# NATIONAL BUREAU OF STANDARDS REPORT

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# Report on Dental Research
# at the
# National Bureau of Standards

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# Report on Dental Research
# at the
# National Bureau of Standards

## Progress Report
## January 1 to June 30, 1964

The dental research program at the National Bureau of Standards is carried on in cooperation with the Council on Dental Research of the American Dental Association; the Army Dental Corps; the Dental Sciences Division of the School of Aerospace Medicine, USAF; and the Veterans Administration.

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U.S. DEPARTMENT OF COMMERCE

NATIONAL BUREAU OF STANDARDS

# REPORT ON DENTAL RESEARCH
# AT THE NATIONAL BUREAU OF STANDARDS

## 1. Introduction

Research on dental materials, dental equipment and natural tooth structures was continued at the National Bureau of Standards during the half year ending June 30, 1964.

A list of reports issued, papers published and summaries of results obtained on work in progress are given below. Copies of the reports are appended.

## 2. Reports Issued

| | |
|---|---|
| NBS Report 8391 | Clinical Evaluation of Complete Dentures Made of Eleven Different Types of Denture Base Material. |
| NBS Report 8395 | Blood-Bone Equilibrium in Calcium Homeostasis. |
| NBS Report 8507 | Early Strength, Flow and Dimensional Changes Obtained on Amalgam Prepared with a Standardized Mechanical Technic. |
| NBS Report 8552 | Viscoelastic Behavior of Dental Amalgam. |

## 3. Papers Published

X-ray spectrometric analysis of noble metal dental alloys. B. W. Mulligan; H. J. Caul; S. D. Rasberry and B. F. Scribner. J. Research NBS (Phy. and Chem.) 68A:5, Jan.-Feb., 1964.

Fluorometric demonstration of tryptophan in dentin and bone protein. K. C. Hoerman and S. A. Mancewicz. J. D. Res. 43:276, March-April, 1964.

Physical properties of cements based on zinc oxide, hydrogenated rosin, o-ethoxybenzoic acid and eugenol. Simon Civjan and G. M. Brauer. J. D. Res. 43:281, March-April, 1964.

Detection of aging effects in cold-worked silver-tin alloys. R. M. Waterstrat and G. E. Hicho. J. Research NBS (Phy. and Chem.) 68A:317, May-June, 1964.

## 4. Work in Progress

### 4.1 Natural Tooth Structure

(a) Structure of Calcified Tissues.

Solubility of hydroxyapatite: The solubility of hydroxyapatite was found to vary depending on the previous treatment of the sample. Annealing the sample in a steam atmosphere for 4 days at 1000°C yields a material that has lower solubility than one that has not been annealed or has been annealed in a dry atmosphere. These results were obtained when equilibrium was approached from under-saturation, and, therefore, they should be indicative of differences in lattice free energy. At least part of these differences in lattice energy can be attributed to a disorder in the positions of the hydroxyl ions in the hydroxyapatite lattice that has been reported recently.

Mechanism of growth of apatitic crystals: When attempts were made to approach equilibrium from supersaturation, the solution composition did not reach the hydroxyapatite isotherm. This result was expected on the basis of a crystal-growth mechanism that we have proposed for hydroxyapatite which requires the intermediate formation of octacalcium phosphate, and substantiates the proposed mechanism.

Blood-bone equilibrium: In cooperation with Dr. James MacGregor (Gardiner Institute, Glasgow, Scotland) data on the solubility of bone in vitro were shown to reveal the presence of octacalcium phosphate in child and calf bone, but not in adult bone. This is in accord with the growth mechanism described above because the crystallites in "young" bone are in a more active state of growth. The presence of octacalcium phosphate in bone introduces a new concept relative to the chemistry of bone formation. A detailed report is appended.

Pyrolysis of octacalcium phosphate: Formation of pyrophosphate on heating of tooth and bone has been used to show that the phosphate enters the lattice in an acidic form as indicated above; we believe as octacalcium phosphate. The products formed in pyrolysis of octacalcium phosphate include $CaHPO_4$, $Ca_5(OH)(PO_4)_3$, $\beta$-$Ca_2P_2O_7$, and $\beta$-$Ca_3(PO_4)_2$, depending on the condition of heating. A chemical and infrared study of the pyrolysis products was carried out to obtain more information on these reactions. Work is continuing, to permit selection of conditions that would make the formation of pyrophosphate quantitative, so that the formation of pyrophosphate may be used as a precise analytical method.

Hydrolysis of octacalcium phosphate: Hydroxyapatite produced by hydrolysis of crystalline octacalcium phosphate is decidely layer-like in appearance as seen with the electron microscope. This appearance is in contrast to that of the directly precipitated hydroxyapatite, and it is in accord with the layer-type structure of octacalcium phosphate and with the observation that the principal mode of hydrolytic action is parallel to the layers. It is expected, therefore, that many of the physical properties of the two types of hydroxyapatite will be quite different, particularly those related to surface, and this would explain the presence of an "inner surface" that has been attributed to hydroxyapatite. Continuation of this study will include comparative surface area measurements before and after hydrolysis.

Tetracalcium phosphate: A single-crystal x-ray study of tetracalcium phosphate, $Ca_4O(PO_4)_2$, showed that it has a structural relationship to hydroxyapatite. The relationship is analagous to the one between hydroxyapatite and octacalcium phosphate, and in the same way this could lead to the formation of interlayered mixtures of the two salts which could account for the existence of apatitic materials with Ca/P ratios greater than that of hydroxyapatite.

(b) Heats of Wetting of Dentin.

Towards the end of the reporting period work on this program was again taken up. The surface area of ground dentin as determined by the BET method was found to be around 10 $m^2/g$. The smaller particle size dentin had a somewhat smaller surface area. The heat of wetting of dentin was found to be $0.8 \pm 0.08$ $cal/m^2$. After extraction of organic matter from dentin with ethylene diamine the surface area was found to have increased to 124 $m^2/g$.

(c) Organic Portion of Tooth Structure.

The role of tryptophan in the crosslinks of collagen was investigated by looking at normal and lathyric collagen. Lathyrism, a disease which can be experimentally induced, is thought to alter the crosslinks of collagen. There were differences in tryptophan content between newly-formed and older collagen from rat skins. These findings might be explained by assuming that only the alpha chain of collagen contains tryptophan. To date attempts to substantiate this thesis have been unsuccessful, mainly because of interfering fluorescence of an unidentified component. However, up to the seven day period of lathrogenic feeding, there apparently were no differences in tryptophan content of normal and lathyric collagen. If tryptophan were involved in the crosslink the tryptophan content would have increased or remained constant. The fluorometric results have not clearly defined the role of tryptophan in the fluorescence of rat skin collagen.

The principles which were used in a filter phosphorometer are being applied to the Aminco-Bowman spectrophotofluorometer, thereby allowing for work with monochromated energy. Most adaptation has been performed on the manner of cooling and dehumidifying the sample compartment. The instrument has been characterized for wavelength and intensity response. Phosphorescent spectra of solids can now be described. Initial spectra of carious dentin seem to agree with those of other investigators in that emission wavelength increases with concentration. Operation at optimal wavelengths will also increase the accuracy of decay curves and phosphorescent lifetime measurements, and may lead to differences in these properties of calcified tissue more pronounced than those shown by measurements with the filter phosphorometer.

## 4.2 Metals

### (a) Amalgam.

Properties of amalgam during the hardening period: Data were obtained on compressive strength, flow and dimensional change during the early hardening period of amalgam. The rate of hardening appears to be linear during the first hour of an amalgam. The initial shrinkage, early strength and flow are interrelated. A detailed report is appended.

Viscoelastic behavior of amalgam: A study of the rheological behavior of dental amalgam (composition approximately: Hg 50%, Ag 35%, Sn 13%, Cu 2%, Zn <0.5%) has been made by means of tensile creep tests. The tensile strain developed in amalgam was found to be a function not only of the applied stress but also of length of time of application of stress. Amalgam was shown to exhibit (1) strain hardening which is both stress and time dependent (2) elastic deformation which is a function of stress only (3) retarded elastic deformation which is both time and stress dependent but is recoverable upon removal of stress and (4) viscous deformation which is both time and stress dependent but is not recoverable upon removal of the stress. Both the retarded elastic and the viscous strains developed in amalgam were nonlinear functions of the applied stress. However, the viscous strain was a linear function of time while the retarded elastic strain was a nonlinear function of time exhibiting a spectrum of exponential form. It was shown that the viscoelastic data obtained from creep tests could be used to calculate tensile stress-strain curves for amalgam which agreed with those obtained experimentally at various stressing rates. This was done by use of the nonlinear form of the Boltzmann superposition principle. It is planned to correlate the basic physical properties with clinical service to determine a more scientific background for material standards and specifications. A detailed report is appended.

### (b) Gallium Alloys.

A program has been initiated to test certain gallium alloys for use as dental fillings by placing them in vital human teeth. The clinical phases of this work are being conducted at the National Institutes of Health. Preliminary results indicate that both gallium-palladium and gallium-gold alloys possess adequate corrosion resistance in the human oral environment for periods of up to 5 weeks. Clinical techniques for preparing and using these alloys are being developed. Pulp reactions will be investigated, however, no adverse reactions are apparent at the present time. The gallium alloys "wet" the surface of human teeth and possess adequate strength for use as a dental filling material. In addition their thermal expansion coefficients more closely match those of tooth structures than does the coefficient of dental amalgam. Gallium alloys are also extremely resistant to "flow" or "creep" under load. Modifications of the basic alloy compositions are presently being investigated in order to provide any further improvement in their properties which would facilitate their use as dental fillings.

## 4.3 Resins

### (a) Silica-Reinforced Direct Filling Resins.

Effective means were found for the treatment of various silica powders with the coupling agent, 3-(trimethoxysilyl)propyl methacrylate. The copolymerization characteristics of a crosslinking monomer (obtained by the addition of methacrylic acid to the diglycidyl ether of bisphenal A) were investigated. This highly reactive monomer is unique in having an unusually long chain segment connecting the methacrylate groups, and in resembling epoxy resins although polymerizing by a free radical mechanism.

### (b) Adhesion Studies.

A method was devised for measuring the tensile stress that develops during the polymerization of adhesive resins. This is done by observing the loading required to maintain constant distance between two opposing walls (adherends) of unit area. The data being obtained will help answer the question: How strong must the adhesion be to the opposing walls of a cavity, to withstand the polymerization shrinkage?

## 4.4 Zinc Oxide-Eugenol Materials

### (a) Reaction Mechanism.

Polarographic studies of the change of the zinc half-wave potentials of solutions containing $Zn^{++}$ and eugenol or o-ethoxybenzoic acid were undertaken to obtain information of the number of liquids complexing with zinc and the determination of chelate stability constants of the complexes formed. Since eugenol forms a five-membered chelate whereas EBA may give a six-membered ring, these constants would show the relative stability of the two ring system.

2,5-dimethoxyphenol was synthesized and found to harden with zinc oxide.

(b) Ionization Constants of Substituted Benzoic Acids.

The thermodynamic pK values of benzoic acid and the substituted benzoic acids in 0 to 62.4 weight percent (0 to 70 volume percent) ethanol were measured. With increasing ethanol concentration the pK values increase markedly from 4.2 for aqueous to 6.5 for the 62.4 weight percent ethanol solution due to the decrease in the ionsolvating power and dielectric constant of the solvent. The rate of change of pK values with solvent concentration increases with increasing ethanol content of the medium until a maximum is reached in the 24.7 to 42.5% ethanol region. With the exception of the p-propenylbenzoic acid the values of the Hammett sigma constants do not change greatly with ethanol concentration.

Ultraviolet absorption spectra of substituted allyl and propenylbenzoic acids and phenols were obtained to determine if shifts in the position of the absorption maxima can be correlated with (1) Hammett reactivity constants and resonance parameters of substituents (2) pK values of the respective compounds (3) the degree of hydrogen bonding. On plotting the displacement of the absorption maxima versus the pK values of the substituted guaiacols the three allyl substituted compounds investigated fall approximately on a straight line with a positive slope and the propenyl compounds on a straight line with a negative slope. Intramolecular hydrogen bonding produces wavelength displacements towards the visible spectrum of all hydrogen derivatives with the exception of the sterically hindered o-eugenol.

## 4.5 Waxes

The flow at 30°C, 37°C, 40°C and 45°C of about 50 binary mixtures of waxes were determined on compositions varying by 10 percent weight increments. As the interest centers around the development of materials by which functional impressions may be taken or in the development of temporary denture relining materials, the binary combinations were selected on the basis of the respective flow above and below body temperature (37°C). Flexowax C light, Ultraflex, C-905 wax and paraffin wax 124 (Cornelius) which had a high flow at 37°C, and below were melted in binary combinations with other waxes which had little or no flow at 37°C. Some of the binary combinations did not mix when melted and some mixed in the melted state but separated out on freezing. This was usually discernable by the asymmetry of the test specimen after being subjected to the load.

Binary combinations of any of the high flow waxes with Singapore Gum, Paraffin wax 138/141, Paraffin wax 160/165 or Ozokerite #870 in any proportions flowed satisfactorily as evidenced by the symmetry of the disks formed when the specimen was loaded.

## 4.6 New Test Methods

(a) Diametral Compression Test.

Evaluation of the diametral compression test as a method for determining the tensile strength of some dental materials has been started. The method involves the application of compressive forces to opposite sides of a cylindrical specimen. This results in a tensile fracture in the plane parallel to the axis of the specimen and the direction of loading. The method has the advantage that specimen preparation is simple, no grips are needed to hold the specimen, and a standard testing machine may be used to determine strengths. Preliminary results on silicate cements suggest that the nature of the padding material used between the specimen and the testing machine platens is one of the most critical factors in the reliability of the test method.

(b) Diffraction Grating Strain Gages.

The use of optical diffraction gratings for measuring strain in small specimens of dental materials is being investigated. The method provides the possibility of using very short gage lengths. A ruling machine has been reconditioned and gratings have been ruled on amalgam specimens. Apparatus for recording the changes in angle of diffraction as the specimen is strained is being developed.

## 4.7 Environmental Testing

In space or at high altitudes, personnel may breathe pure oxygen rather than air. To investigate the possible effect of an oxygen atmosphere on amalgam and silicate cement, compressive strengths were determined on specimens stored at 37°C in air and in oxygen both with and without the presence of water. As shown in the table, for amalgam specimens 2 months old and silicate specimens 3 weeks old no differences were found between specimens stored in oxygen and those stored in air. Specimens stored in water had slightly lower compressive strengths than did specimens stored in either dry air or dry oxygen. These data indicate that inhalation of oxygen would not be deliterious to the physical properties of the two most used filling materials.

Compressive Strength

| | | Storage Medium | |
|---|---|---|---|
| Material | Age | Water | Dry Air |
| Amalgam | 1 mo. | 38,600 psi | 43,900 psi |
| Amalgam | 2 mo. | 39,300 | 43,800 |
| | | Water-oxygen(a) | Dry air-oxygen(b) |
| Amalgam | 2 mo. | 38,200 | 44,400 |
| | | Water | Dry Air |
| Silicate Cement | 24 hr. | 24,400 psi | 29,100 psi |
| Silicate Cement | 3 wk. | 21,400 | 25,800 |
| | | Water-oxygen(c) | Dry air-oxygen(d) |
| Silicate Cement | 3 wk. | 22,[illegible]00 | 26,200 |

(a) Sealed in tube containing oxygen and water after 1 month in water.
(b) Sealed in tube containing dry oxygen after 1 month in air.
(c) Sealed in tube containing oxygen and water after 24 hr. in water.
(d) Sealed in tube containing dry oxygen after 24 hr. in air.

## 4.8 Evaluation of Materials

Materials evaluated for the Federal Dental Services or the American Dental Association by specification or other test methods included amalgam alloys, gold casting alloys, denture base resins, acrylic teeth, a zinc oxide-eugenol impression paste and carbide burs.

## 4.9 Specifications and Standards

A considerable amount of time has been devoted by the staff of the Dental Research Section to the development of specifications and standards for dental materials. This effort involves both laboratory and committee work. The specification developments eventually are formalized as American Dental Association, Federal, American Standards Association and Federation Dentaire Internationale specifications. Dental Materials Group specification subcommittees on which staff members are active as either chairmen or secretaries include: amalgam, burs, direct filling resins, toxicity, gypsum, rubber base impression materials, endodontic files, reamers and points, diamond rotary instruments, radiographic film, casting gold alloy, denture base resin, denture base relining resin, zinc silicophosphate cements, and duplicating materials.

For the Director
By

W. T. Sweeney

W. T. Sweeney
Chief
Dental Research Section

USCOMM-NBS-DC - 26,081

# NATIONAL BUREAU OF STANDARDS REPORT

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# Progress Report

# on

# Clinical Evaluation of Complete Dentures Made of Eleven Different Types of Denture Base Materials

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# Clinical Evaluation of Complete Dentures Made of Eleven Different Types of Denture Base Materials

## Abstract

A subjective clinical examination and the objective laboratory measurements of 63 dentures from three to six years old showed a gradual decrease in serviceability with time without significant dimensional changes or warpage of the dentures in use. The relative effectiveness of a properly made denture depends upon how well the tissues of the wearer tolerate the stresses imposed in use. The findings demonstrate that subjective observations with proper discipline can furnish reliable and valid information in a clinical evaluation of dentures.

## Introduction

A clinical evaluation of a denture is largely subjective. However, it is possible to set up clinical criteria which will aid one in comparing dentures made of different materials and by different processing methods. Moreover, it is important to observe dentures, critically and periodically, over an extended period and if possible to correlate the laboratory with the clinical investigations. Apparently this has not been done previously.

The authors have attempted to do this in a series of reports[1-7] on 73 clinical dentures and 112 technic dentures made of 12 different types of denture base materials. All of these dentures were made from August 1957 to August 1958 for patients who had worn dentures previously or who had been edentulous for some time.

Sixty-three of the 73 dentures worn by 38 patients were still available and were examined during the summers of 1959, 1960 and 1961, and 53 were still available for examination during the summer of 1963. Thus the mortality of the sample was about 14 percent in three to four years and about 27 percent in five to six years.

Changes were noted in centric and eccentric occlusions, in retention and stability of the dentures, in the condition of the tissues beneath the dentures and in the appearance of the dentures. The serviceability of the dentures was judged on the basis of the dentist's examinations and the patients' opinions. Pressure-indicator-paste patterns were used to show the relative apposition of the tissues to the dentures.

## Type of Denture Base Materials

The brand and type of denture base materials from which each denture was constructed, the method of processing the dentures, upper and lower designations and the length of time that each denture was under observation are shown in Table 1, Columns 1-3. These denture bases were selected as being representative of different types of resins or because the bases were processed by special methods and equipment or by a combination of both. Vulcanite (hard rubber) was included as some dentists think that vulcanite dentures fit better and retain the fit longer than dentures made with an acrylic resin base.[8,9] Evidence for such an opinion was not found in the literature.[2]

## Dimensional Changes of Dentures

Processing shrinkage. The molar-to-molar shrinkage during processing (Table 1, Column 4) was higher on the lowers than on the uppers in the 25 sets of dentures with only four exceptions. The average shrinkage on the uppers on these 25 sets was 0.32 percent; on the lowers, 0.44 percent: a difference of 0.12 percent. This difference was not detected clinically as it represents only 0.06 of a millimeter in a 50 millimeter molar-to-molar span.

When the lower denture was thick in cross-section (dentures A-7-L, D-7-L, H-2-L, H-10-L, J-4-L and T-4-L in Column 2) the processing shrinkage of the lower denture was near to that of the upper. The unusually large molar-to-molar shrinkages in two upper dentures (0.70 percent in denture T-5-U and 0.83 percent in denture T-7-U) were caused by decreasing the palatal thickness of the cured dentures that had been waxed up too thick. The thinning procedure in each instance about doubled the shrinkage that occurred when the denture was removed from the cast, because these dentures, after being reduced in cross-section, were not stiff enough to prevent release of some of the strain accumulated during processing. Therefore, the wax denture should be carved to the proper thickness initially.

The differences in processing shrinkage among individual dentures of the same material is caused by variation in the size and shape of the dentures.[3,4] Thickness is especially important since it governs the stiffness, thus limiting the degree of contraction of a denture. If the denture is thin in a critical cross-sectional area, such as in the lower anterior region, much of the strain placed in the lower denture during processing will be released when the denture is removed from its cast. This release of strain causes the contraction which is primarily manifested in the posterior section of the dentures because of their shape.

The average processing shrinkage of the clinical dentures made of each base ranged from a low of 0.18 percent for the Epoxolon dentures to a high of 0.58 percent for the Tilon dentures and, in general, followed the same pattern as technic dentures made from the same materials.[3] The values in Column 4 reflect the shrinkage of the molar-to-molar dimension caused by removal of the denture from the gypsum cast, finishing, polishing and soaking in water for one day just prior to delivering the denture to the patient.

The processing shrinkage (molar-to-molar) of individual dentures ranged from 0.06 percent (0.03 mm.) on upper denture E-6-U to 0.97 percent (0.41mm) on lower denture T-8-L. The value on denture E-6-U was low because it was an upper denture made with an epoxy resin base which had a low polymerization shrinkage especially when processed as recommended at a low temperature (49°C). The value on denture T-8-L was high because it was a large thin lower denture molded in dry heat at a high temperature [110°C (230°F)]. These two dentures fit well on insertion. No post insertion adjustment was necessary on T-8-L.

Dimensional changes in service. Both expansion and contraction occurred in molar-to-molar distance (Table 1, Column 5) and in flange-to-flange distance (Column 8) during use. The dimensional changes in the clinical dentures were almost nil after about three months except for those made of Epoxolon which continued to expand during the three to four years of observation, and for the vulcanite dentures which shrank constantly. The extremes of change ranged from a molar-to-molar expansion of 1.87 percent (0.78 mm) on lower denture E-3-L to a shrinkage of 0.55 percent (0.31 mm) on upper denture Vul-1-U.

Seven of the 63 dentures were so stable in dimension that the molar-to-molar change was no more than 0.05 percent. The flange-to-flange changes ranged from an expansion of 1.27 percent on lower denture E-5-L to a shrinkage of 0.74 percent on upper denture A-6-U. Fifteen of the 63 dentures had a flange-to-flange change of no more than 0.05 percent.

Thus, the values in Columns 5 and 8 (Table 1) reflect the dimensional stability of the dentures in service for from three to six years (Column 3).

Cumulative changes. The cumulative molar-to-molar changes from the time the cured denture was in the flask until the end of the observation period (Column 3) were obtained by adding algebraically the values in Columns 4 and 5 and are given in Column 6. Because all measurements were made at room temperature [23 ± 2°C (73 ± 4°F)] and because the dentures function at mouth temperature [37°C (98.6°F)], a correction for the thermal expansion of the resins has been made for the 14°C range (23 to 37°C). The correction was derived from the coefficient of thermal expansion of each of the denture base resins[6] and amounted to 0.13 percent for Acralite 88, 0.12 percent for Duraflow, 0.10 percent for Epoxolon, 0.12 percent for Hydro-Cast, 0.09 percent for Jectron, 0.10 percent for Luxene 44, 0.07 percent for Miracle 50, 0.06 percent for Mystic 100, 0.12 percent for Tilon, 0.14 percent for Vernonite (powder-liquid) and 0.11 percent for vulcanite. The data in Column 7 were obtained by adding algebraically the foregoing corrections to the values in Column 6.

A near zero value in Column 7 shows that the molar-to-molar dimension of the denture in the mouth at the end of the observation period was practically the same as the molar-to-molar dimension of the cured denture unflasked to the occlusal surfaces. Eight of the dentures were 0.00 ± 0.05 percent. The individual values ranged from +1.74 percent on lower denture E-3-L to -1.10 percent for lower denture Mi-2-L. On the average dentures made from the different bases ranged from a low value of +0.07 percent for the Duraflow dentures to a high of +1.41 percent for the Epoxolon dentures. Considerable variation among dentures constructed of the same material occurred because of size and shape of the dentures varied from patient to patient.

On the average the dimensional changes in the lower dentures over the molar-to-molar span were slightly more (0.42%) than the changes in the upper dentures (0.35%).

On the average, the acrylic resin dentures processed by conventional compression molding (Acralite 88 +0.16%; Duraflow +0.09%; Vernonite, powder-liquid, -0.14%) were just as accurate as those processed by injection molding (Tilon -0.13%) or by hydraulic molding (Hydro-Cast -0.17%) and were more accurate than the polystyrene dentures (Jectron -0.25%), the vinyl-acrylic dentures (Luxene 44 -0.32%) and the vulcanite dentures (-0.70%).

Warpage index. A comparison of molar-to-molar changes occurring during service (Table 1, Column 5) with the flange-to-flange changes (Column 8) shows whether these dimensional changes in the posterior sections of the dentures were proportional. If the molar-to-molar and the flange-to-flange changes were not proportional, then a warpage or a twisting of the denture had occurred. Only five dentures (A-6-U, E-1-U, E-3-L, J-8-U, and T-7-U) had more than a 0.5 percent warpage index. A zero value shows that there was no warpage and that the molar-to-molar and flange-to-flange changes were proportional. Very little distortion occurred in the clinical dentures. On the average, the most warpage occurred in the Epoxolon dentures (0.41%) and the least in the dentures of Luxene 44 (0.10%). Dentures that are to be considered the most accurate in all respects should have low values in Column 7 as well as having a low warpage index (Column 9). On the average the most accurate dentures were made of Duraflow, Hydro-Cast and Vernonite, powder-liquid, as they had the three lowest combined values in Columns 7 and 9; the least accurate dentures were made of Epoxolon, vulcanite and Miracle 50 as they had the three highest combined values.

The average warpage index for the lower (0.13%) was half that of the upper dentures (0.27%). This is probably caused by the thinner and more extended posterior flanges of the upper dentures.

