal of excess mercury and the "mercury" removed from each increment was weighed. A fresh squeeze cloth was used for each increment.

The mean weight of the increments obtained on a chemical balance was found to be 394 milligrams and the mean amount of material expressed through the squeeze cloth was 58 mg. Since this was for an alloy using a 7:5 mercury to alloy ratio the percentage of mercury in the mix before packing was approximately 49.5% although it is realised that some of the constituents of the alloy would have been expressed through the squeeze cloth. The ability to consistently squeeze mercury by hand to approximately 50% was confirmed for all alloys tested. Using pliers to remove mercury resulted in an approximate percentage of mercury of 48.5%. The aid of a dental nurse was enlisted, and it was found that she was able to produce increments of amalgam containing approximately 52% mercury.

It was found that more mercury could be removed from the increments used early than those used later.

When mercury was expressed from the whole mix of amalgam at once the results obtained were 52%, 50% and 56% mercury in the mass of mixed amalgam.
A series of amalgams was set up and their properties examined which were prepared by the standard procedure but with approximate amounts of mercury at the time of introduction into the cavity of 58.5% (original amount), 52%, 50%, 48.5%.

The results obtained for the alloys tested were parallel. Where no mercury was removed before condensation there was a significant decrease in the compressive strength at 1 hour for all alloys. This decrease varied from 950 p.s.i. for alloy 2 to 1,300 p.s.i. for alloy 6. The difference in the 24 hour strengths was negligible.

Where mercury had been removed to either 52%, 50% or 48.5% there was no difference in the strength at 1 hour or 24 hours.

The dimensional change at 24 hours was examined and there was no recordable difference for any of the techniques employed. Likewise flow was not affected by the variation in mercury content before packing.

This investigation was repeated using initial mercury:alloy ratios of 5:5 and 9:5 and it was found that where the ratio of 5:5 was used there was no variation in any of the properties tested irrespective of the methods used to remove mercury prior to packing, although it was found to be possible to remove some mercury by hand or pliers although the dental nurse was not able to do so.

When an initial ratio of 9:5 was used it was found that the removal of mercury before packing assumed a much greater importance.
Where no mercury was expressed prior to packing a considerable reduction in the 1 hour compressive strength resulted.

Alloy 1, Alloy 2, Alloy 3, Alloy 5, Alloy 6,
2,000 p.s.i. 2,500 p.s.i. 1,800 p.s.i. 1,800 p.s.i. 2,800 p.s.i.

These figures are reductions below the values in Table 2 for a ratio of 9:5.

Dimensional change values showed a further increase above the figures in Table 3 while flow values increased slightly above those in Table 4.

A notable feature of specimens packed using an initial ratio of 9:5 without mercury removal, was the inadequacy of the normal amount of amalgam to fill the mould cavity. Very much more plushy amalgam was able to be removed during packing.

While it may be considered to be unimportant to efficiently remove mercury prior to condensation, except where a high initial mercury to alloy ratio is used it is to be realised that a comparatively high condensation load was used in packing these specimens which would tend to efficiently remove mercury during condensation. Consideration of the combination of excess mercury and low condensation loads will be made in the section on condensation.

The effect of the addition of mercury to the partially mixed amalgam or mixed amalgam to render it more plastic was next investigated.
A mix of amalgam 28:20 was made in the standard manner for alloy No. 1 and this was allowed to stand undisturbed. After 5 minutes sufficient mercury was added to the amalgam to restore normal plasticity, as partial set had taken place. A compressive strength specimen packed in the standard manner and crushed at one hour produced an identical result as for a standard specimen. This result could be duplicated and a similar result obtained for all other alloys. There was likewise no change in the compressive strength of fully set specimens.

The investigation was continued and measured amounts of mercury added to the mixed amalgam. Sufficient mercury was added to a 7:5 mix to increase the mercury to a ratio of 10:5 and keep the amalgam mass plastic for 15 minutes. The additional mercury was added at 5, 10, 15 minutes without mulling. Specimen preparation was then carried out in a standard manner, removing excess mercury and condensing normally. Results for a series of such specimens showed no change in the compressive strength. The effect of adding mercury to the partially set amalgam did however produce a very marked effect on the dimensional change of amalgams from all alloys.