## Changes in Occlusion

The centric and eccentric occlusions of the clinical dentures were originally established in the wax dentures on a Hanau Model H articulator by an arbitrary face-bow registration and by centric relation and protrusive interocclusal records which were made in beeswax. The accuracy of all mountings on the articulator was verified by observation of the trial dentures in the mouth during repeated closures. Since teeth with cusps were used on all clinical dentures, inspection in the mouth had considerable reliability. If centric relation in the mouth did not coincide with that on the articulator, a new centric interocclusal record was made and the lower cast was repositioned. Additional jaw relations records and remountings were made, if necessary, until centric relation as determined by testing the wax dentures in the mouth coincided with centric occlusion as established on the articulator. Then a protrusive interocclusal record was made in beeswax and the articulator was adjusted accordingly.

After processing but before the dentures were removed from the gypsum casts, they were precisely repositioned on the articulator using metal split remounting plates (Hanau). Any vertical change in occlusal relations was determined by measuring the increase in height of the incisal guide pin,[3] but no correction was made. The lower denture, with its cast and mounting ring, was removed from the articulator. A remounting facilitator was then fastened on the lower member of the articulator and a gypsum index of the occlusal and incisal surfaces of upper teeth was made to preserve the face-bow registration. The dentures were removed from their gypsum casts, polished and the upper denture was repositioned with plaster on the articulator using the facilitator and gypsum index.[7] To correct for processing and other errors before allowing the patient to wear the dentures, a new centric interocclusal record was made in the mouth so that the lower denture could be positioned correctly on the articulator. The necessary corrections in occlusion were made by selectively grinding the porcelain teeth with a carborundum stone. Thus at the time of insertion the centric and eccentric occlusal relations were well established.

Each patient was recalled in 24 hours, at the end of three days and subsequently, as necessary, to make post-insertion adjustments of the denture. On the average it was necessary to make 2.5 adjustments on each patient during the first month. The number of post-insertion adjustments could not be related to the type of denture base, to the processing method, to the dimensional changes or to the physical and chemical properties of the denture base materials.

Evaluation of centric occlusion and centric relation. Centric occlusion and centric relation were evaluated by inspection in the mouth during the summers of 1959, 1960, 1961 and 1963. No remountings were made on the articulator to correct observed discrepancies except in two instances. The criteria used to judge the relative occlusion in centric relation were:

Excellent - The inspector could detect no fault in centric occlusion when the patient closed the jaw in centric relation. A value of 4 was recorded

Good - A slight objectionable contact, possibly caused by only one cusp, which made the dentures slide slightly (1/2 mm or less) to get into centric occlusion from centric relation. The sliding was not sufficient to tip or dislodge the dentures. A value of 3 was recorded.

Fair - Objectionable occlusal contacts on one side which caused the dentures or the lower jaw to move about a millimeter to get the teeth into centric occlusion from the first contact in centric relation. A value of 2 was recorded.

Poor - there was a gross error in the occlusal relations which could only be corrected by resetting the teeth or by rebasing the denture or both. A value of 1 was recorded.

The values assigned to represent the foregoing and any subsequent clinical criteria are arbitrary as is the assignment of equal importance to all of the conditions evaluated.

The ratings of the dentures are given in Table 1, Column 10. In general, little change in the centric occlusion-centric relation examinations occurred during the first two years. There was a slight drift toward a deterioration during the third and fifth year. About 24 percent of the dentures were rated 4 (excellent) in 1959; about 8 percent in 1963. A comparison of the ratings in 1959 with those in 1961 show a deterioration of the centric occlusion in about 44 percent of the dentures, no change in about 35 percent and an improvement in the relations for about 21 percent. These changes and the further slight deterioration at the 1963 examination cannot be related to the changes in dimension (Table 1, Columns 4-9). Dentures H-9-U and H-10-L were excluded because the posterior teeth on H-10-L, which had been depressed by the hydraulic molding forces during processing (Hydro-Cast), were reset into proper occlusion.

There was a deterioration in dentures J-1-U and J-2-L from 1959 to 1960 caused by the patient's newly acquired habit of pipe smoking. An improvement from 1960 to 1961 was effected by instructing the patient to alter his pipe-holding position.[7]

Evaluation of eccentric occlusion. The criteria used to judge the relations of the teeth in lateral and protrusive eccentric jaw positions were:

Excellent - All of the working and balancing contacts of the teeth occurred without cuspal interference when the mandible was protruded and moved laterally. A numerical rating of 4 was recorded.

Good - The same as excellent except that either a working or a balancing contact was absent. A value of 3 was recorded.

Fair - There was a limited degree of freedom caused by cuspal interference and one or more of the balancing contacts were absent. A value of 2 was recorded.

Poor - There were either no balancing contacts or there was severe restriction of the movements caused by interlocking cusps, or both. A value of 1 was recorded.

The numerical ratings for eccentric occlusions are given in Table 1, Column 11. Like the centric occlusions, the eccentric occlusion had a slight deterioration with time.

This deterioration is attributed to changes in the tissues upon which the dentures rest and not to any warpage or change in dimension of the dentures. A comparison of the warpage index of the various dentures (Table 1, Column 9) with the deterioration in occlusion (Columns 10 and 11) shows no correlation. A similar comparison using total dimensional changes in the dentures (Table 1, Column 7) instead of the warpage index shows no relationship with the slight deterioration in occlusion. The data in Table 1 do not support the opinion that teeth shift position in the acrylic resin dentures and thereby alter the occlusion. Gross errors in the occlusal relationship were found to have been cuased by a shifting of the dentures on the tissue as it changed and by a patient's habit, without any movement of teeth or warpage of the denture.[7]

## Changes in Retention

The degree of retention or resistance to dislodgement of the dentures was estimated by applying force with the fingers and thumb as shown in Figure 1.

The criteria used in judging the relative force, applied in the direction away from the tissues, necessary to dislodge the dentures were:

Excellent - The denture was very difficult to disloge and appeared to have little or no movement before dislodgement. A value of 4 was recorded.

Good - The denture was difficult to dislodge but moved away from the tissues slightly before dislodgement. A value of 3 was recorded.

Fair - The denture was dislodged easily. A value of 2 was recorded.

Poor - The denture offered little or no resistance to dislodgement. A value of 1 was recorded.

(The retention of the lower dentures was tested with the tip of the tongue in the lower incisor region.)

The average relative retention during the observation period (Column 3) is shown in Column 12. Retention decreased on the average about 31 percent. The decline was comparatively small from 1959 to 1960 and then a decline of about 0.3 in the rating occurred annually from 1960 to 1963. Thus dimensionally the tissues were changing slowly but very uniformly while the dimensions of almost all of the dentures were not. The decline in retention is therefore caused by tissue changes. Usually the tissues could not withstand the continued trauma caused by using dentures.

At the 1959 examination 48 percent of the dentures had a rating of 4 (excellent) for retention and only 6 percent were rated as 1 (poor). By 1963 the retention had changed so that only 9 percent of the dentures were in the excellent (4) category and 25 percent were now rated as poor (1). There were only four dentures in which the retention was 4 (excellent) when tested in 1959 and which remained 4 (excellent) at the 1963 examination, namely H-4-L, H-10-L, T-6-L and T-9-U.

Of the 53 dentures that could be examined over the five years, 1959-1963, the upper dentures had, on the average, a score of 2.9 which was a little better than that for the lower dentures 2.6.

## Changes in Stability

The degree of stability or movement of the dentures on the tissues was estimated by applying forces with the fingers or thumb as shown in Figure 2.

The criteria used in judging the relative force, applied in a direction toward the tissues, necessary to move the dentures on the tissues were:

Excellent - The denture had little or no movement on application of strong direct or rotary force. A value of 4 was recorded.

Good - The denture had little or no movement on application of strong rotary force (Fig. 2) but moved and was dislodged when a strong direct force was applied to one side or to the front of the denture. A value of 3 was recorded.

Fair - The denture had considerable movement on application of rotary force and was dislodged by moderate direct force. A value of 2 was recorded.

Poor - A slight force, either rotary or direct, caused the denture to move and become dislodged. The denture should be rebased or relined. A value of 1 was recorded.

The evaluation of the stability is given in Table 1, Column 13. On the average, there was an impairment of stability during 1959-1961. The poor stability in 1961 had not changed by 1963. At the 1959 examination, 30 of the dentures were rated as 4 (excellent), in 1961, only 14 dentures were so rated and 14 were rated as 1 (poor). Contrary to much general opinion among dentists and patients, the lower dentures had as much stability as the upper dentures when first evaluated (3.0 and 3.1, respectively). Furthermore, the clinical scores for the stability of the lowers on the average, decreased less with time than for the upper dentures (2.6 and 2.4, respectively).

A dentist would expect the retention to decrease less with time than stability. This was reflected in observations. The retention of dentures is directly related to the border seal and should not be influenced as much as stability by changes in contour of the tissues that support the denture.

### Evaluation of the Condition of the Tissues Supporting the Dentures

The criteria used to judge the condition of the tissues supporting the dentures were:

Excellent - The tissues were firm and had good color. There were no signs of abrasion or other injury caused by the dentures. A value of 4 was recorded.

Good - The tissues were generally firm and of good color, except for small isolated areas. A value of 3 was recorded.

Fair - On the crest of the ridge there was some moveable tissue which was not present previously, or there were irritated areas with poor color involving as much as one-third of the denture-bearing surface. A value of 2 was recorded.

Poor - There were either large general areas of redness involving one-half or more of the denture-bearing surface, or there was a considerable amount of movable tissue not present previously, or both. A value of 1 was recorded.

The evaluation of the condition of the tissues covered by the dentures is shown in Table 1, Column 14. The healthiness of the tissues decreased during this study under 39 of the 63 dentures. At the 1959 examination, the condition of the tissues beneath 52 of the 63 dentures was classified excellent (4) or good (3) and only two were in poor (1) condition. At the 1961 examination, only 27 of the 63 could be classified as in excellent or good condition, and 14 were now poor.

There was little difference between the healthiness of the tissues beneath the upper and lower dentures of the same set, even though the bearing surface of the upper is much larger than that of the lower denture. This results in a higher stress per unit of surface area in the lower denture and should cause more trauma.

The average value for the condition of the tissues supporting 26 upper dentures which were opposed by lower dentures was 2.6 while an average of 1.6 occurred in 11 instances where the upper denture was opposed in part by natural dentition. This finding confirms the general opinion that the tissues supporting an upper denture are traumatized more when the lower dentition is present in whole or in part than when a lower opposes an upper denture.

The condition of the supporting tissues was on the average, 2.8 for the 16 dentures with an excellent (4) rating for stability and 1.8 for the 14 dentures with a poor (1) rating for stability. This relationship does seem reasonable because stable dentures should irritate the supporting tissues less than unstable dentures.

In only 4 out of the 63 dentures (H-3-U, L-2-U, L-6-U and V-5-L) did the conditions of the oral tissues improve. The improved condition of the tissues supporting upper denture L-2-U occurred after the patient began removing the dentures at night.

### Serviceability - Patients' Opinions

Each patient was asked the following three questions in 1959, 1960, 1961 and 1963:

1. How have you been getting along with your teeth since we last saw you?
2. Do you have any trouble at all with your dentures:

3. Do your dentures fit as well, not as well, or better than one year ago?

The patients' replies were taken down verbatim without their knowledge. These replies were evaluated by the following criteria:

Excellent - Patient was well satisfied. The dentures were better than they had ever expected. They had no difficulty with them. A value of 4 was recorded.

Good - Patient was well satisfied. They got along well in speech and function but had some minor complaint about a small problem. A value of 3 was recorded.

Fair - Patient was getting along alright by making adjustments or acquiring habits to overcome an existing difficulty. A value of 2 was recorded.

Poor - Patient wears the denture with some difficulty, due to looseness or discomfort, or does not wear it at all. A value of 1 was recorded.

During the first two years of observation, all but 5 of the patients rated their dentures as 4 (excellent) or 3 (good). The ratings as given by the patients changed very little with time. Many of the dentures lost much of their former stability and the condition of the oral tissues had deteriorated, but usually the patient had learned to accommodate by acquiring new muscle habits, especially those of the tongue.

Invariably, the patients were unaware of the severity of the irritation or of the deterioration of the condition of the tissues beneath their dentures. Undoubtedly, if similar conditions developed overnight rather than over a period of many months, the patients would be very aware of the discomfort and impaired fit.

Since the patient's opinion is so unreliable periodic examination should be made on all patients wearing dentures.

## Serviceability - Dentist's Opinion

The subjective ratings of one of the authors (J.B.W.) was based upon the following criteria:

Excellent - The best prosthodontic restoration that could be accomplished under existing oral conditions was recorded as 4.

Good - If the functioning of the dentures was satisfactory but could be improved further by making minor adjustments in the bearing surface, or the occlusion, or both, a value of 3 was recorded.

Fair - The dentures functioned and occluded fairly well but were reaching the end of their period of reasonably satisfactory service. Adjustments were needed to prolong the service. A value of 2 was recorded.

Poor - The dentures needed to be rebased or to have the posterior teeth reset or new dentures should be made. Prolonged wearing of the dentures under the existing conditions might harm or destroy some of the supporting tissue. A value of 1 was recorded.

A comparison of the dentist's rating of the dentures (Table 1, Column 16) with that of the patients' (Column 15) demonstrates vividly the inability of the patient to assess the fit and functioning of the dentures they are wearing. This tendency of the patient to rate his dentures improperly increases with time. At the first examination in 1959, the average rating by the patients and the dentist was 3.6 and 2.9, respectively. The difference was 0.7 points. At the third examination in 1961, the difference had almost doubled at 1.3. These data show the ineffectualness of patients in rating dentures and of the futility of personal testimonials as a basis of assessment of fit or function of dentures. Only in 9 out of the 63 dentures worn by 5 out of the 38 patients, did the patients' and the dentist's ratings agree at the third annual examination in 1961 and in no instance was the dentist's rating higher than that of the patient.

## Deterioration

The average total scores for each denture at the end of each observation period are shown in Table 1, Columns 17 through 20. These values were obtained by averaging the scores for the occlusion, retention, stability, condition of tissue and serviceability of each individual denture in 1959, in 1960, in 1961 and in 1963. The efficiency and usefulness of dentures declined with time in about two-thirds of the dentures. The average scores for all of the dentures (bottom of Columns 17 through 20 and in Fig. 3) show a continuous deterioration of the supporting tissues and indicate that this tissue deterioration will continue. Again this assumption is based upon the dimensional stability of the dentures in service.

The highest individual average scores are 3.6 and 3.7 (Column 21), respectively, for dentures H-3-U and H-4-L and can be attributed to the excellent alveolar ridges, which were large, favorably shaped, firm and held up well under the trauma caused by the dentures.

The lowest score 1.6 (dentures My-1-U and My-2-L) (Column 21) reflects the general unsatisfactory behavior of resin denture bases containing glass fibers. The dentures could not be polished satisfactorily as the fibers were exposed. These fibers are not thoroughly impregnated by the resin; hence they come out of the surface.

The dentures made of the epoxy resin (Epoxolon) not only had very large and continued dimensional changes but were porous, stained badly, had poor color stability, and developed large crevices around the porcelain teeth. These crevices together with the porosity and staining made the dentures very unhygienic. Then, too, many posterior teeth came loose.[4]

Patients difference prevents the establishment of any rating of the different denture bases in clinical testing on small numbers of patients. For this reason there is considerable scattering among the clinical scores and the dimensional changes of individual dentures made of the same resin. In fact, one could probably say that if a denture is properly made, its relative effectiveness depends on how well the tissues tolerate the stresses imposed by the denture. In this connection, it would seem good practice to remove the dentures at night to give the tissues an opportunity to recover from the daytime injuries and thus retard the deterioration of the tissues.

There is no definite relation between the molar-to-molar dimensional changes in dentures (Table 1, Column 7) and the general satisfactoriness of dentures as reflected by the average scores in Column 21. This confirms the clinical observation previously expressed in this report that the dimensional changes involved in processing the plastic denture bases and their subsequent dimensional changes in service cannot be detected by the patient or the dentist.

## Evaluation by Pressure-Indicator-Paste Patterns

During the summer of 1958, all patients were recalled to make and photograph manually executed (Fig. 4) pressure-indicator-paste (Mizzy) patterns of the patients' old as well as their new dentures (constructed for this study). These new dentures had been worn from 3 to 12 months when the first patterns were made. Without exception, the patterns made in the new dentures showed a more even distribution of contact with the tissue than did the patterns made in the old dentures.[4]

At this first examination in 1958, through the use of indicator paste, areas of undesirable hard contact between the new denture base and the supporting tissues were discovered in about one-third of the dentures. In these instances a small amount of resin was removed with a number 8 round bur. Thus, the hard contact of the base with the tissue was relieved. Then a new pressure-indicator-paste pattern was made and photographed. In 1959 similar adjustments and photographs of the indicator-paste patterns were made on 11 dentures before and after removing any resin, and in 1960 on only three dentures. None of the denture bases were modified at the 1961 examination as it was desired to observe further deterioration in the serviceability without an attempt to correct discrepancies. There were zones of undesirable hard contact observed in the 1961 patterns in almost half of the dentures. The earlier adjustments to the dentures were made to improve comfort, to relieve irritated tissues, to improve stability and to determine if the patients could detect any difference. Only two patients were able to notice any improvement.

An example of the change in contact between dentures and the supporting tissues that is brought about by relieving the zones of hard contact seen in pressure-indicator-paste patterns is shown in Figure 5.

A comparison of the first set of patterns, made before any alterations (A in Fig. 5), with the fifth set, made after each denture was relieved in the areas of hard contact shown in four successive patterns made at the same sitting, (B in Fig. 4) shows the improved apposition of the dentures to the supporting tissues. The patient could not detect the difference reflected in these patterns. Patterns made in years subsequent to the foregoing 1958 patterns show a marked deterioration in apposition. The 1963 pattern is shown in C of Figure 5. During these five years the greatest dimensional change in the posterior of the dentures was an expansion of only 35 microns (0.0014 in). Here again the deterioration in the apposition of the denture to the tissues was caused by tissue changes and not dimensional changes in the dentures.

Photographs were made of manually executed pressure-indicator-paste patterns in 1958, 1959, 1960 and 1961 on 63 of the original 73 clinical dentures and in 1963 on 53 of the dentures.

This series of pressure-indicator-paste patterns showed that the change in apposition of the tissues to the dentures was not dependent on dimensional change in the dentures. Even though denture E-2-U had a gradual molar-to-molar expansion of 1.55 percent in three and one-half years (Table 1, Column 7) the pressure-indicator-paste patterns did not change appreciably (Fig. 6), which indicates that the tissues accommodated to the larger but gradual expansion. On the other hand, the upper Hydro-Cast denture, which had a molar-to-molar change of only -0.14 percent in three years, gave pressure-indicator-paste patterns that showed deterioration of the apposition of the tissues to the denture (Fig. 7). In this instance, the tissues changed but the denture changed very little in dimension.

## Pressure-Indicator-Past Patterns made with Manual Force and with Occlusal Force

As previously stated, for the yearly examinations, all of the dentures containing pressure-indicator-paste were seated manually as shown in Figure 4, rather than having the patients simulate chewing. The authors felt that in this way better uniformity could be maintained from one year to the next in the forces used to position the dentures, particularly if a change in the occlusal relationships had occured in the interim. With two patients where gross errors in the occlusal relationships were observed, a comparison was made between pressure-indicator-paste patterns in their dentures which were seated manually and those seated by the patients simulating chewing for 30 seconds.

With the set of Jectron dentures (J-1-U and J-2-L) which, because of a pipe habit, occluded prematurely on the right side by about 1mm in the centric relation-centric occlusion position,[7] large zones of hard contact between the denture bases and tissue are observed with the heaviest contacts on the right side when the patient simulated chewing (A and B in Fig. 8). However, when these same dentures containing indicator paste were seated manually, there were considerably fewer areas of hard contact between the dentures and tissues and these areas were more evenly distributed on both sides of the arch (C and D in Fig. 8).

With denture L-8-U, which by 1960 occluded with only nine lower natural anterior teeth, the centric relation-centric occlusion relationship was rated as 2 (fair) because of a premature anterior contact of teeth on the left side. The pressure-indicator-paste pattern made in this denture (L-8-U), by having the patient simulate chewing, reflects that heavy anterior occlusion as a large zone of hard contact is observed in the anterior one-third of the hard palate (A in Fig. 9). However, when another pattern was made in denture L-8-U by seating it manually, the hard contact on the rugae is not seen (B in Fig. 9).

Such occlusal forces as these have a direct effect on the position and size of areas of hard contact between the denture and the supporting tissues. Pressure-indicator-paste patterns which are made in the dentures by having the patient simulate chewing are unreliable and therefore should not be used as a guide to make alterations in the denture bases. Any necessary modifications to the resin bases as determined by the use of pressure-indicator paste should be done prior to occlusal corrections so that the improved occlusion will help maintain the more uniform contact with the supporting structures.

## Summary and Conclusions

A subjective clinical evaluation of 63 dentures, from three to six years old and made of 11 different types of organic denture bases, showed a gradual deterioration with time in the centric and eccentric occlusion, in retention, in stability, in the condition of the oral tissues bearing the dentures and in their serviceability as judged by the patient and by the dentist.

Dimensional changes in dentures caused by processing and by service could not be correlated with the foregoing decline in important requisites. The causes of the gradual reduction in the retention, stability and serviceability of the dentures were changes in the dimensions of the hard and soft tissues upon which the dentures rest and not on any dimensional changes in the dentures.

There is a considerable scatter among the clinical scores of individual dentures because of differences in the tissue reaction of the wearer. In fact one could say that the relative effectiveness of a properly made denture depends upon how well the tissues of the wearer tolerate the stresses imposed by the denture.

There is also a considerable scatter in the dimensional changes in individual dentures made of the same plastic. Differences in the size and cross-sectional areas which occur among the dentures of individuals account for such differences.

Patient differences will usually prevent the establishment of any rating of different denture bases in clinical testing on small numbers of patients.

Contrary to some clinical opinion, the lower dentures were as stable as the upper dentures and retained their stability with time slightly better than the upper dentures.

The condition of tissues beneath upper and lower dentures of the same set was about the same even though the lower has much less bearing surface and consequently a higher stress per unit of area.

The data show the ineffectiveness of the patients' rating of their dentures and hence the futility of personal testimonials as a basis of assessment of the fit and function of dentures.

Dentures made of an epoxy resin gave poor service because they were porous, stained badly, were not color stable, developed large crevices around the porcelain teeth, many of which came out of the dentures, and had a large and long continued expansion caused by water absorption.

Dentures should be removed at night to enable the tissues to recover from daytime injuries.

Pressure-indicator-paste patterns provide a convenient and reliable method of showing the apposition of dentures and tissues. This apposition is not altered by the very small dimensional changes in the dentures but by a resorption of the tissues supporting the dentures.

Manually executed patterns obtained by pressure-indicating-paste are far more reliable as a guide for making alterations in a denture than are patterns obtained by having the patient simulate chewing.

The findings demonstrate that subjective observations with proper disciplines and controls can furnish reliable and valid information in the clinical evaluation of dentures.

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Table 1. Dimensional Changes In and Serviceability of Clinical Dentures

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Trade Brand, Type of Material and Processing Method | Denture Designation(1) | Observation Period--Months | Molar-To-Molar Change (4) | | | | Flange-To-Flange Change (6) % | Warpage Index (7) | Occlusion | | Retention | Stability | Oral Tissues | Serviceability | | Total Average Scores (17) | | | | |
| | | | Shrinkage during processing % (2) | in Service (3) % | Total Change | | | | Centric | Eccentric | | | | Patient's Opinion | Dentist's Opinion | 1959 | 1960 | 1961 | 1963 | All Years |
| | | | | | Room Temp. % | Mouth (5) Temp. % | | | Average Points (16) | | Average Points (16) | | | Average Points (16) | | | | | | |
| Acralite 88 | (8) A-1-U | 72 | 0.25 | +0.42 | +0.17 | +0.30 | +0.28 | 0.14 | 4.0 | 3.5 | 3.2 | 3.0 | 3.0 | 4.0 | 3.2 | 3.7 | 3.4 | 3.3 | 3.3 | 3.4 |
| Copolymer methacrylate, powder-liquid, cross-linked, self-curing. | (8) A-2-U | 72 | 0.35 | +0.33 | -0.02 | +0.11 | +0.04 | 0.29 | 3.7 | 3.5 | 3.7 | 3.7 | 3.5 | 4.0 | 2.7 | 4.0 | 3.7 | 3.1 | 2.7 | 3.4 |
| | (8) A-3-U | 72 | 0.13 | +0.45 | +0.32 | +0.45 | +0.01 | 0.44 | 2.7 | 3.0 | 2.5 | 3.2 | 2.7 | 3.2 | 2.7 | 2.8 | 3.1 | 2.7 | 2.8 | 2.8 |
| | A-4-U | 39 | 0.15 | +0.23 | +0.08 | +0.21 | +0.17 | 0.06 | 2.3 | 3.0 | 4.0 | 4.0 | 4.0 | 3.6 | 3.3 | 3.7 | 3.4 | 3.3 | - | 3.5 |
| | A-5-L | 39 | 0.29 | +0.24 | -0.05 | +0.08 | +0.25 | 0.01 | 2.3 | 3.0 | 4.0 | 4.0 | 3.6 | 3.6 | 3.3 | 3.7 | 3.4 | 3.1 | - | 3.4 |
| Compression molding (room temperature). | A-6-U | 36 | 0.16 | +0.04 | -0.12 | +0.01 | -0.74 | 0.78 | 2.6 | 2.6 | 2.6 | 2.0 | 2.3 | 3.6 | 2.3 | 2.8 | 2.4 | 2.6 | - | 2.6 |
| | A-7-L | 36 | 0.15 | -0.33 | -0.48 | -0.35 | -0.36 | 0.03 | 2.6 | 2.6 | 3.0 | 3.3 | 3.0 | 3.6 | 2.3 | 3.0 | 3.0 | 2.8 | - | 2.9 |
| | (9) A-8-L | 60 | 0.51 | +0.51 | 0.00 | +0.13 | +0.39 | 0.12 | 2.2 | 2.5 | 2.0 | 2.2 | 2.0 | 3.2 | 2.2 | 2.0 | 2.1 | 2.6 | 2.7 | 2.3 |
| Duraflow | D-3-U | 66 | 0.21 | +0.29 | +0.08 | +0.20 | +0.03 | 0.26 | 3.2 | 2.7 | 3.0 | 3.2 | 2.2 | 4.0 | 2.7 | 3.6 | 2.7 | 3.1 | 2.7 | 3.0 |
| Acrylic resins, powder-liquid cross-linked. | D-4-L | 66 | 0.43 | +0.41 | -0.02 | +0.10 | +0.25 | 0.16 | 3.2 | 2.7 | 2.5 | 3.5 | 2.7 | 4.0 | 2.7 | 3.7 | 2.6 | 3.0 | 3.0 | 3.1 |
| | D-6-U | 36 | 0.21 | +0.10 | -0.11 | +0.01 | +0.02 | 0.08 | 2.3 | 2.3 | 3.0 | 3.0 | 2.3 | 4.0 | 3.0 | 2.8 | 3.1 | 2.6 | - | 2.8 |
| Compression molding (no trial packing). | D-7-L | 36 | 0.19 | +0.11 | -0.08 | +0.04 | +0.18 | 0.07 | 2.3 | 2.3 | 2.7 | 2.7 | 2.3 | 4.0 | 3.0 | 3.1 | 3.3 | 2.7 | - | 3.0 |
| | (8) D-8-U | 60 | 0.42 | +0.19 | -0.23 | -0.11 | +0.02 | 0.17 | 2.7 | 2.7 | 3.2 | 2.0 | 1.7 | 4.0 | 2.5 | 2.6 | 3.0 | 2.7 | 2.6 | 2.7 |
| Epoxolon | (8) E-1-U | 41 | 0.15 | +1.71 | +1.56 | +1.66 | +0.98 | 0.73 | 3.0 | 2.3 | 3.0 | 3.3 | 3.3 | 3.6 | 2.0 | 3.4 | 2.7 | 2.1 | - | 2.7 |
| | E-2-U | (10) 41 | 0.08 | +1.53 | +1.45 | +1.55 | +1.12 | 0.41 | 2.5 | 2.0 | 3.5 | 3.7 | 3.5 | 3.0 | 1.7 | 3.3 | 2.7 | 2.6 | 2.8 | 2.8 |
| Epoxy resin, slurry-liquid. | E-3-L | (10) 41 | 0.23 | +1.87 | +1.64 | +1.74 | +0.92 | 0.95 | 2.5 | 2.0 | 2.0 | 2.0 | 2.5 | 3.0 | 1.5 | 2.7 | 2.0 | 1.7 | 2.4 | 2.2 |
| | E-4-U | 65 | 0.13 | +1.23 | +1.10 | +1.20 | +1.14 | 0.09 | 3.2 | 3.0 | 1.7 | 1.7 | 2.5 | 3.2 | 2.0 | 2.7 | 3.3 | 2.3 | 1.7 | 2.5 |
| | E-5-L | 65 | 0.38 | +1.39 | +1.01 | +1.11 | +1.26 | 0.13 | 3.2 | 3.0 | 2.7 | 2.7 | 2.7 | 3.2 | 2.0 | 3.1 | 3.3 | 2.7 | 1.8 | 2.7 |
| Pouring (dry-low heat cure). | E-6-U | 61 | 0.06 | +1.44 | +1.38 | +1.48 | +1.00 | 0.44 | 2.0 | 2.5 | 2.0 | 1.5 | 1.2 | 3.5 | 1.0 | 2.0 | 2.0 | 2.1 | 1.7 | 2.0 |
| | E-7-L | 61 | 0.21 | +1.27 | +1.06 | +1.16 | +1.13 | 0.14 | 2.0 | 2.5 | 1.7 | 1.7 | 3.0 | 3.5 | 1.0 | 2.4 | 1.8 | 2.1 | 1.8 | 2.0 |

Table 1. (con't)

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Hydro-Cast | H-1-U | 68 | 0.51 | +0.27 | -0.24 | -0.12 | +0.16 | 0.11 | 2.7 | 3.0 | 3.2 | 2.7 | 3.0 | 4.0 | 2.7 | 3.4 | 3.1 | 3.1 | 2.6 | 3.0 |
|  | H-2-L | 68 | 0.50 | +0.56 | +0.06 | +0.18 | +0.36 | 0.20 | 2.7 | 3.0 | 3.2 | 2.7 | 2.7 | 4.0 | 2.7 | 3.3 | 3.3 | 3.1 | 2.4 | 3.0 |
| Copolymer of acrylic resins, powder-liquid. | H-3-U | 64 | 0.44 | +0.13 | -0.31 | -0.19 | 0.00 | 0.13 | 3.5 | 3.0 | 3.5 | 4.0 | 3.5 | 4.0 | 3.5 | 3.8 | 3.7 | 3.8 | 3.0 | 3.6 |
|  | H-4-L | 64 | 0.52 | -0.04 | -0.56 | -0.44 | -0.00 | 0.04 | 3.5 | 3.0 | 4.0 | 4.0 | 3.5 | 4.0 | 3.5 | 4.0 | 3.6 | 3.8 | 3.3 | 3.7 |
| Directional hydraulic compression (Hydro-Cast technic). | H-6-U | 63 | 0.54 | +0.39 | -0.15 | -0.03 | -0.02 | 0.41 | 2.7 | 2.2 | 3.0 | 2.0 | 3.7 | 4.0 | 2.7 | 3.6 | 3.0 | 2.8 | 2.3 | 2.9 |
|  | H-7-L | 63 | 0.67 | +0.56 | -0.11 | +0.01 | +0.40 | 0.16 | 2.7 | 2.2 | 2.0 | 2.7 | 3.2 | 4.0 | 2.7 | 3.6 | 3.0 | 2.4 | 2.3 | 2.8 |
|  | (8)<br>H-8-U | 36 | 0.28 | +0.02 | -0.26 | -0.14 | +0.09 | 0.07 | 2.3 | 2.0 | 2.3 | 1.3 | 3.3 | 3.0 | 2.3 | 2.6 | 2.1 | 2.4 | - | 2.4 |
|  | H-9-U | 61 | 0.60 | +0.21 | -0.39 | -0.27 | +0.08 | 0.13 | 2.5 | 1.7 | 3.5 | 3.7 | 2.7 | 3.2 | 2.7 | 2.6 | 3.0 | 3.4 | 2.6 | 2.9 |
|  | H-10-L | 61 | 0.63 | +0.20 | -0.43 | -0.31 | +0.24 | 0.04 | 2.5 | 1.7 | 4.0 | 3.7 | 2.0 | 3.2 | 2.7 | 2.6 | 3.0 | 3.3 | 2.6 | 2.9 |
| Jectron | J-1-U | 66 | 0.37 | +0.07 | -0.30 | -0.21 | -0.13 | 0.20 | 2.2 | 2.2 | 3.2 | 3.0 | 2.7 | 3.7 | 2.2 | 3.3 | 3.0 | 2.3 | 2.6 | 2.8 |
|  | J-2-L | 66 | 0.57 | 0.00 | -0.57 | -0.48 | +0.01 | 0.01 | 2.2 | 2.2 | 3.2 | 2.0 | 2.0 | 3.7 | 2.2 | 3.1 | 2.7 | 2.1 | 2.1 | 2.5 |
| Polystyrene, precured bar. | J-3-U | 66 | 0.34 | 0.00 | -0.34 | -0.25 | -0.10 | 0.10 | 3.0 | 2.5 | 2.0 | 1.7 | 2.5 | 3.5 | 2.5 | 3.6 | 2.8 | 1.6 | 2.1 | 2.5 |
|  | J-4-L | 66 | 0.35 | +0.10 | -0.25 | -0.16 | +0.02 | 0.08 | 3.0 | 2.5 | 2.7 | 2.5 | 2.5 | 3.5 | 2.5 | 3.7 | 3.0 | 2.1 | 2.1 | 2.7 |
|  | J-5-U | 64 | 0.32 | -0.09 | -0.41 | -0.32 | -0.06 | 0.03 | 3.5 | 3.0 | 3.7 | 2.5 | 2.7 | 3.7 | 2.7 | 3.4 | 3.1 | 3.0 | 3.0 | 3.1 |
| "Transinjection molding". | J-6-L | 64 | 0.43 | -0.01 | -0.44 | -0.35 | +0.01 | 0.02 | 3.5 | 3.0 | 3.5 | 2.7 | 3.0 | 3.7 | 2.7 | 3.3 | 3.6 | 3.0 | 2.8 | 3.2 |
|  | (8)(14)<br>J-8-U | 64 | 0.26 | +0.55 | +0.29 | +0.38 | -0.21 | 0.76 | 2.7 | 2.7 | 2.2 | 1.0 | 1.0 | 3.0 | 1.2 | 1.8 | 2.4 | 2.1 | 2.0 | 2.1 |
| Luxene 44 | (8)<br>L-1-U | 68 | 0.39 | -0.10 | -0.49 | -0.39 | -0.20 | 0.10 | 2.7 | 2.0 | 2.5 | 2.0 | 1.7 | 3.5 | 1.5 | 2.7 | 2.7 | 1.8 | 1.8 | 2.2 |
|  | L-2-U | 68 | 0.20 | 0.00 | -0.20 | -0.10 | -0.23 | 0.23 | 2.2 | 2.5 | 3.2 | 2.7 | 2.0 | 3.7 | 2.2 | 2.4 | 2.7 | 2.7 | 2.8 | 2.6 |
| Vinyl-acrylic copolymer, gel. | L-3-L | 68 | 0.46 | +0.07 | -0.39 | -0.29 | -0.14 | 0.21 | 2.2 | 2.5 | 2.2 | 1.7 | 2.7 | 3.7 | 1.7 | 2.7 | 2.8 | 2.1 | 2.0 | 2.4 |
|  | L-6-U | 62 | 0.20 | -0.19 | -0.39 | -0.29 | -0.13 | 0.06 | 2.7 | 2.2 | 3.2 | 3.7 | 3.7 | 4.0 | 3.2 | 3.4 | 3.6 | 3.0 | 3.1 | 3.3 |
| Injection-moist heat ("Pressure Cast" process). | L-7-L | (11)<br>62 | 0.34 | -0.11 | -0.45 | -0.35 | -0.13 | 0.02 | 2.7 | 2.2 | 3.0 | 3.0 | 2.5 | 4.0 | 3.0 | 3.3 | 3.3 | 2.7 | 2.4 | 2.9 |
|  | (8)<br>L-8-U | 63 | 0.40 | -0.21 | -0.61 | -0.51 | -0.22 | 0.01 | 2.0 | 2.2 | 2.0 | 1.5 | 1.5 | 2.2 | 1.5 | 2.3 | 2.3 | 1.6 | 1.3 | 1.9 |
| Miracle 50 and Mystic 100 | Mi-1-U | 66 | 0.33 | +0.11 | -0.22 | -0.15 | -0.13 | 0.24 | 2.2 | 2.5 | 3.2 | 3.5 | 3.7 | 2.5 | 2.5 | 3.4 | 3.1 | 2.7 | 2.3 | 2.9 |
|  | Mi-2-L | 66 | 0.42 | -0.75 | -1.17 | -1.10 | -0.58 | 0.17 | 2.2 | 2.5 | 1.2 | 1.2 | 2.5 | 2.5 | 2.0 | 2.4 | 2.6 | 1.6 | 1.6 | 2.0 |

Table 1. (con't)

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Acrylic resin powder-liquid, cross-linked, containing 14% and 21% glass fibers by weight respectively, Compression molding. | My-1-U | (12) 66 | 0.35 | -0.08 | -0.43 | -0.36 | -0.20 | 0.12 | 2.5 | 2.2 | 1.2 | 1.5 | 1.0 | 1.7 | 1.0 | 2.3 | 1.6 | 1.3 | 1.3 | 1.6 |
| | My-2-L | (12) 66 | 0.63 | -0.12 | -0.75 | -0.68 | -0.01 | 0.11 | 2.5 | 2.2 | 1.2 | 1.2 | 1.2 | 2.0 | 1.0 | 2.3 | 1.6 | 1.3 | 1.3 | 1.6 |
| Tilon | T-3-U | 65 | 0.31 | +0.20 | -0.11 | +0.01 | -0.08 | 0.28 | 1.7 | 1.7 | 2.5 | 2.0 | 2.7 | 4.0 | 2.5 | 2.6 | 2.4 | 2.3 | 2.6 | 2.7 |
| | T-4-L | (13) 36 | 0.34 | +0.21 | -0.13 | -0.01 | +0.10 | 0.11 | 2.0 | 1.6 | 3.0 | 3.3 | 2.3 | 4.0 | 2.3 | 3.0 | 2.7 | 2.3 | - | 2.7 |
| Copolymers of methyl and ethyl methacrylate uncross-linked, gel. | T-5-U | 62 | 0.70 | +0.35 | -0.35 | -0.23 | -0.02 | 0.37 | 2.5 | 2.7 | 3.2 | 3.7 | 3.7 | 4.0 | 2.5 | 3.3 | 3.3 | 3.4 | 2.7 | 3.2 |
| | T-6-L | 62 | 0.40 | +0.40 | 0.00 | +0.12 | +0.25 | 0.15 | 2.5 | 2.7 | 4.0 | 4.0 | 3.0 | 4.0 | 2.5 | 3.3 | 3.3 | 3.4 | 3.0 | 3.2 |
| Injection-dry heat ("Unidirectional Curing"). | T-7-U | 62 | 0.83 | +0.62 | -0.21 | -0.09 | -0.11 | 0.73 | 2.5 | 2.7 | 3.0 | 2.0 | 2.2 | 3.7 | 2.5 | 3.0 | 3.0 | 2.7 | 2.0 | 2.7 |
| | T-8-L | 62 | 0.97 | +0.36 | -0.61 | -0.49 | +0.26 | 0.10 | 2.5 | 2.7 | 3.0 | 2.7 | 1.7 | 3.7 | 2.2 | 3.0 | 3.0 | 2.8 | 1.8 | 2.6 |
| | (9) T-9-U | 60 | 0.51 | +0.30 | -0.21 | -0.09 | -0.22 | 0.52 | 2.5 | 2.5 | 4.0 | 4.0 | 2.0 | 3.2 | 3.2 | 3.3 | 2.7 | 3.0 | 3.0 | 3.0 |
| Vernonite | (8) V-1-U | 48 | 0.33 | -0.32 | -0.65 | -0.51 | -0.15 | 0.17 | 3.0 | 1.6 | 2.6 | 2.3 | 2.6 | 3.3 | 2.3 | 2.6 | 3.1 | 1.8 | - | 2.5 |
| | V-2-U | 66 | 0.39 | +0.06 | -0.33 | -0.19 | -0.19 | 0.25 | 3.5 | 3.2 | 3.5 | 2.7 | 3.5 | 4.0 | 3.2 | 3.8 | 3.3 | 3.3 | 3.1 | 3.4 |
| | V-3-L | 66 | 0.63 | +0.26 | -0.37 | -0.23 | +0.01 | 0.25 | 3.5 | 3.2 | 2.7 | 4.0 | 2.5 | 4.0 | 3.2 | 3.7 | 3.1 | 3.3 | 3.1 | 3.3 |
| Acrylic resin, powder-liquid, uncross-linked. | V-4-U | 65 | 0.21 | +0.20 | -0.01 | +0.13 | -0.09 | 0.29 | 3.2 | 3.5 | 3.2 | 2.0 | 3.2 | 4.0 | 3.0 | 3.4 | 3.1 | 3.1 | 3.0 | 3.2 |
| | V-5-L | 65 | 0.40 | +0.37 | -0.03 | +0.11 | +0.27 | 0.10 | 3.2 | 3.5 | 2.5 | 2.5 | 2.7 | 4.0 | 3.0 | 3.3 | 3.3 | 3.0 | 2.7 | 3.1 |
| Compression molding. | V-6-U | 63 | 0.28 | +0.12 | -0.16 | -0.02 | +0.08 | 0.04 | 1.5 | 1.2 | 2.5 | 1.7 | 2.5 | 1.7 | 1.2 | 2.0 | 1.7 | 2.0 | 1.4 | 1.8 |
| | V-7-L | 63 | 0.48 | +0.15 | -0.33 | -0.19 | +0.19 | 0.04 | 1.5 | 1.2 | 1.7 | 2.2 | 2.0 | 1.7 | 1.2 | 2.0 | 2.1 | 1.6 | 1.1 | 1.7 |
| | (8)(15) V-8-U | 62 | 0.26 | +0.29 | +0.03 | +0.17 | -0.03 | 0.32 | 2.7 | 2.5 | 2.7 | 3.0 | 2.2 | 3.2 | 2.5 | 4.0 | 2.8 | 1.7 | 2.3 | 2.7 |
| Vulcanite Hygienic Dental Rubber Vulcanization | Vul-1-U | 62 | 0.22 | -0.55 | -0.77 | -0.66 | -0.16 | 0.39 | 2.7 | 2.5 | 2.7 | 2.2 | 2.2 | 3.0 | 2.5 | 3.1 | 2.4 | 2.4 | 2.1 | 2.5 |
| | Vul-2-L | 62 | 0.39 | -0.46 | -0.85 | -0.74 | -0.30 | 0.16 | 2.7 | 2.5 | 2.7 | 3.0 | 2.7 | 3.0 | 2.5 | 3.6 | 2.6 | 2.6 | 2.3 | 2.8 |
| Average | | 60.4 | 0.37 | +0.34 | -0.22 | -0.02 | +0.11 | 0.22 | 2.7 | 2.5 | 2.7 | 2.7 | 2.6 | 3.5 | 2.4 | 3.1 | 2.8 | 2.6 | 2.4 | 2.7 |

Table 1. (con't)

(1) The initial letter(s) indicate(s) the denture base material as shown in Column 1. The numeral identifies each denture in each base material. The letters U and L designate upper and lower, respectively, and the sets of upper and lower dentures are bracketed.

(2) Molar-to-molar change of cured denture in flask through polishing and one day in water.

(3) The amount of gain (+) or loss (-) in molar-to-molar distance during clinical service (Column 3).

(4) The total molar-to-molar dimensional change from the time the denture was unflasked to the occlusal surfaces through the measuring period (Column 3).

(5) Value in Column 6 corrected for thermal expansion of the denture from room temperature (22°C.) at which it was measured to mouth temperature (37°C.).

(6) The amount of gain (+) or loss (-) in flange-to-flange distance during clinical service (Column 3).

(7) Difference between molar-to-molar change (Column 5) and flange-to-flange change (Column 8) during clinical service (Column 3).

(8) Upper denture opposed in part by lower natural teeth.

(9) A-8-L and T-9-U comprise the only set of dentures made with two different bases.

(10) After 41 months these Epoxolon dentures were replaced.

(11) Worn 50 months and kept in water subsequently.

(12) Worn 6 months and kept in water subsequently except when worn by the patient when the replacement dentures (A-8-L and T-9-U) were being measured.

(13) Denture lost during hospitalization.

(14) Denture was inserted in April 1958 and has fractured four times in service, namely in August 1960, October 1961, January 1962 and April 1962.

(15) Denture was inserted in June 1958 and was fractured in June 1960 when the patient was struck in the face.

(16) During the period of observation (Column 3).

(17) Average scores of ratings for centric occlusion, eccentric occlusion, retention, stability, condition of oral supporting tissues and serviceability.

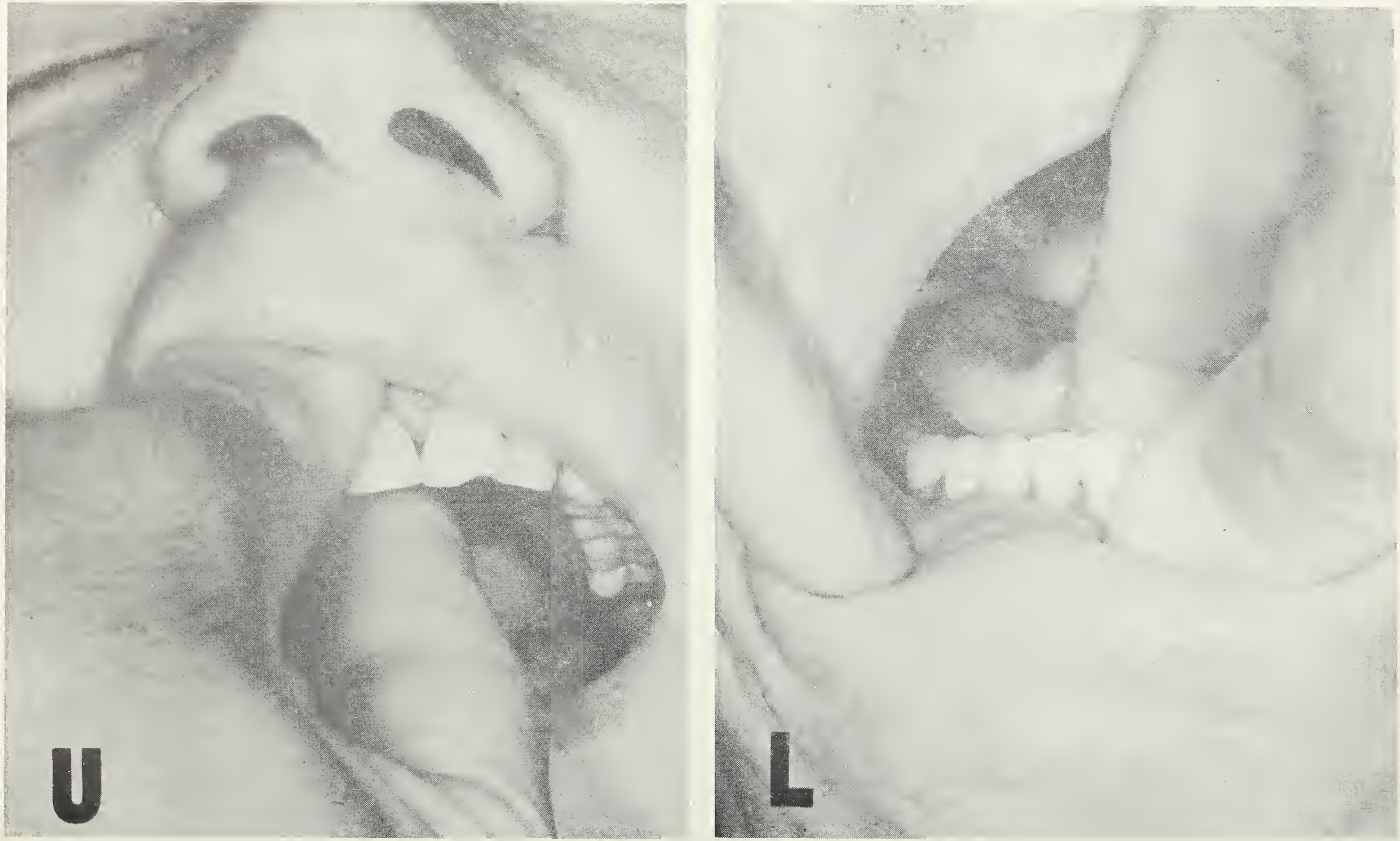

Figure 1. Position of the index finger and thumb when applying force to estimate resistance to dislodgement of upper denture, U, and of lower denture, L.

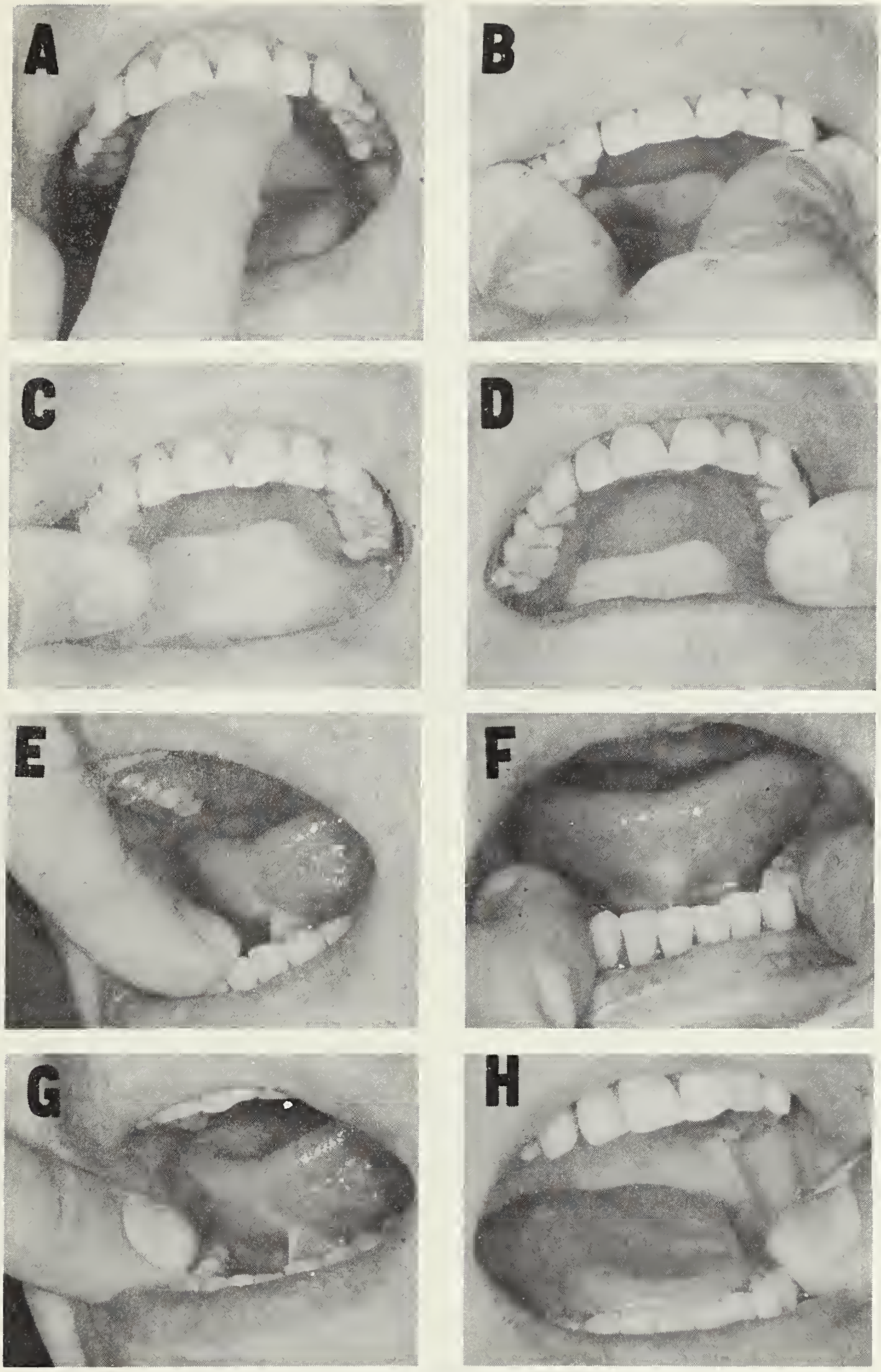

Figure 2.