All alloys produced an initial contraction followed by an expansion (see Fig. 5) but this expansion was followed by a secondary contraction which continued for a very much longer time than usual and resulted in a contraction for alloys 5 and 6 and a very low expansion for the other alloys, when only a small amount of mercury was added and the amalgam allowed to stand for only 5 minutes. Where the amalgam was kept
plastic for 15 minutes as outlined previously, all amalgams resulted in a contraction.

Table 5 shows the dimensional change values for replasticized amalgam.

The flow values were not affected by this procedure.

**TABLE 5.**

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Plastic to 5 min.</th>
<th>Plastic to 15 min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>+1</td>
<td>-2</td>
</tr>
<tr>
<td>2.</td>
<td>+2</td>
<td>-2</td>
</tr>
<tr>
<td>3.</td>
<td>+2</td>
<td>-2</td>
</tr>
<tr>
<td>4.</td>
<td>+3</td>
<td>-1</td>
</tr>
<tr>
<td>5.</td>
<td>-5</td>
<td>-15</td>
</tr>
<tr>
<td>6.</td>
<td>-1</td>
<td>-4</td>
</tr>
<tr>
<td>7.</td>
<td>+2</td>
<td>-4</td>
</tr>
</tbody>
</table>

**TRITURATION.**

The standard method of trituration adopted was the use of a very light pestle load (1\(\frac{1}{2}\) pounds) with a high speed of rotation of the pestle (approximately 200 r.p.m.). In a well conditioned mortar this produced
a standard mix in a relatively short time (see Table 1). It should be noted here that in a new mortar (S.S. White No. 8) the time taken to produce the same stage of trituration under the same conditions was approximately 1 minute and not until the mortar and pestle had been conditioned approximately twenty times was this reduced time achieved which then remained standard. In a round bottomed mortar (S.S. White No. 7) the time taken to produce the same stage was approximately 70 seconds in a "well" conditioned mortar and 95 seconds in a non conditioned mortar.

It is interesting to note that in the time of approximately 40 seconds standard trituration, the first 20 seconds is taken up by almost dry grinding after which the alloy begins to take on its wetted appearance. This initial period is the period, during which, the oxide film formed on the alloy particles is broken down. This is necessary before the mercury can go into solution in the alloy. In the case of alloy 6 (preamalgamated) "wetting" of the surface commences within the first 5 seconds of mixing.

The effects of varying mortar and pestle trituration, on the physical properties of amalgams are shown in Figure 6.

Trituration times were increased and periods ranging from 90 seconds to 8 minutes were investigated. A reduction in trituration time to 20 seconds was made and investigations carried out.

At 20 seconds the mix was obviously undertriturated, being still somewhat granular and when an attempt was made to shake it into a ball, the
result was a large number of small pellets. At 90 seconds the mix was quite obviously overtriturated, having taken on a very thin appearance, being difficult to keep under the pestle and attempts to shake the mix into a single ball were complete failures. At this and longer trituration times the mix had to be mechanically removed from the walls of the mortar. The mortar required conditioning after every mix where overtrituration was used. Where trituration times as long as 8 minutes were used no difficulty was experienced in condensing the amalgam in the normal manner.

The effect on compressive strength, at 1 hour, of varying the trituration time is shown in Table 6.

**TABLE 6.**

<table>
<thead>
<tr>
<th>Alloy</th>
<th>20</th>
<th>40</th>
<th>90</th>
<th>180</th>
<th>300</th>
<th>480</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>12,200</td>
<td>15,400</td>
<td>19,000</td>
<td>21,500</td>
<td>21,500</td>
<td>19,000</td>
</tr>
<tr>
<td>2.</td>
<td>14,000</td>
<td>17,500</td>
<td>19,500</td>
<td>21,800</td>
<td>21,500</td>
<td>20,500</td>
</tr>
<tr>
<td>3.</td>
<td>11,500</td>
<td>14,400</td>
<td>18,000</td>
<td>20,500</td>
<td>21,000</td>
<td>18,500</td>
</tr>
<tr>
<td>4.</td>
<td>14,000</td>
<td>16,500</td>
<td>18,500</td>
<td>21,000</td>
<td>21,000</td>
<td>18,000</td>
</tr>
<tr>
<td>5.</td>
<td>10,500</td>
<td>14,200</td>
<td>17,500</td>
<td>20,500</td>
<td>20,200</td>
<td>19,000</td>
</tr>
<tr>
<td>6.</td>
<td>15,000</td>
<td>17,500</td>
<td>20,500</td>
<td>21,000</td>
<td>20,500</td>
<td>20,000</td>
</tr>
<tr>
<td>7.</td>
<td>13,500</td>
<td>14,500</td>
<td>18,000</td>
<td>20,000</td>
<td>20,500</td>
<td>18,400</td>
</tr>
</tbody>
</table>
It is obvious that undertrituration considerably lowers the 1 hour compressive strength of the amalgam while overtrituration increases the strength up to a limit and beyond this the compressive strength begins to fall again. Alloy 6 was least affected by undertrituration. The setting rate for all alloys is also considerably increased, the maximum compressive strength being reached very much earlier where the alloy is overtriturated. The compressive strength values for fully set amalgams is only slightly affected by extra trituration (see Fig. 6).