Position of fingers and thumb when applying force to estimate the stability (resistance to movement) of dentures. A, direct force applied in center of upper denture; B, application of rotary forces to upper denture; C, application of direct force on right side and D, on left side of upper denture; E, direct force applied on incisors of lower denture; F, application of rotary forces to lower denture; G, application of direct force on right side; and H, on left side of lower denture.

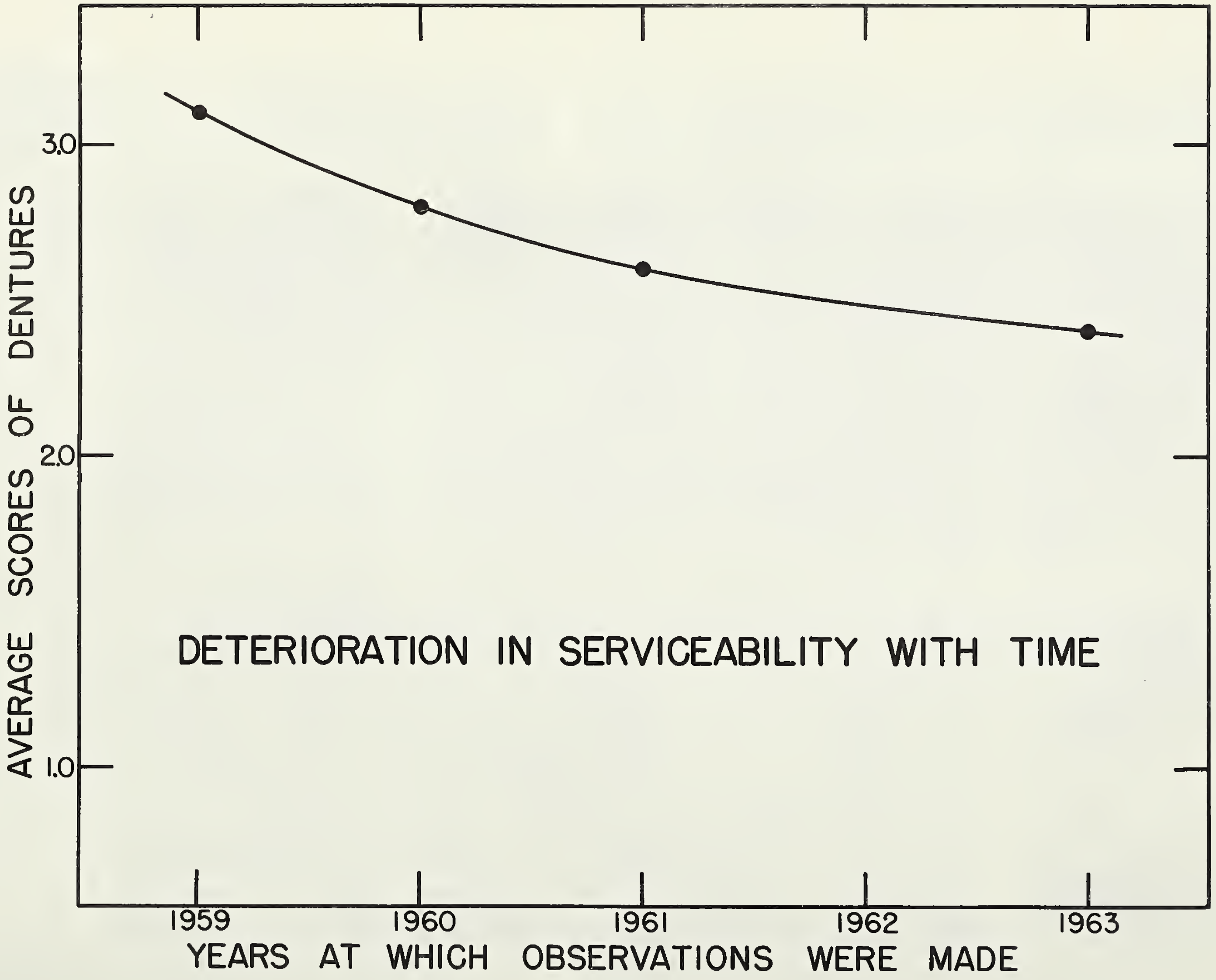

Figure 3. The relation between the average value of the scores used in rating each denture and the time the observations were made. The higher the score, the better is the rating.

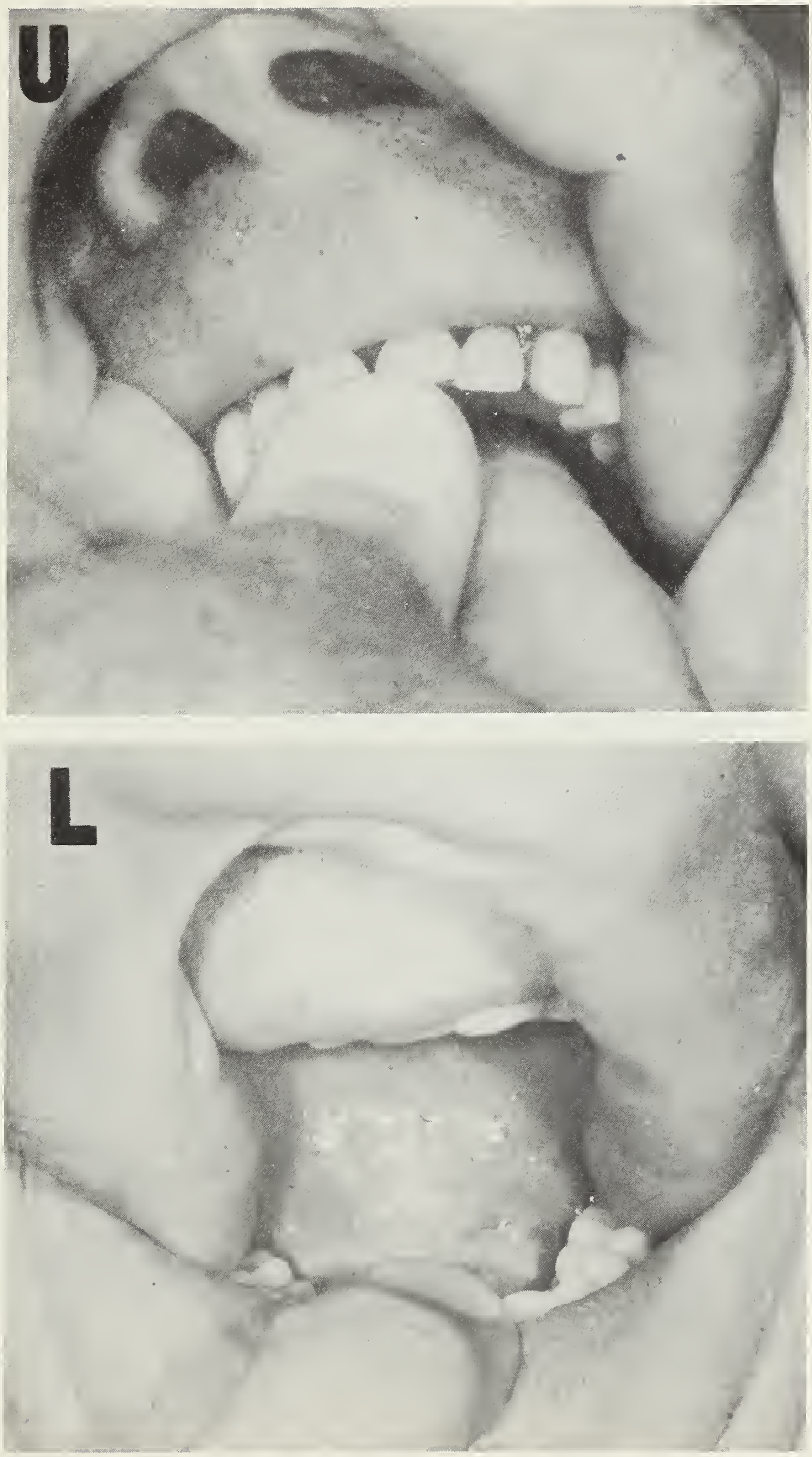

Figure 4.

Position of fingers and thumb on an upper denture, U, and on a lower denture, L, when applying force in obtaining manually executed patterns with indicator paste.

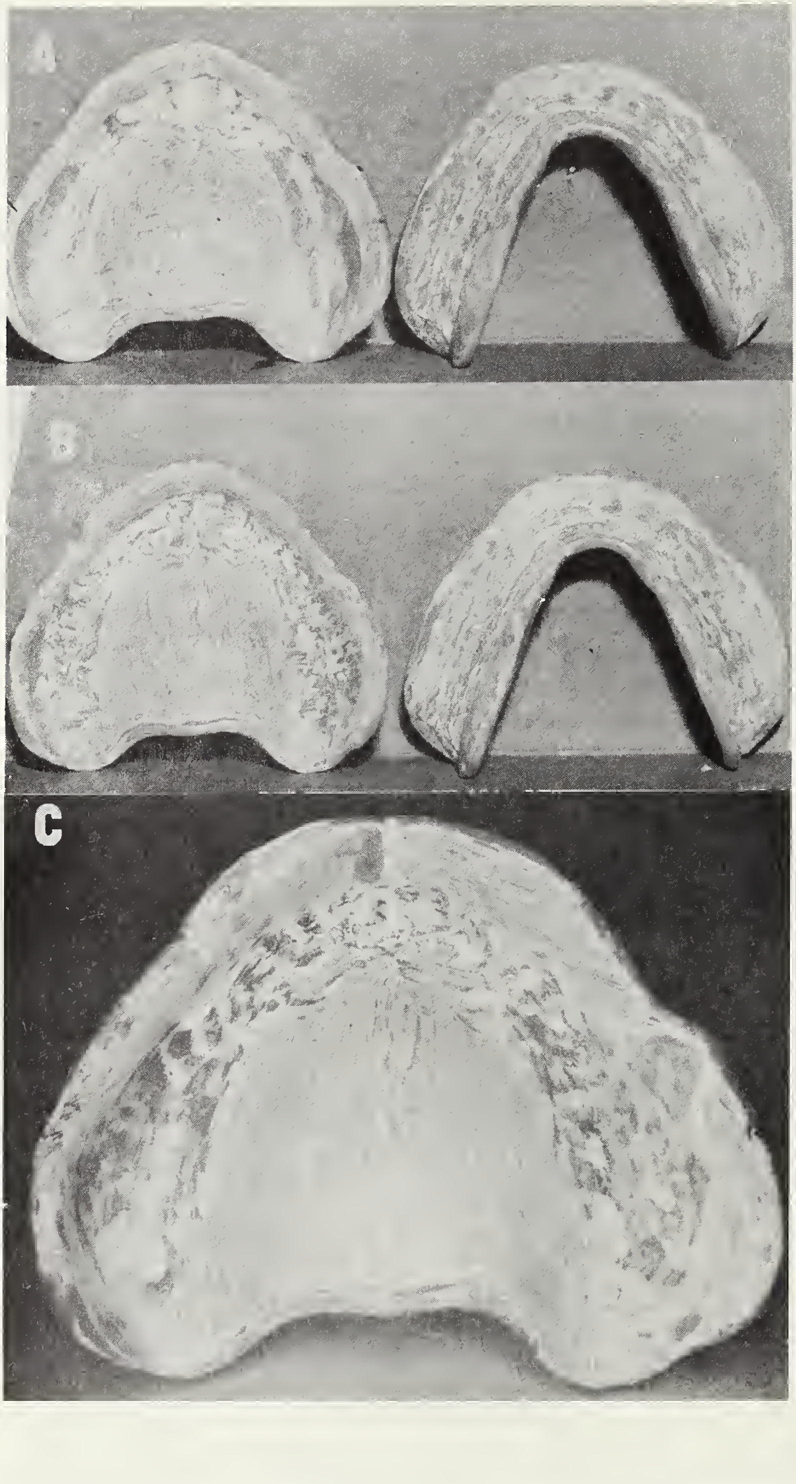

Figure 5.

Improvement in the apposition of tissues to dentures caused by the selective grinding of areas of hard contact as shown by pressure-indicating-paste patterns. A, patterns made on August 30, 1958, after dentures had been worn for six months. B, patterns made on August 30, 1958, but after relieving areas of contact in four successive patterns, C, patterns made five years later on August 27, 1963, of the upper centure shows many more areas of hard contact which indicate a deterioration in the apposition of the tissues.

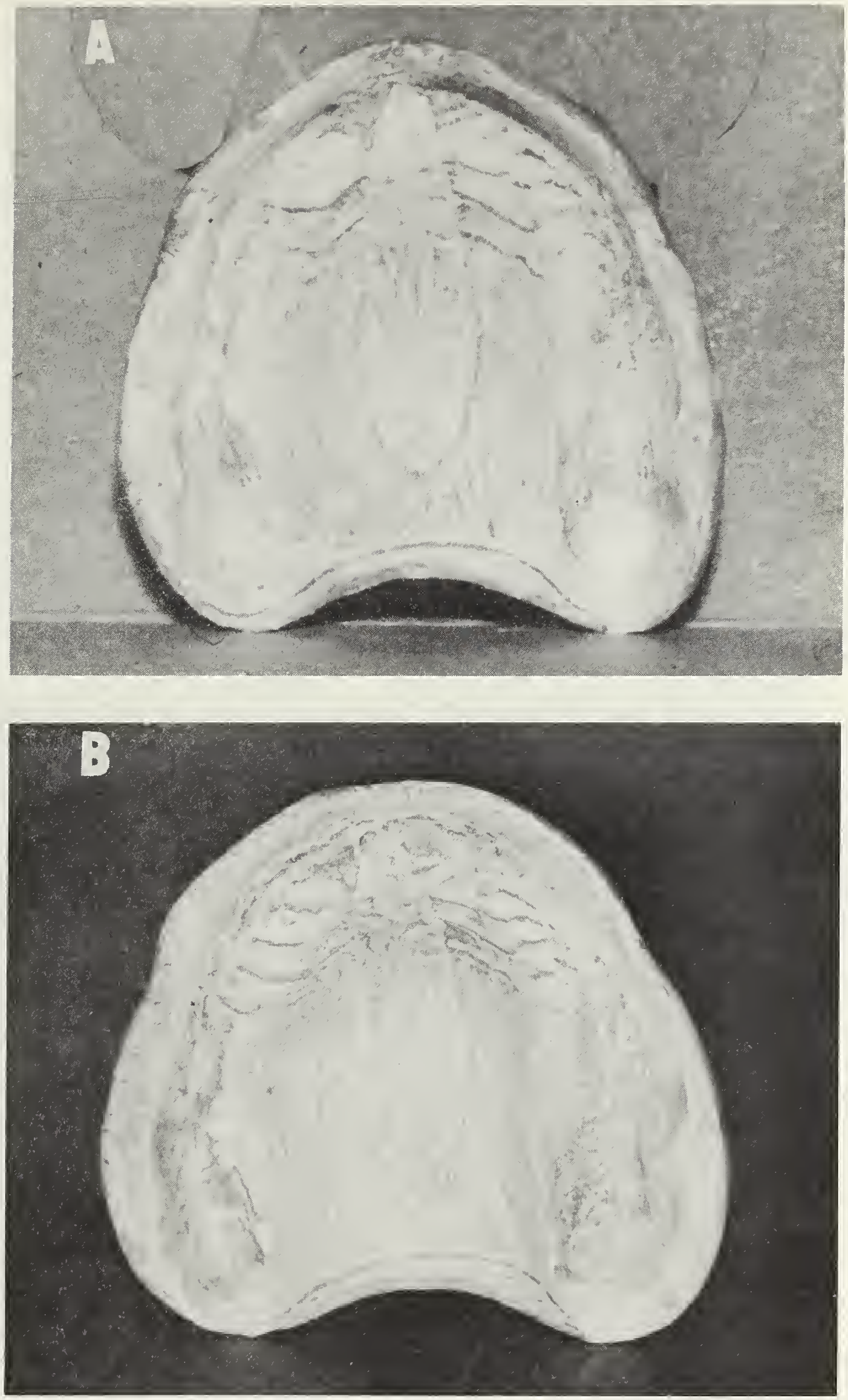

Figure 6.

Pressure-indicator-paste patterns of an upper denture of Epoxolon. Pattern A was taken on July 23, 1958, when the new denture was given to the patient. Pattern B was taken three years later on July 12, 1961. During this three year period the molar-to-molar expansion was 1.55 percent. The tissues accomodated to this large but gradual change as shown by the similarity of patterns A and B.

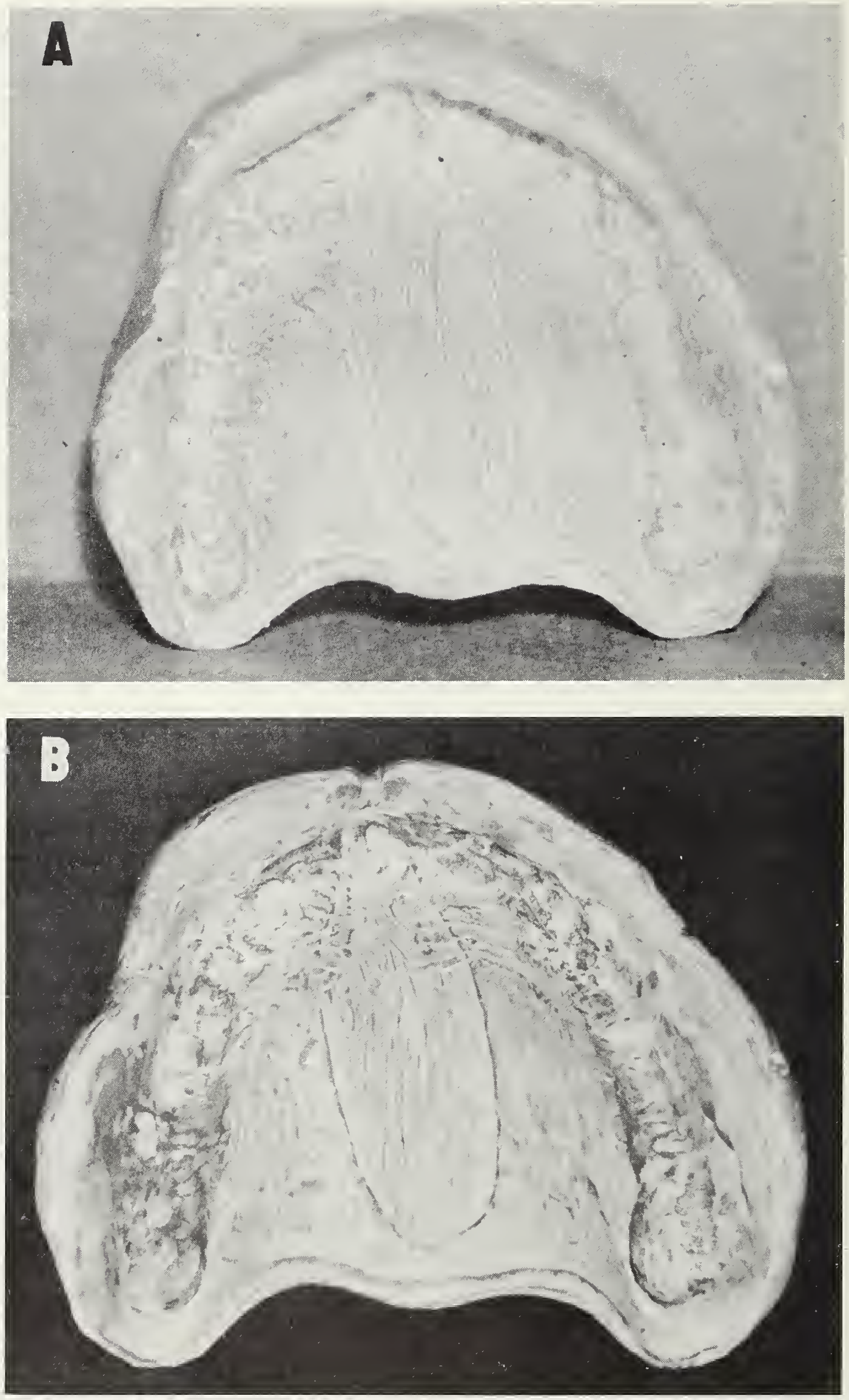

Figure 7.

Pressure-indicator-paste patterns of an upper Hydro-Cast denture. Pattern A was taken July 28, 1958, at the time the denture was delivered to the patient. Pattern B, taken about three years later (July 31, 1961, shows large and heavy contact areas not present in Pattern A. The denture remained stable in dimension during these three years as the molar-to- molar change was only -0.14 percent.

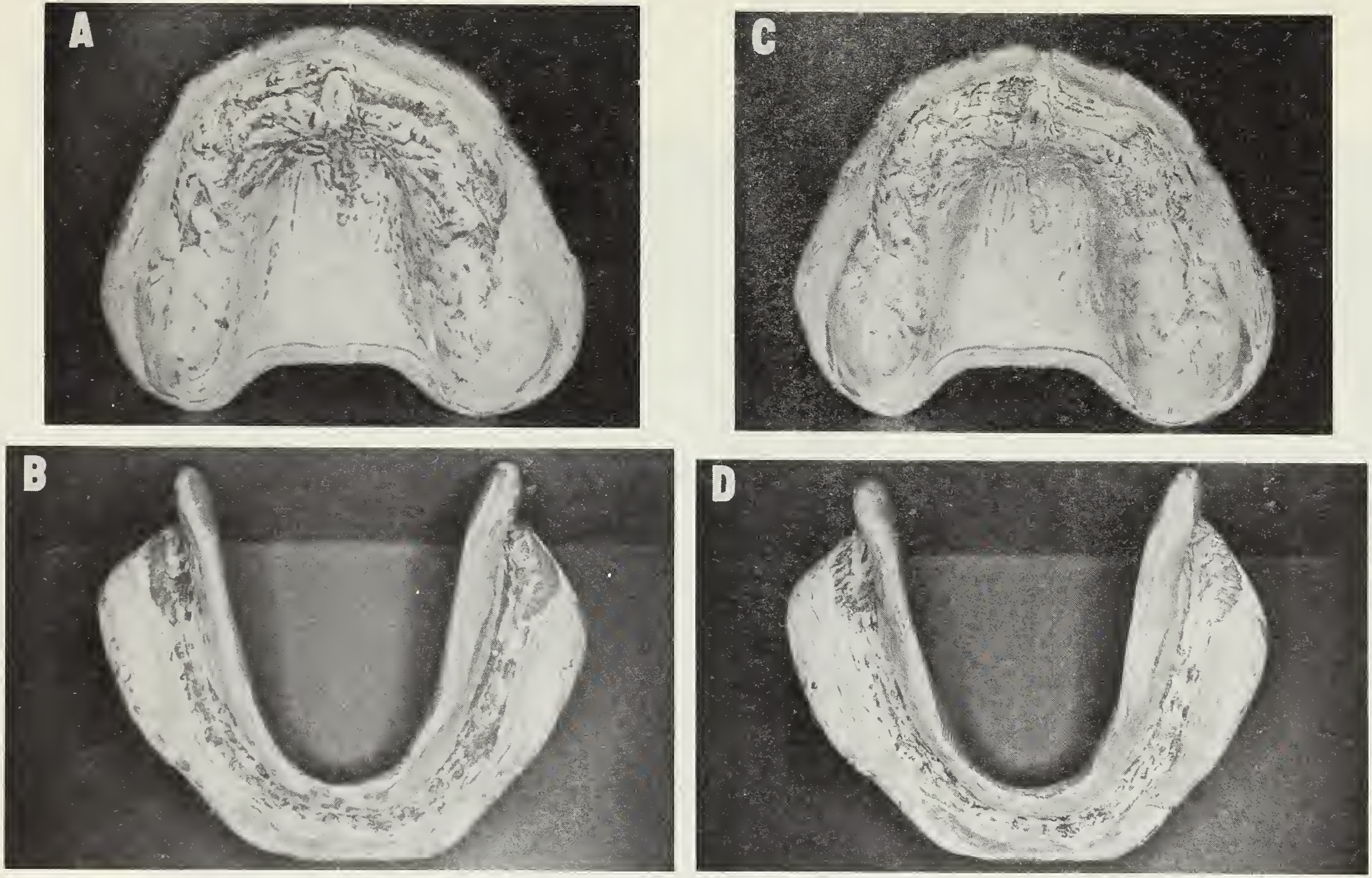

Figure 8. Pressure-indicator-paste patterns of a set of Jectron dentures. Patterns A and B were obtained by the patient simulating chewing for 40 seconds. Patterns C and D were manually executed by the dentist. A pipe smoking habit caused this patient to have premature contacts on the right side which are reflected in patterns A and B. Patterns C and D show good apposition of the tissues.

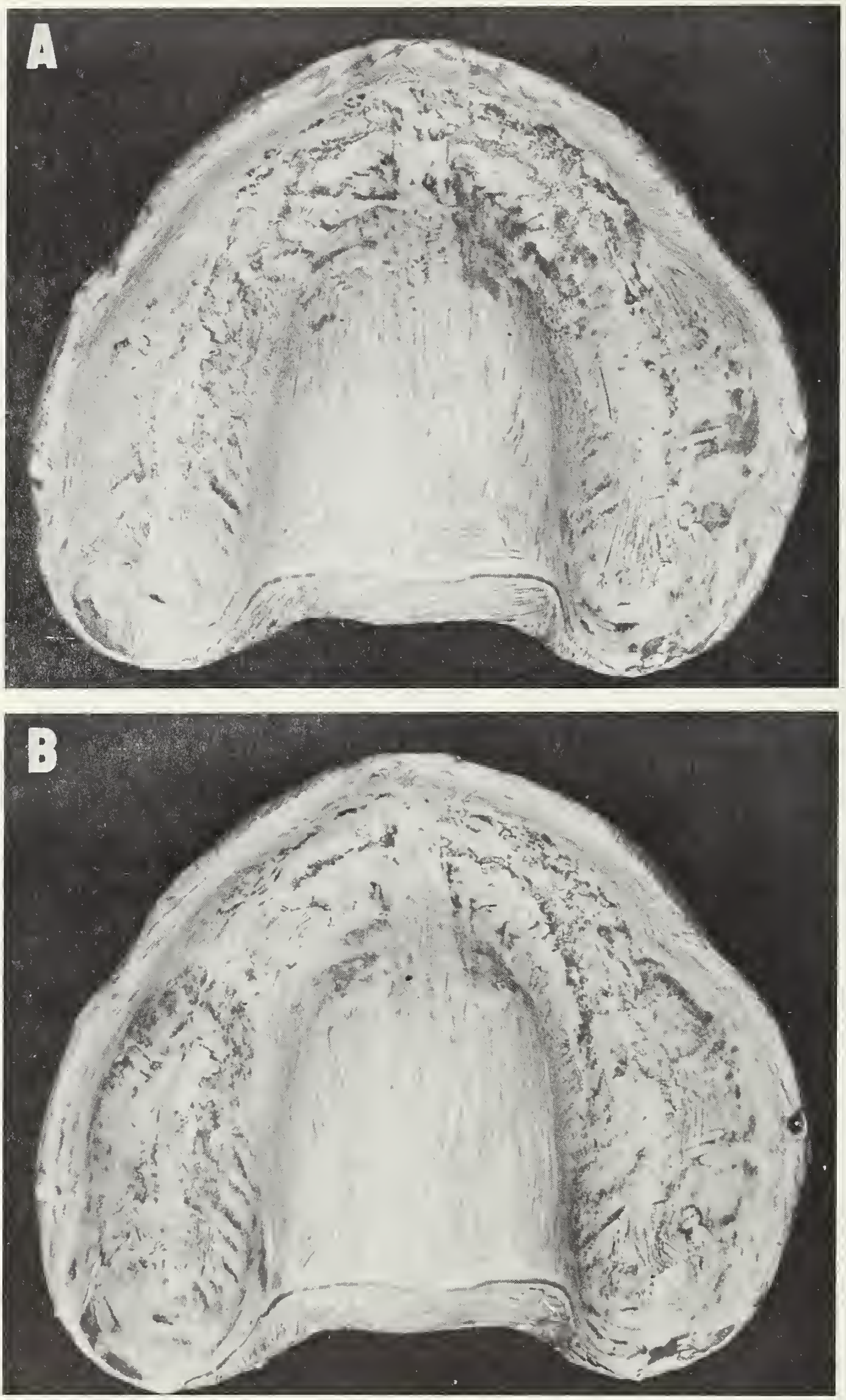

Figure 9.

Pressure-indicator-paste patterns of an upper Luxene 44 denture that occluded only with nine natural lower teeth. Pattern A was made with the patient simulating chewing and shows areas of hard contact in the anterior one-third of the hard palate. Pattern B, which was executed manually, shows a comparatively even distribution of contact of the denture with the tissues.

**NATIONAL BUREAU OF STANDARDS REPORT**

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# Progress Report
# on
# Blood: Bone Equilibrium in Calcium Homeostasis

By

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# Blood: Bone Equilibrium in Calcium Homeostasis

## Abstract

Data on the solubility of bone *in vitro* are plotted so that the slope of the line through the experimental points yields the Ca/P ratio of the equilibrating salt. The slopes of the lines for child and calf bone corresponded best to the ratio 8/6, that of octacalcium phosphate, $Ca_8H_2(PO_4)_6 \cdot 5H_2O$; the slope for adult bone approximated that of hydroxyapatite, $Ca_{10}(OH)(PO_4)_6$.

This is the first direct evidence that octacalcium phosphate is present in bone. Its presence in "young" bone, as contrasted to "adult" bone, is taken as further evidence for the participation of octacalcium phosphate as a precursor in the formation of the apatitic crystallites *in vivo*. The presence of octacalcium phosphate in bone introduces a new concept to be considered in relation to the chemistry of growth, healing, and diseases of bone, and it helps to explain factors that control composition of serum.

The nature of the dynamic equilibrium between the mineral component of human bone and its ions in solution in tissue fluid is unknown. A few years ago the theory advanced by MacLean and Urist (1) was generally accepted, and experimental studies on the mode of action of parathyroid hormone (e.g. Talmage et al (2), Copp (3)) were designed in terms of it. Their hypothesis held that normal tissue fluids were 'supersaturated' with respect to whole bone; that the bone mineral maintained only 'hypoparathyroid levels' of calcium and phosphorus by physical processes and that a parathyroid controlled 'vital activity' caused destruction of whole bone, release of mineral and increase in the concentration of calcium in the tissue fluids. Maintenance of a calcium concentration gradient therefore not only required continual secretion of parathormone but also continual osteoclastic resorption of whole bone.

In 1957, Nordin (4) suggested that the situation be re-examined in terms of solubility theory and showed that calf bone powder would maintain reproducible levels of Ca and P *in vitro*. Later, MacGregor and Nordin extended the work and showed that in both trihydroxymethylaminomethane: HCl (5) and carbonic acid: potassium bicarbonate buffer systems (6) at ion strengths of 0.15 and at 20°C, human dead bone powder would maintain the levels of Ca and P found in normal tissue fluids if the pH at the solid: solution interface was about 7.0 to 7.1 (6). This pH is intermediate between the intracellular pH of 6.8 and the extracellular pH of 7.4, and the concept of a pH gradient across the blood:bone barrier is not unattractive.

The equilibration studies showed that the ion-concentration product, $[Ca^{++}]^3\ [PO_4^{\equiv}]^2$, expressed as a negative logarithm (pK) was relatively constant over the pH range 6.4 to 7.6 and ranged from 26.0 to 26.8 and was not related to pH. On the other hand, the ion-concentration product $[Ca^{++}][HPO_4^{=}]$, correlated highly with pH (5). On this evidence, MacGregor and Nordin postulated that, on purely empirical grounds, the cubed-squared product best represented the physico-chemical relation between normal human bone and its ions in 'equilibrium' with it. It was pointed out that no theoretical significance could be attached to the tricalcium phosphate product on this basis and that the authors did not necessarily consider that this salt was the relevant solid phase of 'the exchangeable calcium pool'.

The chemical nature of the micro-crystals in bone and the physical properties of its surfaces have been the subject of discussion for many years. The conventional view (7) is that hydroxyapatite (HA) is the relevant compound. The basic model is modified in detail by internal lattice faults with entrapped contaminants, and the free surfaces are believed to have suffered heteroionic exhange with ions such as $Sr^{++}$, $Mg^{++}$, $Na^+$, $P^-$, $CO_3^{=}$, and citrate. In consequence, quantitative analysis of whole bone has yielded no definitive data.

In view of the possibility that one or more sparingly soluble calcium phosphates $\{Ca_5OH(PO_4)_3$, $Ca_8H_2(PO_4)_6 \cdot 5H_2O$, $\beta\text{-}Ca_3(PO_4)_2$, $CaHPO_4$, $CaHPO_4 \cdot 2H_2O\}$ could be present in bone, assumptions regarding the stoichiometry of the equilibrium between the solid phase and the aqueous solution are undesirable. It is easily shown (8,9,10) that solubility data can be treated in a way so that such assumptions become unnecessary. The formula for any calcium phosphate may be written in generalised form as $Ca_xH_y(PO_4)\cdot zH_2O$, where $2x + y = 3$ and $y$ may have negative values (e.g. for HA, $x = 5/3$ $y = -1/3$ and $z = 1/3$). The condition for equilibrium between an aqueous solution and the generalised salt required that there be no change in free energy for the reaction:

$$x\,Ca(OH)_2 + H_3PO_4 + (z - 2x)\,H_2O = Ca_xH_y(PO_4)\cdot zH_2O$$

That is, the sum of the chemical potentials of the components given on the left side of the equation is equal to the standard free energy of formation of the salt on the right-hand side.

$$x\,\mu_{Ca(OH)_2} + \mu_{H_3(PO_4)} + (z - 2x)\,\mu_{H_2O} = \Delta F^\circ_{xz}$$

If we then assume that the chemical potential of the water is essentially constant for dilute solutions and that the chemical potentials of $Ca(OH)_2$ and $H_3PO_4$ may be expressed as usual in terms of their ionic activities, then:

$$\mu_{Ca(OH)_2} = \mu^\circ_{Ca(OH)_2} + RT\ \ln\ (Ca^{++})\ (OH^-)^2$$

$$\mu_{H_3PO_4} = \mu^\circ_{H_3PO_4} + RT\ \ln\ (H^+)^3(PO_4^{\equiv})$$

and we can derive the result:

$$\log\ (Ca^{++})\ (OH^-)^2 = -\frac{1}{x}\log\ (H^+)^3(PO_4^{\equiv}) + K_{xz}$$

where the parentheses represent ionic activities and $K_{xz}$ is a constant specific for the applicable salt.

Therefore if negative log $(Ca^{++})\ (OH^-)^2$ is plotted against negative log $(H^+)^3(PO_4^{\equiv})$ for a series of solutions in equilibrium with the above salt, the points should fall on a straight line with a slope of $-\frac{1}{x}$ where x is the Ca/P molar ratio in the salt. In other words, the slope is dictated by the stoichiometry of the reaction. The position of the line is determined by the value of $K_{xz}$ which cannot be calculated without assumptions concerning the appropriate activity coefficients. However, in a series of experiments at constant ionic strength it is usually assumed that the activity coefficients are also constant, and therefore the slope of the line is valid even though its position is made somewhat obscure by the replacement of $Ca^{++}$ and $PO_4^{\equiv}$ ion activities by their concentrations.

This method has been applied to the data of MacGregor and Nordin (5) and MacGregor (11). In the first case negative log $(Ca^{++})\ (OH^-)^2$ was plotted against negative log $(H^+)^3(PO_4^{\equiv})$ using the data from experiments in which adult dead bone powder was equilibrated with 'tris' and cacodylate buffers. Unitary activity coefficients were assumed for the calcium and phosphate ions. The graph is shown in Fig. 1. The slope of the line of best fit obtained by linear regression analysis was -0.632 which represents a Ca/P ratio for the solid phase of 9.494/6.

Secondly, the results of experiments with child dead bone powder were treated by this method and the results are shown in Fig. 2. In this case the slope was -0.743 representing a Ca/P ratio of 8.076/6.

It would appear then that in the child the solid phase in equilibrium with body fluids is octacalcium phosphate (OCP), $Ca_8H_2(PO_4)_6\cdot 5H_2O$, while in the adult the Ca/P ratio of the equilibrating solid phase is more nearly that of HA. This interpretation is dependent on the degree of confidence we can place in the slopes of the fitted lines.

A standard correlation analysis yields a correlation coefficient, r and a regression coefficient, b. The significance of these coefficients can be decided using standard and distribution transformations. Applying this method to the human bone powder we find: -

| Bone | r | Significance | b | Significance | Interpretation |
|---|---|---|---|---|---|
| Adult | -0.983 | 0.001 | -0.632 | 0.001 | Ca/P probably $\frac{9}{6}$ or $\frac{10}{6}$ |
| Child | -0.976 | 0.001 | -0.743 | 0.001 | Ca/P probably $\frac{8}{6}$ |

Considering the complex form of the basic distribution of the regression coefficients, it is extremely difficult to decide whether the observed value of $-\frac{1}{x}$ i.e. the Ca:P ratio, is most likely to represent an integer ratio of 9/6 or 10/6. In practice, and in view of the relatively low S.E. of the slopes, we can say that it is unlikely that the adult bone powder has yielded a Ca/P stoichiometric ratio of 8/6.

The above data indicate that the solid phase saturating the solutions in the experiments with child bone is OCP but they should not be construed to mean that bone salt in children is exclusively OCP. The behavior is also consistent with the view that the major phase is unreactive or does not have a definite solubility, while OCP - even though a minor phase - governs the apparent solubility. It has been shown in fact that the rates of dissolution and recrystallization of OCP appear to be considerably greater than those of HA (12).

Other indications support the idea that OCP may be present in bone. The 'platey' habit of crystallites in bone has been interpreted as indicating that OCP participated in their formation (12). Evidence has been presented recently to support the view that growth of HA crystals depends on a mechanism in which OCP is the initial precipitate (13, 14). The incorporation of phosphate into the apatite lattice has been shown by Dallemagne et al to involve initially an acidic phosphate which we believe is OCP (15). On the other hand, the adult bone data indicate that HA probably dominates the equilibration in aged formed bone.

Thus, the burden of evidence now available supports our considered view that bone mineral is first formed as OCP and that there are spontaneous ageing changes (hydrolysis) which convert OCP into HA. In the child, where bone growth is maximal, there tends to be more OCP in the microcrystals than in the older subject where the reorganization of the OCP to HA overtakes the precipitation process.

Since the new feature of our theory concerns the presence of OCP in 'young' bone, we have applied the calculation to the data of Nordin (4) who studied the equilibration of powdered calf bone with 0.15 M tris and cacodylate buffers at 37°C (Fig.3). His data yielded a correlation coefficient of -0.9930 which is significant at the 0.1% level and a regression coefficient of -0.7378 which is also significant at the 0.1% level. This regression coefficient represents a Ca/P stoichiometry of 8.13/6. It is highly improbable that this observed ratio could have arisen by chance if the true Ca/P ratio was 7/6 or 9/6. ($P < 0.001$) and confirms the dominance of the OCP stoichiometry in equilibrations with 'young'bone.

Since there appears to be a difference in the physico-chemical behaviour of young and adult bone, it may be possible to explain the raised serum inorganic phosphate concentrations in children by the greater solubility of OCP as compared to that of HA. MacGregor and Nordin were aware that their 'ion product' theory of blood:bone exchange and the role of the parathyroid glands in maintaining the level of the equilibrium (16) might not explain adequately the situation in child plasma. Possible explanations advanced by MacGregor (17) included the suggestions that child plasma might contain a larger proportion of Ca and P binding substances, that the metabolic pH gradient between the bone mineral tissue fluid interface and the circulating body fluids might be steeper due to increased metabolic activity in the young, or thirdly, that there might be a qualitative difference in child bone. This last now appears more plausible and explains the observed differences, at least in part.

Our present ability to re-interpret the child data, which to some extent conflicted with the 'solubility' hypothesis of parathyroid function, now yields substantial support to that thesis, since it allows rational explanation of the fact that plasma inorganic phosphates in young mammals are higher than in adult man, whereas the plasma calcium concentration is usually fairly constant at about 2.5 mM. In point of fact it is the calcium homeostasis which is the specific necessity for optimal cellular function.

The biological significance of these findings remains to be examined in detail. They suggest that OCP is present in greatest amounts in newly formed bone, whether child or adult, and that the chemical properties and interactions of two crystalline species must be taken into account for a full understanding of bone growth and remodelling, bone healing, and various pathological conditions.

In a sense this paper can only be a preliminary communication on the basis of retrospective analysis of equilibration data. It is now necessary to re-examine this line of inquiry. In particular, the pure salts should be subjected to the same chemical treatment as the bone powder to verify the position of the fitted lines as well as their slopes. Regardless of the uncertainty suggested by this point, the present evidence for the presence of OCP *in vivo* is substantial and it introduces a new factor of considerable physiological potential. Far from detracting from the concept of a metabolic process modulating a physico-chemical exchange mechanism, these data may now help to explain many of the inconsistencies previously observed with the *in vitro* models of skeletal behaviour.

Acknowledgement

We are indebted to Mr. J. W. McInnes, B.Sc., Statistics Officer to the United Kingdom Western Regional Hospital Board for the statistical treatment of these data.

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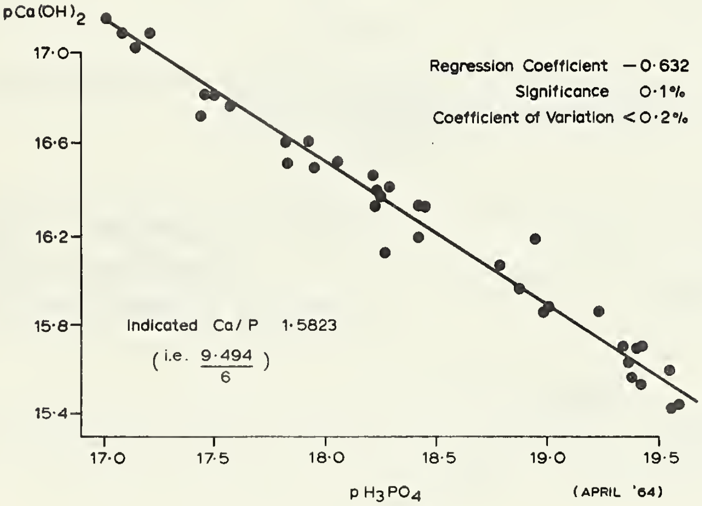

Figure 1. Chemical Potential Relation in Equilibration Studies With Powdered Adult Bone

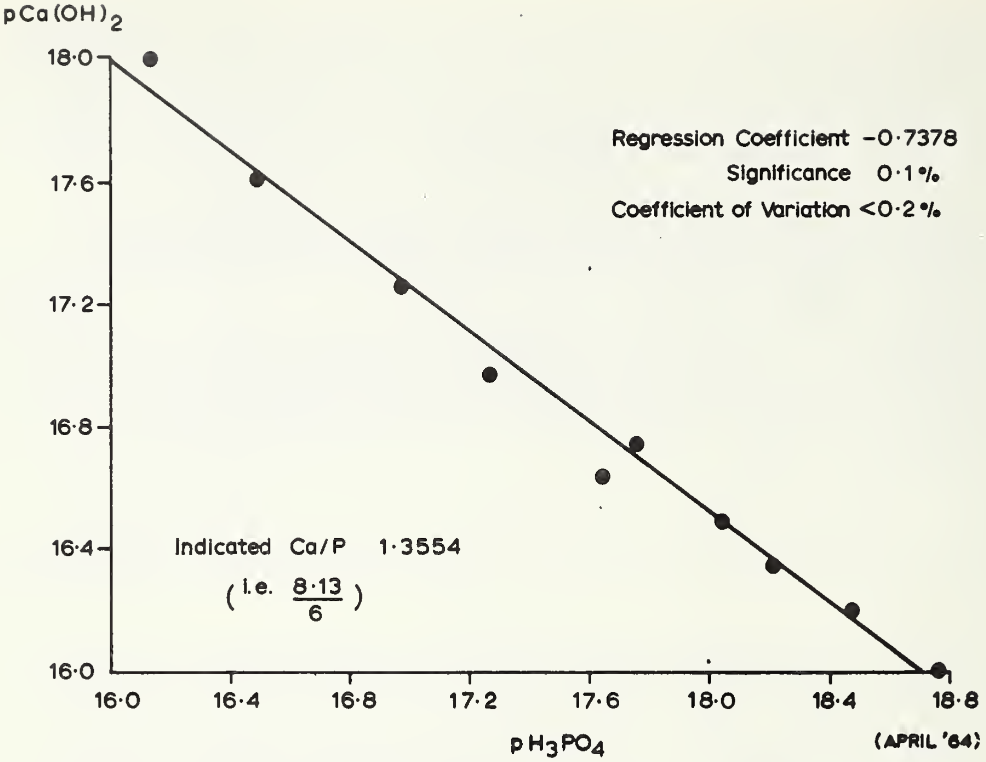

Figure 2. Chemical Potential Relation in Equilibration Studies With Powdered Calf Bone

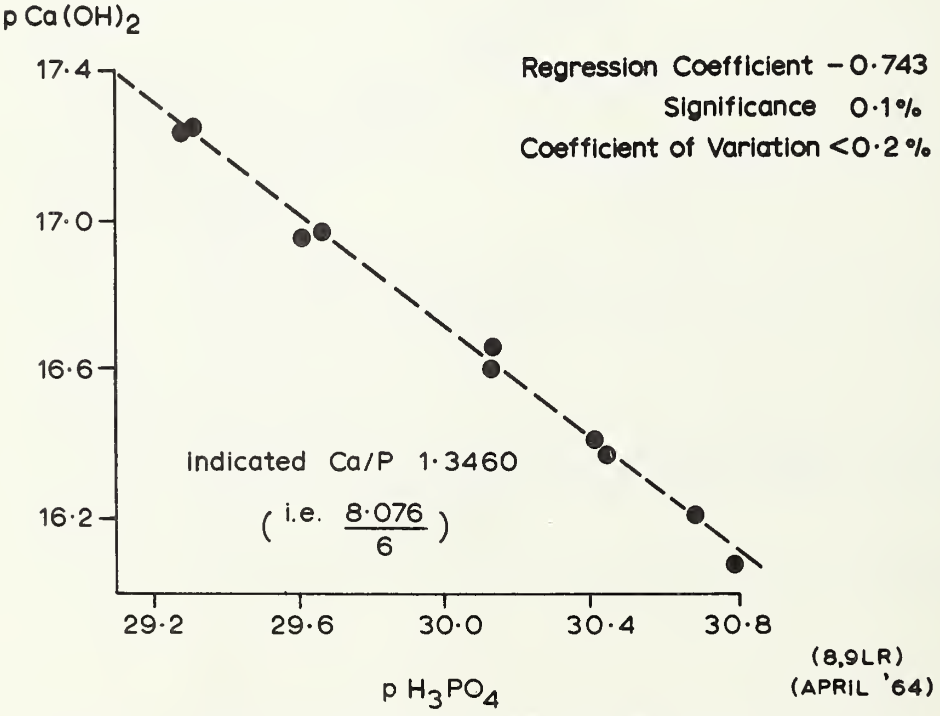

Figure 3. Chemical Potential Relation in Equilibration Studies With Powdered Child Bone

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# Progress Report
# on
# Early Strength, Flow and Dimensional Changes Obtained on Amalgam Prepared With a Standardized Mechanical Technic

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NBS

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NATIONAL BUREAU OF STANDARDS

# Early Strength, Flow and Dimensional Changes Obtained on Amalgam Prepared with a Standardized Mechanical Technic

## Abstract

The rate of hardening of five amalgams was nearly linear with time during the first hours, as shown by the increase in compressive strength or the decrease in flow. There were large differences in strength and in flow among the alloys. The data indicate that an early flow test should be given consideration as a method of specifying the setting time of amalgam. The magnitude of initial shrinkage during hardening appears to be directly related to strength and flow. The time of maximum shrinkage occurs at about the same time for all five amalgams but there are large differences in the dimensional changes which occur during the hardening of the five amalgams. The relation of the dilatometric and flow data is linear up to the time of maximum shrinkage. Specimens were prepared by an all-mechanical method so that observations could begin within three minutes from the end of trituration.

## 1. Introduction

The rate of hardening of an amalgam determines the time during which it is sufficiently plastic for packing, the time during which it can be carved, and the early strength, flow and dimensional change. In the literature there are no data on physical properties determined on specimens from 3 to 19 minutes old [1-8] except for some setting changes by Souder and Peters [9] and a preliminary report on flow by Kimmel [10]. Most of the available information on the physical properties of amalgam was obtained on specimens prepared manually. Ware and Docking [11] developed an all-mechanical procedure for trituration and for condensing the amalgam. Their data were more reproducible when obtained on mechanically prepared than on manually prepared specimens.

In this investigation specimens 3 to 60 minutes old, prepared by an all-mechanical procedure, were used to obtain values for compressive strength and flow. Also, an initial observation on setting changes was obtained 5 minutes after the end of trituration.

## 2. Procedures

The same alloys as those in a recent paper on compressive strength [12] were used in the present study (Table 1).

The flow and strength specimens were cylinders 4 ± 0.01 mm in diameter and 8 ± 0.4 mm long. The setting change specimens (about 9 mm long) were made in the same mold except the plunger had a 90° inverted cone depression on one end.

The time for preparing specimens by the all-mechanical procedure [12] was shortened from 4 minutes to 1-1/2 minutes by decreasing the time of load application during packing of the specimen. This had no effect on the strength.

The standardized procedure in making all specimens follows:

1. To make each specimen, 0.60 ± 0.01 gm of alloy and 0.90 ± 0.03 gm of mercury were triturated with a low-energy mechanical amalgamator* controlled by an electric stop clock to within 1/4 second.

2. The time of trituration (without pestle) was the minimum time that would develop the maximum or near maximum 24-hour strength [12]. Other trituration times were used in developing some of the data, as will be noted subsequently.

3. After trituration the entire mass of mixed amalgam was placed in the mold [12] and the following schedule was observed:

---

* S. S. White Amalgamator #1

| | Min-Sec |
|---|---|
| (a) End of trituration | 0 - 00 |
| (b) Place mix in mold, apply 150 kg/cm$^2$ (2,100 psi) at | 0 - 30 |
| (c) Remove pressure at | 1 - 30 |
| (d) Brush away mercury and eject specimen at | 1 - 45 |

4. All specimens were made, stored and tested at 23°C. (73°F).

The age of the specimen, when compressive strength was determined, and the crosshead speed are shown in Figure 1.