The effect of trituration on the dimensional change is much more marked (see Fig. 6 and Table 7).

It may be seen that undertrituration causes a slight increase in the dimensional change for all alloys, while overtrituration, produced a decrease for all alloys. All alloys exhibited a contraction for trituration times of 90 seconds. This was slight for all alloys except alloy 5 where it was considerable. With the trituration time increased beyond 90 seconds all alloys exhibited considerable contractions. For increased trituration time all alloys exhibited an initial contraction as well as a secondary contraction.

It is to be noted that both fine grained and medium grained alloys have a reduced expansion with overtrituration. The suggestion that fine grained alloys are not reduced in size during trituration cannot be given credence as the explanation for the theory that overtrituration has little effect on the dimensional change of these alloys.
Alloy 1.

Alloy 2.

Fig. 7.
### Table 7.

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Trituration (seconds)</th>
<th>20</th>
<th>40</th>
<th>90</th>
<th>180</th>
<th>300</th>
<th>480</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- 1</td>
<td>- 3</td>
<td>- 4</td>
</tr>
<tr>
<td>2.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- 1</td>
<td>- 4</td>
<td>- 4</td>
</tr>
<tr>
<td>3.</td>
<td></td>
<td>13</td>
<td>11</td>
<td>0</td>
<td>- 3</td>
<td>- 5</td>
<td>- 6</td>
</tr>
<tr>
<td>4.</td>
<td></td>
<td>12</td>
<td>11</td>
<td>- 1</td>
<td>- 3</td>
<td>- 4</td>
<td>- 7</td>
</tr>
<tr>
<td>5.</td>
<td></td>
<td>5</td>
<td>4</td>
<td>- 5</td>
<td>- 10</td>
<td>- 13</td>
<td>- 19</td>
</tr>
<tr>
<td>6.</td>
<td></td>
<td>9</td>
<td>9</td>
<td>0</td>
<td>- 3</td>
<td>- 5</td>
<td>- 7</td>
</tr>
<tr>
<td>7.</td>
<td></td>
<td>12</td>
<td>10</td>
<td>- 1</td>
<td>- 4</td>
<td>- 5</td>
<td>- 6</td>
</tr>
</tbody>
</table>

Fig. 7 shows alloys No. 1 and 2 before and after grinding in a mortar for 25 seconds. Both alloys have reduced in size.

The effect of an alteration in trituration time on flow was not very marked.

Alloys 1, 2, 3, 4 and 7 showed no alteration in flow with increased trituration while alloys 5 and 6 showed a very slight decrease in the flow with an increase in trituration. This decrease in flow however was so slight as to almost be disregarded.

In view of the very marked effect, on the compressive strength
and setting rate of amalgam, of trituration it is surprising that this variable has a negligible effect on the flow.

An investigation was carried out on alloys No. 1 and No. 5 to determine the effects of triturating to the same stage by different hand methods. This is in line with the concept of "trituration work" as proposed by Ware and Docking. It was found that where trituration work was achieved by increasing the pestle load and decreasing the number of revolutions there was no alteration in the compressive strengths of specimens for the same degree of trituration. There was however a slight decrease in the 24-hour dimensional change for alloy 1 and a slightly greater decrease for alloy 5 which is obviously very susceptible to manipulative variables from the dimensional change point of view.

The use of heavy loads for trituration tend to be unsatisfactory because of a tendency to grind the amalgam into the mortar, even when it is undertriturated.

The use of a poorly conditioned, round bottomed mortar where the time to achieve the same degree of trituration was greater, resulted likewise in a reduced dimensional change for the same compressive strength.

It appears that the additional time taken in dry grinding influences the dimensional change.