A series of flow determinations were made by initially applying a 50-kg/cm$^2$ stress on specimens that were 3, 6, 9, 12 and 15 minutes old as calculated from the end of trituration. Values for one amalgam were also obtained when the initial stress was placed on specimens 30 and 60 minutes old. To determine flow, a fiducial reading was taken at the time of load application and other readings were taken at 2, 4, 5, 8, 10, 15, 20, 30, 45 and 60 minutes, thereafter. From these readings the flow or percentage of shortening in length was calculated. The effects of the mercury-alloy ratio and the length of trituration on the early flow of one amalgam were determined.

Dimensional changes during hardening were measured in an interferometer. The schedule for making observations follows.

| | Time minutes |
|---|---|
| End of trituration | 0 |
| Every 5 minutes | 5 - 30 |
| Every 10 minutes | 30 - 60 |
| Every 15 minutes | 60 - 90 |
| Every 30 minutes | 90 - 180 |
| Final observation | 24 hours |

In the literature, most dimensional change data are based on the 15-minute fiducial reading. In this investigation, either a 5- or 15-minute observation was used as the fiducial or zero reading for dimensional change.

The residual mercury content of the specimens was estimated by the following procedure.

The weights of alloy, expressed mercury and specimen were determined on an analytical balance. The residual mercury in the specimen was then calculated by the equation:

$$\%\mathrm{Hg} = \frac{A-(B-C)}{A} \times 100$$

where A = weight of specimen

B = weight of alloy used in making the specimen

C = weight of tin and silver in expressed mercury which is obtained by multiplying the weight of expressed mercury by 0.012 as the impurity in mercury expressed from amalgam is about 1.1 percent tin and 0.1 percent silver [13].

## 3. Results and Discussion

Graphs of the compressive strengths (average of five specimens) in relation to age of the specimens (3 to 60 minutes) in Figure 1 are nearly linear and show the rates of hardening of the five amalgams. Amalgams No. 1 and 2 show the same rate of hardening but are slightly different in strengths. Amalgams No. 3, 4 and 5 show a slower rate of hardening and lower compressive strengths than amalgams No. 1 and 2.

The absence of abrupt changes in slope in these curves indicates that the reaction is continuous at a nearly constant speed during the first hour.

The only variable in the preparation of the specimens of the different alloys was the time of trituration (Fig. 1). These are the minimum times at which maximum or near maximum strength was obtained with 24-hour specimens.

The effect of age of specimens on the flow of amalgam is shown in Figure 2. The load was applied at various times between 3 and 60 minutes from the end of trituration. For example, by applying the load when the specimen was 15 minutes old instead of 3 minutes old, the flow was reduced 50 percent (4% to 2%) on specimens stressed for 10 minutes. The data for each point is the average of two or three determinations.

Figure 3 shows the flow of amalgam specimens after 10 minutes of load application for various ages. These flow data show that the rate of hardening is very similar to that shown by the compressive strength data (Fig. 1). The numerical identification of the curves in Figure 3 is the same as that in Figure 1. As strength is increasing linearly with the time the flow is decreasing at a nearly linear rate (Fig. 3). The weakest alloy has the largest flow. These data indicate that the flow measurements are relatively more sensitive than the strength determination for evaluation of the hardening rate of the alloys tested at early age of specimens.

The determination of flow of amalgam is relatively simple in comparison with the determination of its ultimate compressive strength because a dead load is used for stressing the specimen. The dead load eliminates the difficulties of obtaining constant stress or strain rates required in the determination of compressive strength. The stress or strain rates govern the values obtained in tests for ultimate compressive strength of amalgam and are not easy to control on the variety of available testing machines [12,14]. Therefore, different laboratories should be able to agree better with each other's values for flow than for compressive strength.

The estimated values for mercury content of the five amalgams given in parentheses in Figure 3 and in Table 2 had ranges of 0.1 to 0.9 percent which indicate good reproducibility for the method. These values for mercury content are in accord with values which are expected to be found in well-condensed amalgam.

Increasing the trituration decreases the flow as shown in Figure 4. The minimum trituration time that will give a maximum 24-hour strength in alloy No. 2 is 20 seconds [12]. Increasing the trituration time to 80 seconds did not materially increase the 24-hour compressive strength of alloy No. 2 [12], but Figure 4 shows that the early rate of hardening is accelerated as manifested by its decrease in flow.

On 6-minute-old specimens, loaded for 10 minutes, an increase in the mercury-alloy ratio to as much as three had no effect on flow (Fig. 5). The average value was 2.5 percent ($\sigma = 0.1$) which is within the precision of the data. The flow at a ratio of four was slightly higher.

The effect of trituration on the 24-hour dimensional change during hardening (Fig.6) shows the expected result of decreasing dimensional change with increased trituration. The unexpected result is that all of the alloys have a zero setting change when triturated for 40 ± 10 seconds. In other words , when amalgam is triturated for less than the 30 seconds, under the experimental conditions of this paper, expansion occurs and when triturated for more than 50 seconds shrinkage takes place.

The standardized all-mechanical technic prescribed for the five amalgams uses a fixed weight of mercury and alloy, and the same mechanical mixing and packing procedure so there should be about the same amount of work done in preparing each specimen. These data on dimensional change show that it is feasible to prescribe a standardized technic including a fixed trituration time for the five amalgams even though their rate of hardening is different as the data on flow and compressive strength indicate.

Since the five amalgams became stiffer and stronger with time, but at different rates, it was thought that these effects could be detected in the data on dimensional changes with time. Seemingly, the time of occurrence of the maximum early shrinkage during hardening should occur earlier with the amalgam (No. 2) having the most rapid rate of hardening as shown by the data on early flow and strength. Any such assumption is hazardous because all of the physical and chemical changes occurring in the early hardening of amalgam are not well known and have not been definitely related to the shape of the curves of dimensional change during hardening.

To see if the foregoing correlation with respect to dimensional change existed, each of the five amalgams was triturated for 30 or 40 seconds. The results are in Figure 7.

A comparison of the five amalgams (Fig. 7) shows that a wide variation in the amount of shrinkage occurs and that the setting change curve reaches a minimum at 45 ± 10 minutes. Amalgam No. 5 with the greatest shrinkage has the largest flow while amalgam No. 1 with the least shrinkage has the smallest flow. Thus it would appear that the curves in Figure 7 reflect the rate of hardening.

Doubling the trituration time from 30 to 60 seconds with amalgam No. 3 (Fig. 8) and with amalgam No. 1 had very little effect on the time at which the maximum shrinkage occurred but nearly doubled the amount of shrinkage. On amalgams No. 2, 4 and 5 there was little effect on either the time at which the maximum shrinkage occurred or on its amount. When the trituration time was doubled, the curve for amalgam No. 1 showed a continuing shrinkage for as long as 24 hours when the readings were discontinued. The other curves were shaped similar to those in Figure 8.

Reference to Figures 7 and 8 shows that the time of maximum setting cannot be selected accurately. In Figure 7 the change from 30 to 90 minutes through the maximum shrinkage averages about 1 micron per centimeter with the largest change being 1.7 microns. These results indicate that there is a difference in the setting or reaction rate of the five alloys as shown by the dilation curves (Fig. 7 and 8).

Since both the data on early dimensional change during hardening and on early flow should delineate the rate of matrix formation, there should be a linear relation between the two sets of data. Plots of the relationship in Figure 9 show this to be true up to the time of maximum shrinkage.

## 4. Summary

An all-mechanical, standardized procedure for preparing test specimens was found suitable for five widely used amalgams employing 0.60 grams of alloy, 0.90 grams of mercury and 40-second trituration in a nylon capsule without a pestle in a low-energy mechanical amalgamator.

These amalgams became stiffer and stronger with time but at different rates according to data on flow and compressive strength obtained on specimens with ages as low as 3 minutes. The mercury-alloy ratio up to 3 did not appreciably affect flow. Data on dimensional change during hardening also reflected the different rates of hardening.

The early flow test is a convenient method of specifying the setting time of dental amalgam.

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Table 1

Commercial Dental Alloys Tested

| Brand | Batch Number | Manufacturer |
|---|---|---|
| Aristaloy | 540548 | Baker Dental Division<br>Engelhard Industries, Inc. |
| New True Dentalloy | 17860358 | S. S. White Dental Manufacturing Co. |
| Silver Crown Medium | 511 | General Refineries, Inc. |
| Twentieth Century Fine Cut | 13G61F | L. D. Caulk Co. |
| Twentieth Century Micro Non Zinc | 30H61 | L. C. Caulk Co. |

Table 2

Estimated Mercury Content

|  | Percent Mercury |  |
|---|---|---|
| Amalgam | Individual determinations | Average |
| 1 | 48.9; 48.9; 48.6 | 49 |
| 2 | 47.8; 47.8; 48.2 | 48 |
| 3 | 48.9; 48.9; 49.0 | 49 |
| 4 | 50.0; 50.4; 50.4 | 50 |
| 5 | 45.7 46.6 45.7 | 46 |

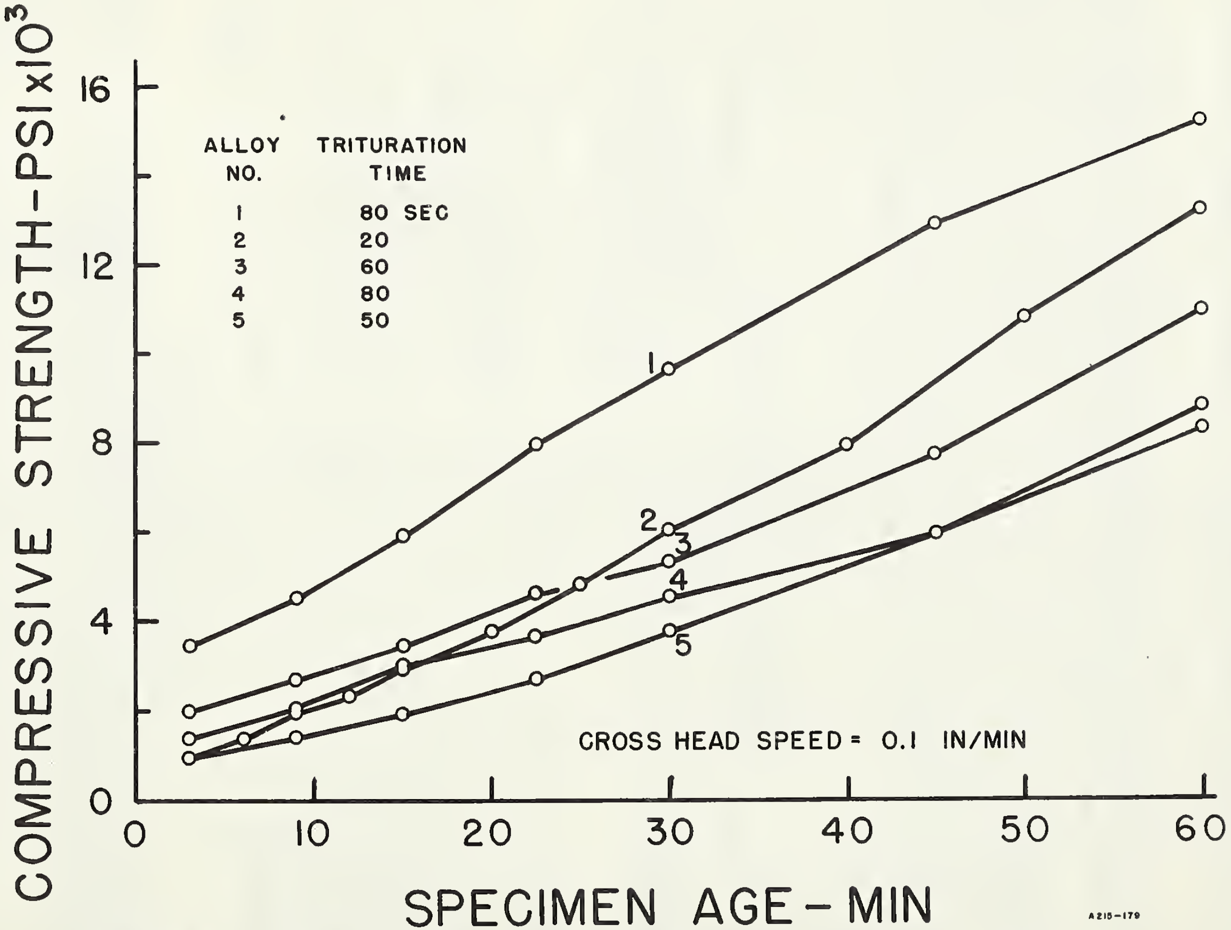

Figure 1. Relation between compressive strength and age of specimen. Age of specimen is the time from the end of trituration to the time the load is applied. The times of trituration given in the upper lefthand corner are the minimum times at which the approximate highest compressive strength at one day was attained. A Model 1, S. S. White amalgamator, and a nylon capsule without pestle were used. The numbers given to the amalgams are in sequence with decreasing compressive strength on 30-minute-old specimens.

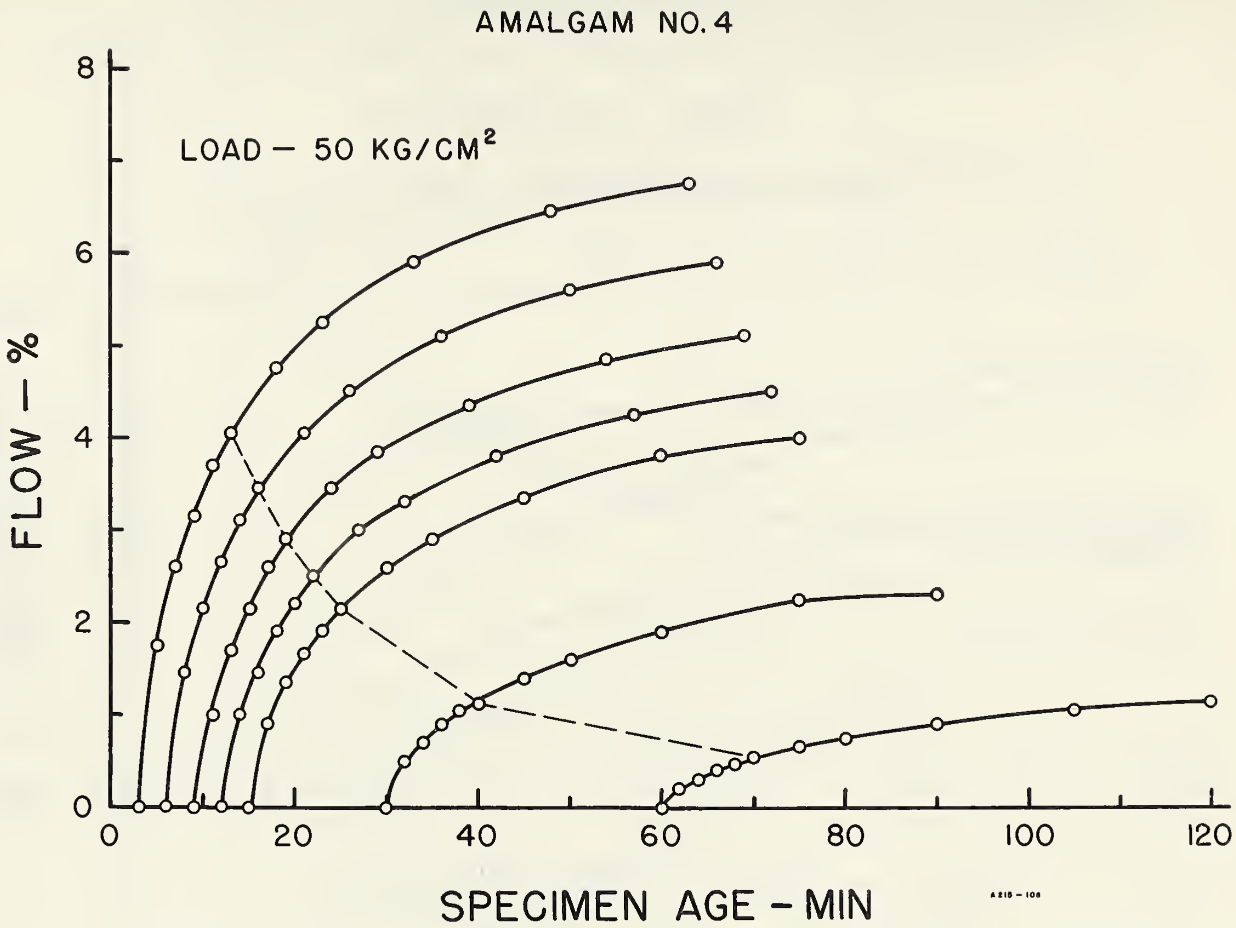

Figure 2. Family of curves showing the effect of the age of the specimen on flow of amalgam No. 4. Broken line connects points at 10 minutes after application of load.

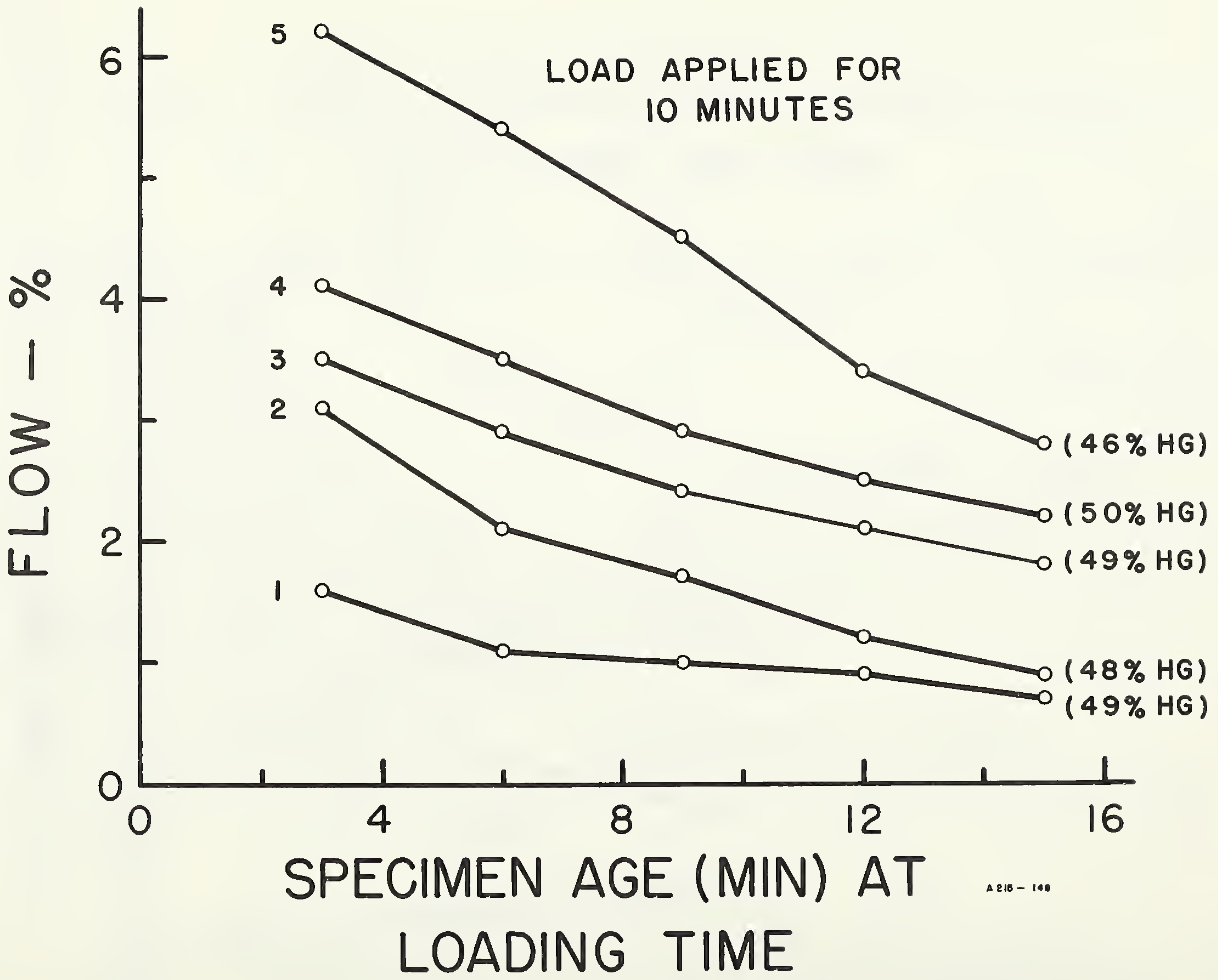

Figure 3. Relation between the age of specimen and flow for the five amalgams. The initial points on the left of the curves represent the amount of flow in percent which occurred when 3-minute-old specimens were subjected to a stress of 50 kg/cm$^2$ for 10 minutes; the second points for 6-minute-old specimens, and so on. The figures in parentheses at the right end of the curves give the calculated percentages of residual mercury in the specimens.

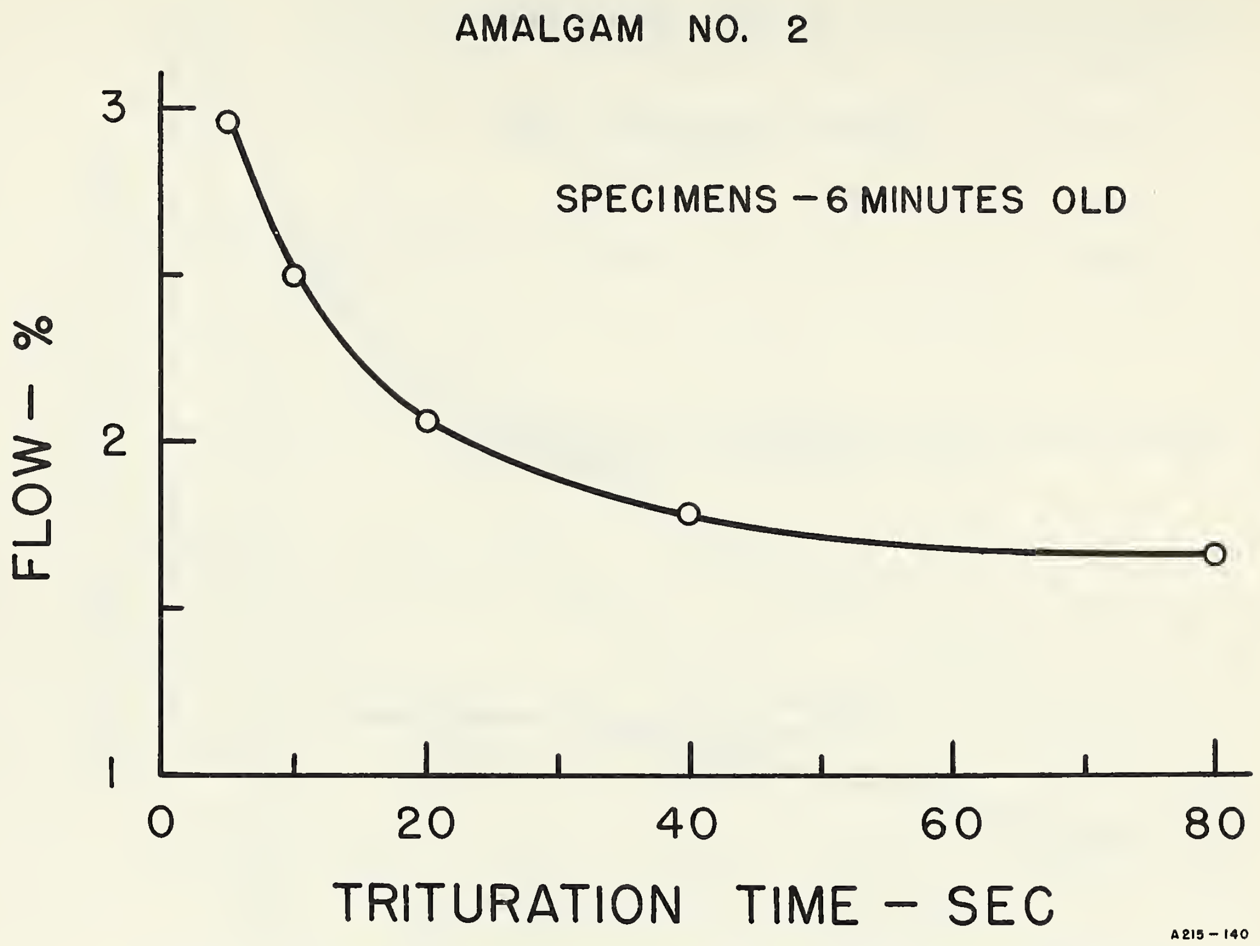

Figure 4. Effect of trituration time on flow of 6-minute-old specimens of amalgam No. 2. A 50-kg/cm2 stress was applied for 10 minutes.

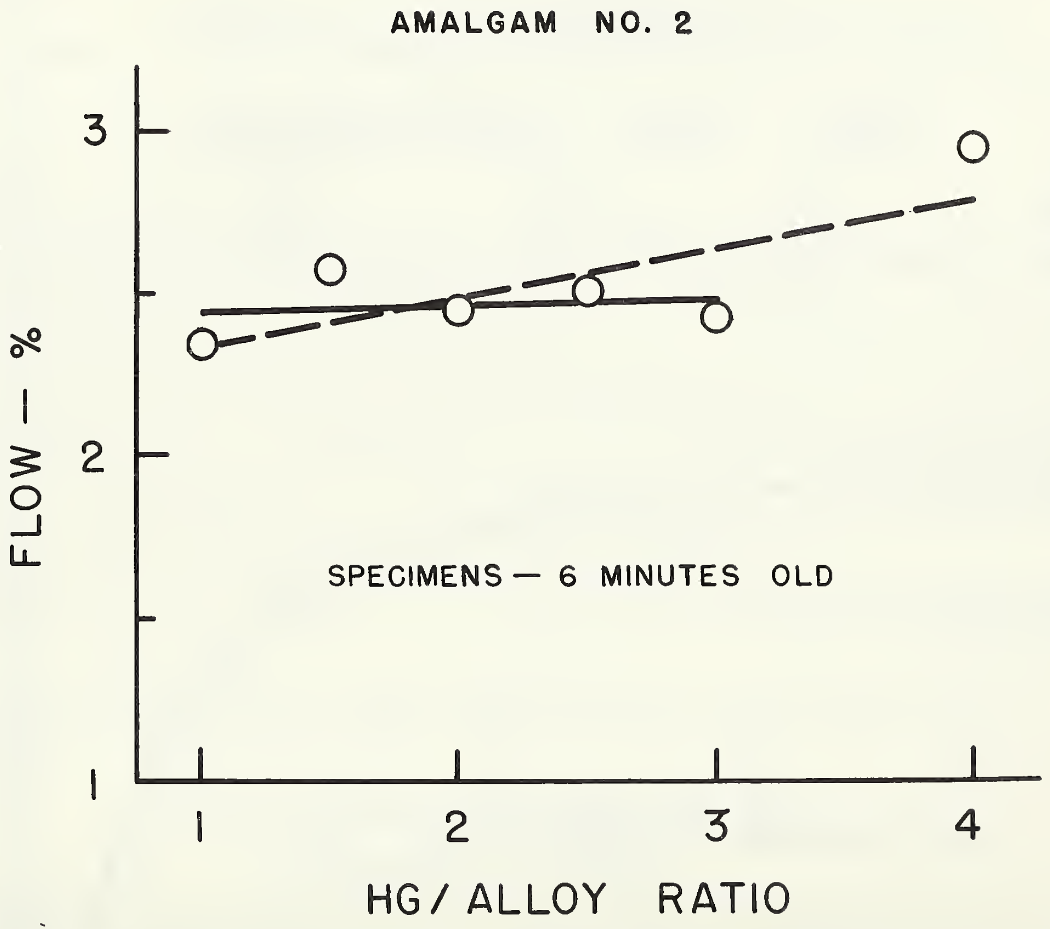

Figure 5. Effect of mercury-alloy ratio on flow. A 50-kg/cm$^2$ stress was applied for 10 minutes. The horizontal line represents the data for mercury-alloy ratios from 1 to 3, and the broken line, the 1 to 4 ratios. Both lines were fitted by the least squares method.

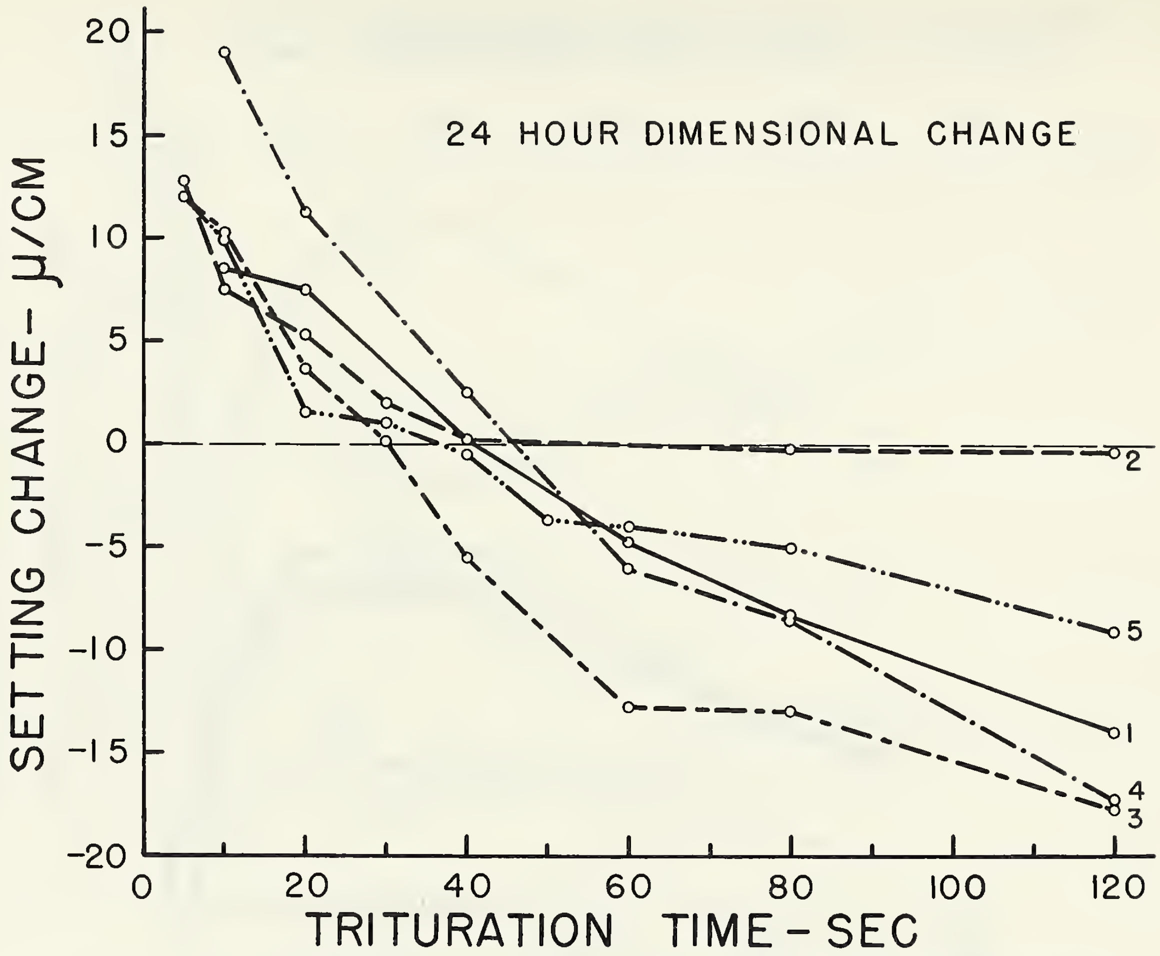

Figure 6. Effect of trituration time on 24-hour dimensional change. The dimensional change during 24 hours represents the difference in the length of the specimen (μ/cm) determined at 15 minutes after the end of the trituration and at 24 hours.

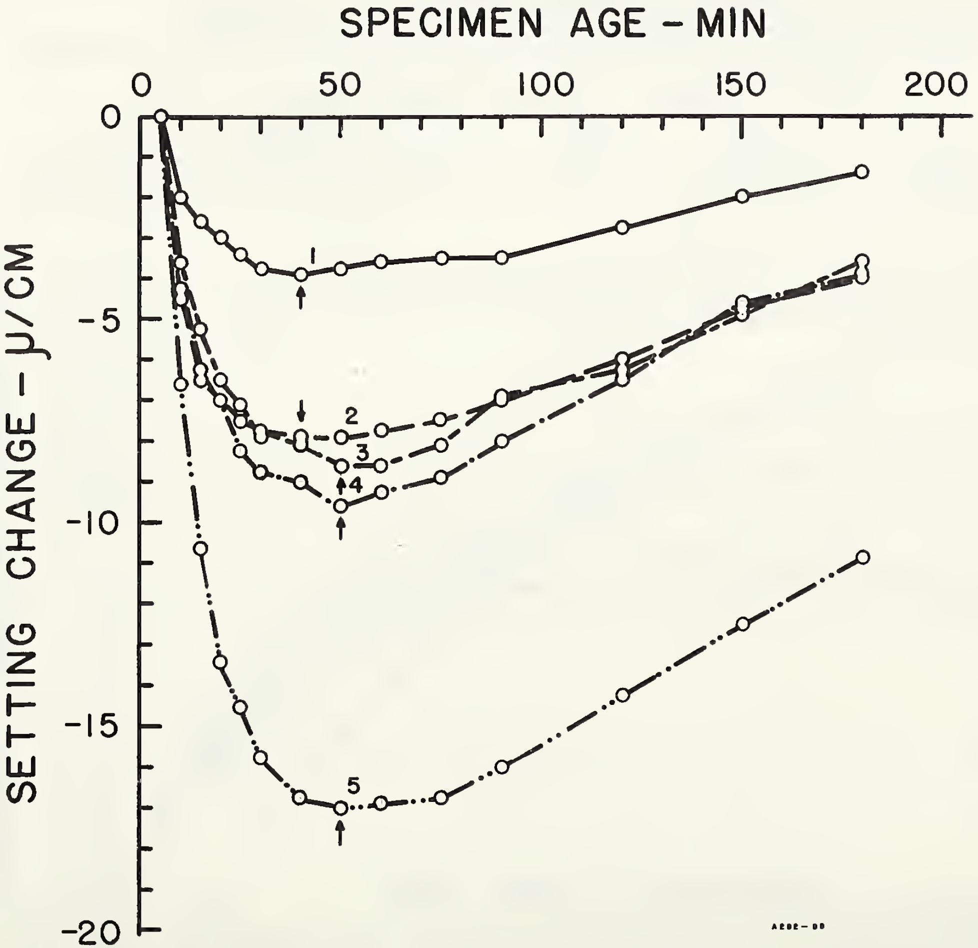

Figure 7. Dimensional setting change occurring during the first three hours. The fiducial reading was taken when the specimen was 5 minutes old. Arrows indicate time of maximum shrinkage.

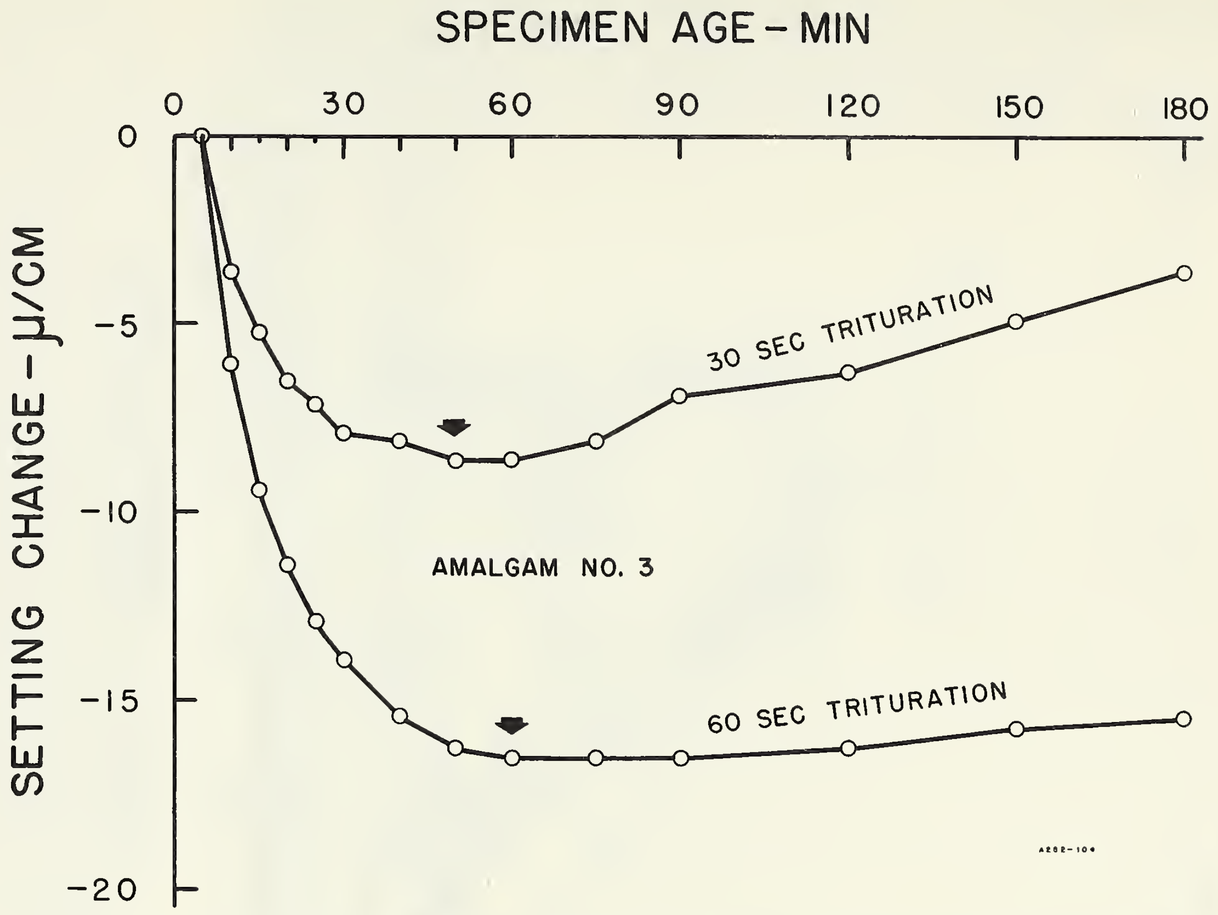

Figure 8. Effect of doubling the trituration time of amalgam No. 3 on setting change during the first three hours and on the time of maximum shrinkage (arrows).

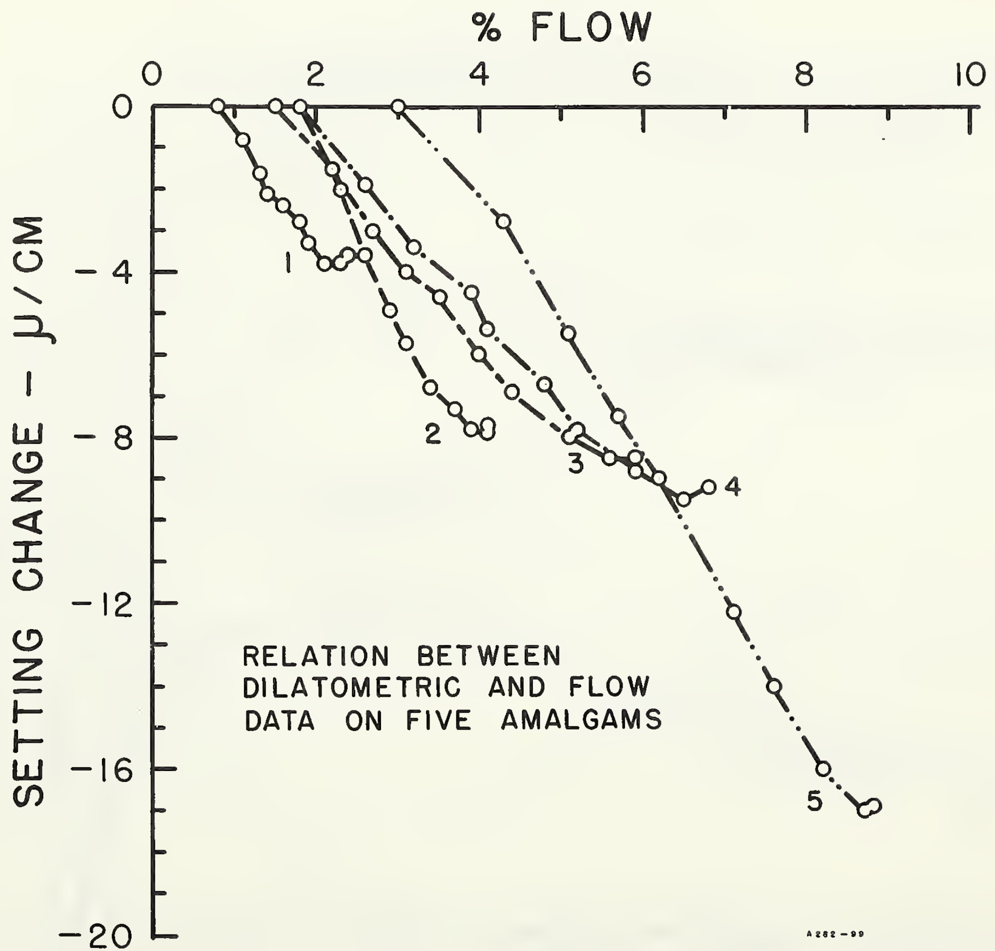

Figure 9. The relation between linear dimensional change occurring during the hardening of five dental amalgams and their flow under a pressure of 50 kg/cm$^2$. The individual points on the curves represent readings taken when the specimens were 5, 7, 9, 11, 13, 18, 23, 33, 48 and 63 minutes old. Ages of the specimens were reckoned from the end of trituration.

# NATIONAL BUREAU OF STANDARDS REPORT

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# Progress Report on Viscoelastic Behavior of Dental Amalgam

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NATIONAL BUREAU OF STANDARDS

# Viscoelastic Behavior
# of Dental Amalgam

## Abstract

Measurements made on dental amalgam in tension indicate that amalgam exhibits three types of viscoelastic phenomena: (1) instantaneous elastic strain, (2) retarded elastic strain (transient creep) and (3) viscous strain (steady state creep). The combination of elastic plus retarded strain can be represented by an equation of the form $\epsilon = A\sigma + B^2\sigma^2$ where A and B are functions of time but not of the stress, $\sigma$. The viscous strain rate can be represented by an equation of the form $\dot{\epsilon}_v = K\sigma^m$ where K and m are constants of the material. By applying a nonlinear generalization of the Boltzmann superposition principle to a general equation describing the creep behavior of amalgam, the results of creep tests can be directly related to the results of stress-strain tests.

## 1. Introduction

There is no information in the literature on the viscoelastic behavior of dental amalgam except the limited amount that could be obtained from stress-strain relationships [1,2]. The stress-strain data indicate that amalgam exhibits a nonlinear relation between stress and strain over the entire range of stress investigated; this has been found to be true in both tension and compression. It has been further noted that the shape of the stress-strain curves vary with the strain rate indicating that the strain developed is not only dependent on the applied stress but also upon time of application of the stress. The results of tensile stress-strain investigation by Rodriguez [1] indicated that dental amalgam might be a viscoelastic material. It was, therefore, the objective of this study (1) to make an exploratory investigation of the types of viscoelastic phenomena exhibited by dental amalgam in tension (2) to describe the viscoelastic phenomena exhibited by amalgam in terms of available viscoelastic theory and (3) to make available a practical example and method of application of viscoelastic theory to a dental material.

## 2. Theory

In general a strain-hardened material will exhibit at least one or more of three types of viscoelastic phenomena; (1) elastic behavior (2) viscous behavior (steady-state creep) and (3) retarded elastic behavior (transient creep).

The elastic behavior is a linear function of the applied stress and completely recoverable; that is to say the elastic strain developed in a material is directly proportional to the applied stress and disappears instantaneously upon removal of the stress. The instantaneous elastic response of a material is commonly represented by a spring as shown in Figure 1-A.

$$\epsilon_e = J_o\sigma \qquad (1)$$

where

$\epsilon_e$ = the elastic strain developed in the material

$\sigma$ = the applied stress

$J_o$ = the proportionality constant called the elastic compliance which is the reciprocal of the elastic modulus.

In the case of viscous behavior (steady-state creep), the strain developed in the material is a function of both the applied stress and the time of application of the stress, but the viscous strain is nonrecoverable upon removal of the stress, that is to say it is permanent. The viscous strain is a linear function of time, and the viscous strain rate for any given stress value is a constant; on the other hand the viscous strain may or may not be a linear function of the applied stress. The viscous response of a material can be represented by the behavior of a dashpot as shown in Figure 1-B.

The viscous strain rate $\dot{\epsilon}_v$ for a material exhibiting a linear relation to the applied stress is as follows:

$$\dot{\epsilon}_v = \frac{1}{\eta}(\sigma - \sigma_o) \qquad (2)$$

where

$\dot{\epsilon}_V$ is the viscous strain rate

$\eta$ is the coefficient of viscosity of the material

$\sigma$ is the applied stress

$\sigma_o$ is the applied stress level at which the material first begins to exhibit viscous flow and is called the yield stress.

A material exhibiting viscous flow in accordance with the above relation is said to exhibit ideal plastic behavior (Bingham behavior) while a material which obeys the above equation in its viscous flow but has a value of $\sigma_o$ which is zero, is said to be a material which exhibits Newtonian flow. Materials demonstrating viscous flow which is a nonlinear function of the applied stress; usually obey one of the following two equations in their viscous response

$$\dot{\epsilon}_V = K (\sigma - \sigma_o)^m \qquad \text{or} \qquad \dot{\epsilon}_V = A \sinh B\sigma \tag{3), (4}$$

where K and m, and A and B are constants at any given temperatures. A material exhibiting nonlinear viscous response described by the first equation is said to be pseudo plastic [3] in its viscous behavior in either case whether $\sigma_o$ is or is not zero. On the other hand a material demonstrating nonlinear viscous response according to the second equation does not have a yield stress $\sigma_o$ and is said to exhibit viscous flow (plastic flow) in accordance with the Kauzmann rate theory [4]. This type of viscous flow occurs in metals at temperatures near their melting points [5,6,7,8].

The retarded elastic response of a material may be represented by a single model or by either a finite or an infinite series of models called Voigt elements. A Voigt element is shown in Figure 1-C.

Retarded elastic behavior (transient creep) which can be described by means of a single Voigt element is represented by the following equation:

$$\epsilon_R = J \left(1-e^{-t/\tau}\right) \sigma \tag{5}$$

where

$\epsilon_R$ is the retarded elastic strain developed in the material

$\sigma$ is the applied stress

$J$ is the retarded elastic compliance

$\tau$ is the retardation time of the material.

Retarded elastic behavior corresponding to a finite series of Voigt elements is represented by the following equation:

$$\epsilon_R = \sigma \sum_{i=1}^{n} J_i\left(1-e^{-t/\tau_i}\right) \tag{6}$$

where

$\epsilon_R$ is the retarded elastic strain developed in the material

$\sigma$ is the applied stress

$J_i$ is the retarded elastic compliance of each Voigt element

$\tau_i$ is the retardation time of each Voigt element

Retarded elastic response represented by an infinite number of Voigt elements is described by the following equation:

$$\epsilon_R = \sigma \int_0^{\infty} J(\tau) \left(1-e^{-t/\tau}\right) d\,\tau \tag{7}$$

In all of the above equations for the representation of retarded elastic strain, the relation between the retarded strain and the applied stress is assumed to be linear; however, in many materials this relation has been found to be nonlinear. The mathematical description of the nonlinear case has been treated in various publications [9, 10]. For example, if the retarded elastic strain (transient creep) is related to the applied stress by a second degree equation in which the time response can be represented by an infinite number of Voigt elements, such a retarded elastic response can be described by the following equation:

$$\epsilon_R = \sigma \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau + \sigma^2 \int_0^\infty B(\gamma)\left(1-e^{-t/\gamma}\right) d\gamma \qquad (8)$$

where

$\epsilon_R$ is the retarded elastic strain

$\sigma$ is the applied stress

$J(\tau)$ is the continuous distribution of retarded elastic compliance as a function of the variable retardation time $\tau$ of the continuous distribution of Voigt responses in the linear stress response element of the material

$B(\gamma)$ is the continuous distribution of retarded elastic compliances as a function of the variable retardation time $\gamma$ of the continuous distribution of Voigt elements in the nonlinear second degree stress response of the material.

In a material which exhibits all three types of behavior (elastic, viscous, and retarded elastic), and in which the three types of behavior are additive (obey a superposition relation), the creep strain developed in the material at any time t under an applied stress $\sigma$ may be related to the strains due to the individual behaviors by the following equations:

$$\epsilon = \epsilon_o + \epsilon_R + \epsilon_v \qquad (9)$$

where

$\epsilon$ is the creep strain developed in the material at any time t under an applied constant stress

$\epsilon_o$ is the instantaneous elastic strain developed in the material

$\epsilon_R$ is the retarded elastic strain developed in the material

$\epsilon_v$ is the viscous strain developed in the material.

These individual responses as was noted earlier may be linearly or nonlinearly related to the stress. For example, in a material which exhibits all three phenomena linearly related to the applied stress then using the linear equation of each response given earlier, the above equation takes the form:

$$\epsilon_R = J_o \sigma + \sigma \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau + \frac{\sigma t}{\eta} \qquad (10)$$

Next, consider an example of a material which exhibits all three types of behavior, but in which the viscous strain developed is nonlinearly related to the applied stress as follows:

$$\epsilon_v = K(\sigma - \sigma_o)^m t \qquad (11)$$

while the retarded elastic strain is nonlinearly related to the applied stress by an equation of second degree as given earlier and the instantaneous is assumed to be linear; thus, the above superposition equation for creep strain takes the following form:

$$\epsilon = J_o \sigma + \sigma \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau + \sigma^2 \left[\int_0^\infty B(\gamma)\left(1-e^{-t/\gamma}\right) d\gamma\right]^2 + K(\sigma - \sigma_o)^m t \qquad (12)$$

Where the creep strain is linearly related to the applied stress, the strain can be written as a product of a general creep compliance, which is a function of time only, and the applied stress as given by the following equation:

$$\epsilon = J(t)\sigma \tag{13}$$

where

$\epsilon$ is the creep strain developed

$\sigma$ is the applied stress

$J(t)$ is the general creep compliance

For example, in the linear creep behavior as seen in equation (10), the general creep compliance $J(t)$ is of the form:

$$J(t) = J_o + \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\,\tau + \frac{t}{\eta} \tag{14}$$

In the case that creep strain is linearly related to the applied stress, the stress-strain behavior for a material may be related to the creep behavior by means of the linear superposition principle developed by Boltzmann [11] as shown below:

$$E\,(\sigma_t) = \int_0^{\sigma_t} J(T-\theta)\,d\,\sigma\,(\theta) \tag{15}$$

or

$$E\,(T) = \int_0^T J(T-\theta)\,\frac{d\sigma(\theta)}{dt}\,d\,\theta \tag{16}$$

where

$\sigma$ is the applied stress related to the experimental time $\theta$ in a prescribed experimental functional relation.

$E(T)$ is the strain of a stress-strain relation measured at a time $T$ subsequent to the variable time $\theta$.

$\frac{d\sigma(\theta)}{d\theta}\,d\,\theta$ is the increment of stress from time $\theta$ to time $(\theta + d\,\theta)$.

For example, applying the Boltzmann superposition principle to the example of creep given in equation (10) gives the following relation between creep behavior and the strain measured in a stress-strain test [12]:

$$E\,(T) = J_o\sigma + \int_0^\infty\int_0^\infty J(\tau)\left(1-e^{-\frac{T-\theta}{\gamma}}\right) d\,\tau\,\frac{d\sigma(\theta)}{d\,\theta}\,d\,\theta + \int_0^T \frac{\sigma(\theta)}{\eta}\,d\,\theta \tag{17}$$

An excellent discussion of the application of the Boltzmann superposition principle as applied to linear viscoelastic phenomena is given by Leaderman [13]. However, if a material exhibits a nonlinear relation in its retarded strain (transient creep) and the applied stress, as discussed earlier, the linear Boltzmann superposition principle cannot be applied in its present form to relate strain of a stress-strain test on the material to the creep strain. A generalization of Boltzmann's superposition principle developed by Nakada [10] may be used to relate nonlinear retarded elastic creep behavior to the stress-strain behavior of the material as given:

$$E_R\,(\sigma) = \int_0^{\sigma(T)} \Psi(T-\theta)\,d\,\sigma\,(\theta) + \int_0^{\sigma(T)}\int_0^{\sigma(T)} \Phi\,(T-\theta)\,\Phi\,(T-\phi)\,d\,\sigma\,(\theta)\,d\,\sigma\,(\phi)$$

$$+ \int_0^{\sigma(T)}\int_0^{\sigma(T)}\int_0^{\sigma(T)} E(T-\theta)\,E(T-\phi)\,E(T-\alpha)\,d\,\sigma\,(\theta)\,d\,\sigma\,(\phi)\,d\,\sigma\,(\alpha) \quad + ----- \tag{18}$$

Now for example, consider the application of the nonlinear superposition principle of Nakada as applied, to the case of retarded strain (transient creep) related to the applied stress by a second degree equation as shown earlier:

$$\epsilon_R = \sigma \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau + \sigma^2 \left[\int_0^\infty B(\gamma)\left(1-e^{-t/\gamma}\right) d\gamma\right]^2 \tag{19}$$

now

$$\psi(t) = \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau$$

and

$$\Phi(t) = \int_0^\infty B(\gamma)\left(1-e^{-t/\gamma}\right) d\gamma$$

where $\psi(t)$ and $\Phi(t)$ are the respective first degree and second degree retardation functions as given in the nonlinear superposition principle. Therefore, applying the nonlinear superposition principle to the above retarded creep example one has:

$$E_R\left(\sigma(T)\right) = \int_0^{\sigma(T)} \int_0^\infty J(T)\left(1-e^{-\frac{T-\theta}{\tau}}\right) d\tau\, d\sigma(\theta)$$

$$+ \int_0^{\sigma(T)} \int_0^{\sigma(T)} \int_0^\infty \int_0^\infty B(\gamma)^2\left(1-e^{-\frac{T-\theta}{\gamma}}\right)\left(1-e^{-\frac{T-\phi}{\gamma}}\right) d\gamma\, d\gamma\, d\sigma(\theta)\, d\sigma(\phi) \tag{20}$$

Considering an example of a material which exhibits all three phenomena but in which the creep strain is a nonlinear function of the applied stress according to equation (12), then applying the nonlinear superposition principle, the stress-strain behavior of the material may be related to the creep behavior by the following equation:

$$E = \sigma J_0 + \int_{\infty}^{T} \int_0^\infty J(\tau)\left(1-e^{-\frac{[T-t]}{\gamma}}\right) d\tau\, d\sigma(t)$$

$$+ \int_{-\infty}^{T} \int_{-\infty}^{T} \int_0^\infty B(\gamma)\left(1-e^{-\frac{[T-\theta]}{\gamma}}\right)\left(1-e^{-\frac{[T-\phi]}{\gamma}}\right) d\gamma\, d\sigma(\theta)\, d\sigma(\phi)$$

$$+ \text{-------} \qquad + \int_0^T K\,\sigma^m(t)\, dt \tag{21}$$

## 3. Materials

The specimens were prepared using a commercial alloy for dental amalgam certified to comply with American Dental Association Specification No. 1. This alloy (composition approximately Ag 70%, Sn 26%, Cu 3.5%, Zn 0.5%) was mixed with mercury and condensed into a mold as described by Rodriguez and Dickson [1] to produce a specimen with dimensions as shown in Figure 2. Specimens were aged for at least one week to obtain essentially full mechanical strength [14].

## 4. Procedure

The dumbbell-shaped specimen was placed in the grips and Tuckerman optical strain gages were mounted on opposite sides of the specimen as shown in Figure 3. To obtain the creep curve, readings were taken on the strain gages, a weight was suspended from the lower grip and a second strain gage reading was taken immediately. Strain readings were then made at 15 second intervals for 4 minutes and at increasingly longer intervals until the strain rate became constant (usually after approximately 1.5 hours). At the end of this period, the load was removed, a strain reading was taken immediately and the recovery curve was followed by reading first at 15 second intervals and then at longer intervals until the strain became constant.

Strain readings obtained on the two sides of the specimen were averaged and strain was plotted against time to obtain the loaded creep and unloaded recovery curves. Readings on the strain gages were normally made to the nearest $2 \times 10^{-5}$ inch. Since a gage length of 0.25 inch was used this is equivalent to a strain of $8 \times 10^{-5}$. Thus in the results given below differences in strain of $1 \times 10^{-4}$ are approximately equal to the minimum reading difference.

Loads placed on the specimen (with a nominal 0.01 $in^2$ cross sectional area) varied from 5 to 40 lbs. giving stresses from approximately 500 to 4,000 psi. Most specimens were used for several runs, first at high and then at lower stresses. The first loaded creep run was considered a strain hardening treatment and data obtained on these runs were not used in the calculation of results other than for viscous strain rate. All runs were made at 23 ± 1°C.

## 5. Results and Discussion

The creep curves (both loaded and recovery curves) of strain hardened dental amalgam as shown for a number of different stress levels in Figure 4 indicate that at room temperature amalgam exhibits three different types of viscoelastic phenomena: (1) instantaneous elastic strain (2) retarded elastic strain (transient creep) and (3) viscous strain (steady-state creep).

The viscous strain rate was determined from the loaded portion of the creep curve by taking the slope of the straight line portion of the curve, and was also determined from the recovery portion of the creep curve by dividing the value of the recovery strain (the permanent strain in the specimen) by the total time the load was on the specimen. The viscous strain rates for any given creep curve as calculated from the loaded and recovery portions of the curve were found to agree fairly well as shown in Table 1. The log of the viscous strain rate was found to be a linear function of the log of the applied stress, as shown in Figure 5; thus the viscous strain rate could be related to the applied stress by the following equation:

$$\dot{\epsilon}_v = K \sigma^m \tag{22}$$

where

$\dot{\epsilon}_v$ is the viscous strain rate

$\sigma$ is the applied stress

K and m are constant of the material

The value of m for amalgam is the value of the slope of the curve in Figure 5. While the value of K is the antilog of the viscous strain rate value at a value of applied stress $\sigma$ of 1 psi. The values of K and m for the dental amalgam used in this investigation were found to be K equals 2.85 and $4.98 \times 10^{-19}$ and m equals 3.99 and 3.92 from loaded and unloaded data respectively.

The strain developed in amalgam due to the other two phenomena (1) instantaneous elastic strain and (2) retarded strain can be determined from the strain recovery since these two types of strain are recoverable while the viscous strain is not. Thus, at any given load the strain values taken from the creep curve after the sample has been unloaded (that is in the recovery portion of the creep curve) are subtracted from the strain value on the creep curve at the instant just before unloading of the specimen. This difference is plotted against recovery time $t_1 = T_1 - T_u$ where $T_u$ is the time at which the specimen was unloaded and $T_1$ is the time of the strain value on the recovery portion of the curve. These difference values, $\epsilon'$, are seen plotted against the recovery time for each load or stress in Figure 6. These plots are a measure of the combination of the elastic and retarded elastic strain behavior of dental amalgam as a function of time for various stress levels. In theory the same plot may be obtained from the loaded portion of the creep curve by taking values off the loaded creep curve and subtracting the viscous strain accumulated in the specimen at that time. The accumulated viscous strain at any time may be calculated by multiplying the viscous strain rate by the time corresponding to that value on the creep curve. Thus the difference between the creep curve value on the loaded protion and the viscous strain value at a corresponding time is a measure of the combination of the instantaneous and retarded elastic strain. However, a small error in the viscous strain rate causes a large error in the difference value. Therefore, the plot of the combination of elastic and retarded elastic strain versus time as obtained from the loaded creep curve is subject to large possible error; for this reason the combination elastic behavior is obtained from the recovery portion of the creep curves.

The combination elastic strain (instantaneous and retarded elastic strain) values are seen in Table 2 tabulated against time for various applied stress levels. The combination strain becomes asymptotic with time in accordance with theory as seen in Figure 6. The combination elastic strain values were plotted as a function of the various stress levels for corresponding time; as shown in Figure 7. The combination elastic strain is seen to be a nonlinear function of the applied stress. When the combination strain values were divided by their corresponding stresses and then plotted against the corresponding stress for a fixed time a linear plot was obtained for each fixed time as illustrated in Figure 8; this result indicated that the combination elastic behavior of amalgam as a function of applied stress under the test conditions could be represented by an equation of the form:

$$\epsilon' = A(t)\,\sigma + B^2(t)\,\sigma^2 \qquad (23)$$

where

$\epsilon'$ is the combination of elastic and retarded elastic strain

$\sigma$ is the applied stress

$A(t)$ and $B(t)$ are constants for any given time value and are functions of time but not of stress. The value of $A(t)$ for any time value is the intercept at $\sigma = 0$ of the plot for that time value as shown in Figure 8 while $B^2(t)$ is the slope of the straight line for that time value. It is also noted in Figure 8 that as a function of stress the combination strain divided by the stress is a straight line for all values of t. This indicated that over all ranges of t the combination elastic strain obeys the same functional relation to the stress. Thus from Figure 6 it was concluded that the retarded elastic strain as a function of time could be described by means of conventional viscoelastic theory assuming either a finite or infinite number of Voigt elements while the combination elastic strain being a nonlinear function of stress indicated the need for the use of nonlinear viscoelastic theory as developed first by Leaderman[9] and later more generally by Nakada [10]. Therefore, the values for $A(t)$ and $B^2(t)$ were determined by fitting curves to the data by the method of least squares and were tabulated as a function of time as shown in Table 3. The $A(t)$ values were plotted as a function of the corresponding t values as shown in Figure 9. The $A(t)$ values are seen to approach an asymptote as $t \longrightarrow \infty$, thus when the values of the asymptote minus the $A(t)$ values are plotted as a function of the corresponding time values, Figure 10, the curve is seen to fall off in a complicated exponential behavior which could be represented by viscoelastic theory assuming an infinite number of Voigt elements or a continuous spectrum. Thus, it has been assumed that $A(t)$ could be represented by the following equation from linear viscoelastic theory since $A(t)$ is the linear term in stress:

$$A(t) = J_o + \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\tau \qquad (24)$$

$$A(t) = J_o + \int_0^\infty J(\tau)\, d\tau - \int_0^\infty J(\tau)\, e^{-t/\tau} d\tau \qquad (25)$$

$$A(t) = A_{as} - \int_0^\infty J(\tau)\, e^{-t/\tau} d\tau \qquad (26)$$

$$A_{as} - A(t) = \int_0^\infty J(\tau)\, e^{-t/\tau} d\tau \qquad (27)$$

where

$A(t)$ is the creep compliance term which is linear in stress

$J_o$ is instantaneous elastic compliance

$J(\tau)$ is the retarded elastic compliance spectrum as a function of the retardation time $\tau$

$A_{as}$ is the asymptote value of A(t) as t becomes very large.

Thus, equation (26) indeed does describe the asymptotic behavior of A(t) as a function of time as seen in Figure 9 while equation (27) could be used to describe the complicated exponential-like behavior seen in Figure 10. The linear creep compliance term A(t) is plotted as a function of log t to obtain a sigmoidal curve as shown in Figure 11. The first plateau of the sigmoidal plot at very small values of t should correspond to $J_o$ (the instantaneous elastic compliance)[15], however, the curve does not extend to short enough time values to determine the value of the plateau which would be $J_o$. Therefore, it is concluded that instantaneous elastic behavior of amalgam cannot be separated from the combination elastic behavior and thus is indeterminate from the data obtained in this investigation. The plot in Figure 11 indicates the need for improved experimentation on amalgam in which strain measurements can be made at very short times after loading or removal of the load from the specimen.

Next the nonlinear creep compliance term $B^2(t)$ was plotted against t as shown in Figure 12. The value of $B^2(t)$ is seen to approach an asymptote with increasing time. When the asymptote value of B(t) minus B(t) is plotted against time it decreases in a complicated exponential-like form as shown in Figure 13. Thus using the nonlinear theory of Nakada [10] it is concluded that the experimental B(t) for amalgam could be described by the following equation:

$$B(t) = \sqrt{B^2(t)} = \int_0^\infty B(\gamma)\ \left(1-e^{-t/\gamma}\right) d\ \gamma \qquad (28)$$

which would indeed describe the behavior of the curves seen in Figure 12 and Figure 13. It is therefore concluded that the combination elastic behavior of dental amalgam in creep under the test conditions used can be described by means of the following equation from viscoelastic theory [10,15]:

$$\epsilon' = A(t)\ \sigma + B^2(t)\ \sigma^2 \qquad (29)$$

$$\epsilon' = J_o\ \sigma + \sigma \int_0^\infty J(\tau)\ \left(1-e^{-t/\tau}\right) d\ \tau + \left[\int_0^\infty B(\gamma)\ \left(1-e^{-t/\gamma}\right) d\ \gamma\right]^2 \cdot \sigma^2 \qquad (30)$$

and since the loaded portion of the creep curve for amalgam is also composed of viscous strain $\epsilon_v$ as well as combination elastic strain $\epsilon'$ then the strain on the loaded portion is composed of the sum of the two as follows:

$$\epsilon = \epsilon' + \epsilon_v \qquad (31)$$

Thus

$$\epsilon = J_o\ \sigma + \sigma \int_0^\infty J(\tau)\left(1-e^{-t/\tau}\right) d\ \tau + \left[\int_0^\infty B(\gamma)\ \left(1-e^{-t/\gamma}\right) d\ \gamma\right] \sigma^2 + K\ \sigma^m t \qquad (32)$$

Therefore applying both linear and nonlinear viscoelastic theory to the experimental behavior of amalgam under the test conditions used, a general equation describing the creep behavior of amalgam as given by equation (32) above is obtained.

Applying the nonlinear generalization of the Boltzmann superposition principle as developed by Nakada [10], to the above creep equation for amalgam, the stress-strain curves for various stress rate conditions were calculated for amalgam from the creep data and compared to the experimental stress-strain curves obtained under those conditions. It is seen in Figure 14 that good agreement is obtained between the calculated and experimental stress strain curves. Thus, it is concluded that in the case of dental amalgam the results of creep tests can be directly related to those of stress-strain tests by use of viscoelastic theory [10,15].

## 6. Conclusion

Dental amalgam exhibits three types of viscoelastic phenomena: (1) instantaneous elastic strain (2) retarded elastic strain (transient creep) and (3) viscous strain (steady state creep).

The instantaneous elastic strain is assumed to be proportional to the applied stress but the methods used in this study did not provide an independent determination of instantaneous strain.

The combination of elastic plus retarded strain is a nonlinear function of stress and can be represented by an equation of the following form:

$\epsilon' = A\sigma + B^2 \sigma^2$ where A and B are functions of time but not of stress

The viscous strain rate is also a nonlinear function of stress and can be represented by an equation of the form:

$\dot{\epsilon}_V = K\sigma^m$ where K and m are constants of the material

A general equation describing the creep behavior of amalgam was obtained by the application of both linear and nonlinear viscoelastic theory to the experimental behavior of amalgam under the test conditions used. By applying the nonlinear generalization of the Boltzmann superposition principle to this equation the results of creep tests can be directly related to results of stress-strain tests of amalgam.

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## TABLE 1

## VISCOUS STRAIN RATES

| Specimen - Run | Stress | Strain per Minute | |
|---|---|---|---|
| | PSI | Loaded* X $10^{-5}$ | Unloaded** X $10^{-5}$ |
| 41-40-1 | 3807 | 5.6531 | 6.3511*** |
| 41-40-2 | 3807 | 5.9759 | 5.8037 |
| 39-40-1 | 3731 | 4.9184 | 5.7449*** |
| 39-40-2 | 3731 | 5.2273 | 5.0822 |
| 40-40-1 | 3731 | 7.4476 | 8.0372*** |
| 43-35-1 | 3458 | 3.1200 | 3.7086*** |
| 43-35-2 | 3458 | 3.2339 | 3.2762 |
| 43-35-3 | 3458 | 3.4171 | 3.4389 |
| 44-30-1 | 2959 | 1.9183 | 1.9778*** |
| 44-30-2 | 2959 | 1.6821 | 1.6769 |
| 44-30-3 | 2959 | 1.7387 | 1.7185 |
| 41-30-3 | 2855 | 2.8429 | 2.9555 |
| 44-25-4 | 2465 | 0.9067 | 0.8867 |
| 35-25-1 | 2375 | 1.0521 | 1.3312*** |
| 35-25-2 | 2375 | 0.9762 | 1.0621 |
| 35-25-3 | 2375 | 1.0288 | 1.0803 |
| 38-25-1 | 2366 | 1.0909 | 1.3967*** |
| 38-25-2 | 2366 | 0.7832 | 0.8211 |
| 38-25-3 | 2366 | 0.7846 | 0.8061 |
| 35-20-4 | 1900 | 0.3843 | ------ |
| 35-20-5 | 1900 | 0.4192 | 0.4348 |
| 38-20-4 | 1892 | 0.3487 | 0.3581 |
| 35-15-6 | 1425 | 0.1109 | 0.1133 |
| 38-15-5 | 1419 | 0.0761 | 0.0768 |
| 35-10-7 | 950 | 0.0306 | 0.0338 |
| 38-10-6 | 946 | 0.0232 | 0.0248 |
| 38-10-7 | 946 | 0.0170 | 0.0178 |
| 35-05-8 | 475 | 0.0083 | 0.0066 |

* From slope of straight portion of loaded creep curve

** From strain remaining in specimen after unloading and recovery

*** These values include effects of strain hardening and were not used in calculating the relation between stress and viscous strain rate.

## TABLE 2

## RECOVERY OF ELASTIC (INSTANTANEOUS PLUS RETARDED) STRAIN

| Time (Min.) | Strain* at: | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | 475 psi | 947 psi | 1422 psi | 1896 psi | 2384 psi | 2933 psi | 3458 psi | 3761 psi |
| | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ | X $10^{-4}$ |
| 0.25 | 0.745 | 1.300 | 2.113 | 3.062 | 3.729 | 5.281 | 6.301 | 7.085 |
| 0.50 | 0.945 | 1.301 | 2.163 | 3.212 | 4.186 | 5.831 | 7.101 | 7.964 |
| 1.00 | 0.845 | 1.301 | 2.297 | 3.495 | 4.660 | 6.431 | 7.601 | 8.530 |
| 2.00 | 1.045 | 1.303 | 2.597 | 3.663 | 5.030 | 6.740 | 8.268 | 9.370 |
| 3.00 | 1.045 | 1.336 | 2.763 | 3.763 | 5.254 | 7.073 | 8.846 | 10.021 |
| 5.00 | 1.046 | 1.450 | 2.914 | 3.963 | 5.563 | 7.506 | 9.468 | 10.727 |
| 10.00 | 1.047 | 1.675 | 3.065 | 4.471 | 5.997 | 8.239 | 10.224 | 11.991 |
| 20.00 | 1.147 | 1.968 | 3.199 | 4.662 | 6.468 | 9.106 | 11.201 | 13.244 |
| 30.00 | 1.286 | 1.971 | 3.421 | 4.962 | 6.700 | 9.307 | 11.668 | 13.827 |
| 40.00 | 1.366 | 1.985 | 3.654 | 5.228 | 7.055 | 9.656 | 11.969 | 14.343 |
| 50.00 | 1.445 | 2.000 | 3.787 | 5.295 | 7.265 | 9.857 | 12.169 | 14.704 |
| 60.00 | 1.445 | 2.014 | 3.830 | 5.362 | 7.340 | 10.140 | 12.602 | 14.963 |
| 70.00 | 1.445 | 2.029 | 3.864 | 5.396 | 7.415 | 10.413 | 12.735 | 15.250 |
| 80.00 | 1.445 | 2.043 | 3.982 | 5.429 | 7.498 | 10.428 | 12.891 | 15.400 |
| 90.00 | 1.445 | 2.058 | 4.110 | 5.472 | 7.582 | 10.476 | 13.035 | 15.542 |

* Each value is an average of 2 to 7 determinations of the difference between strain recorded at the time indicated and strain recorded immediately prior to removal of the tensile stress.

## TABLE 3

### VALUES OF $A(t)$ AND $B^2(t)$ IN EQUATION

$$\epsilon' = A(t)\,\sigma + B^2(t)\,\sigma^2$$

| Time<br>Min. | $A(t)$<br>$\times 10^{-7}$ | $B^2(t)$<br>$\times 10^{-11}$ |
|---|---|---|
| 0.25 | 0.922 | 2.707 |
| 0.50 | 1.122 | 2.873 |
| 1.00 | 1.303 | 2.828 |
| 2.00 | 1.309 | 3.367 |
| 3.00 | 1.280 | 3.904 |
| 5.00 | 1.322 | 4.284 |
| 10.00 | 1.363 | 4.986 |
| 20.00 | 1.383 | 5.804 |
| 30.00 | 1.409 | 6.110 |
| 40.00 | 1.544 | 6.082 |
| 50.00 | 1.589 | 6.190 |
| 60.00 | 1.581 | 6.454 |
| 70.00 | 1.570 | 6.678 |
| 80.00 | 1.588 | 6.728 |

$\epsilon'$ is combination of instantaneous and retarded elastic strain

$\sigma$ is stress

$A(t)$ and $B^2(t)$ are constants for any time and are functions of time but not of stress.

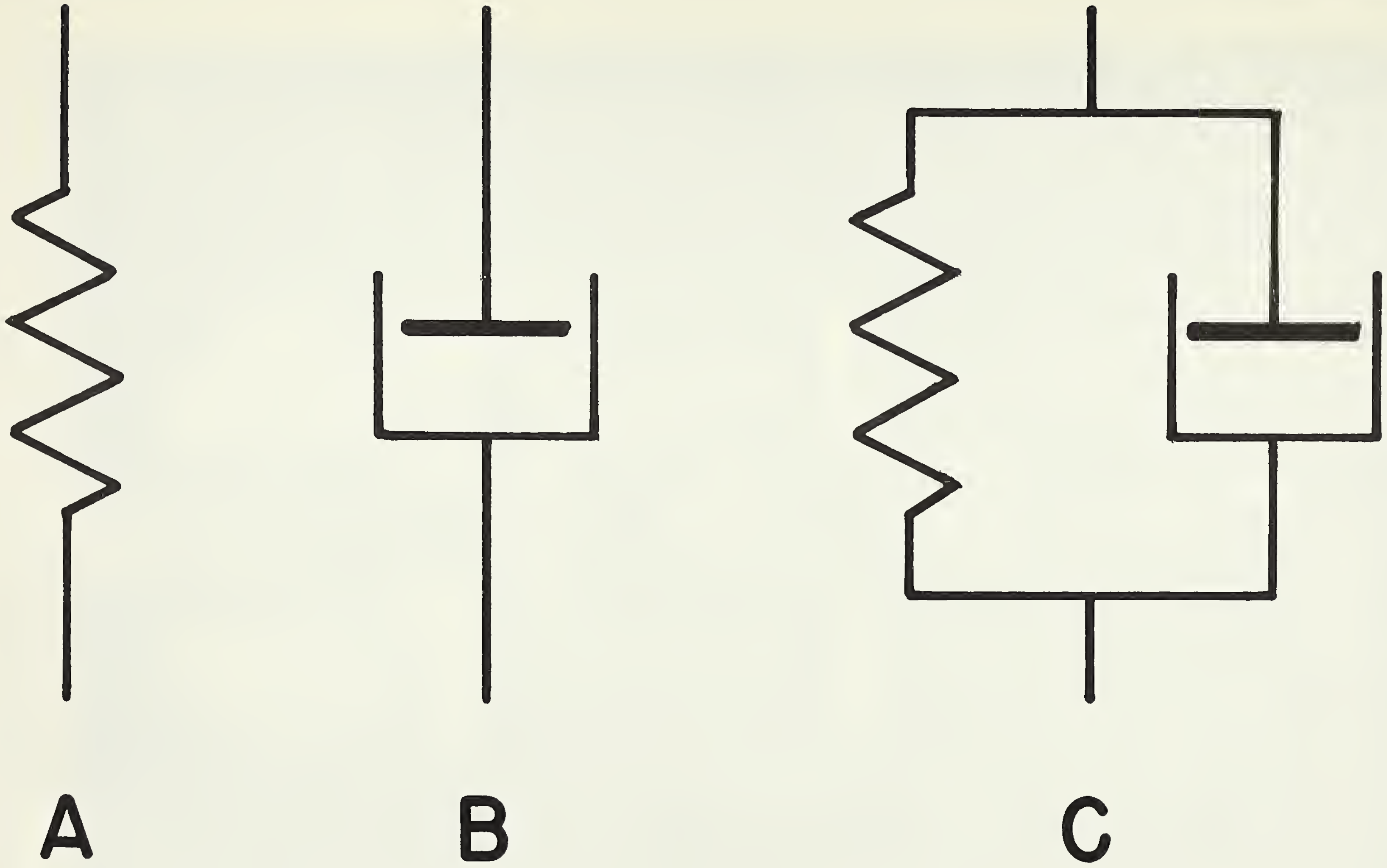

Figure 1. Models representing types of viscoelastic behavior: A-spring-elastic behavior, B- dashpot - viscous behavior, C-Voigt element - retarded elastic behavior.

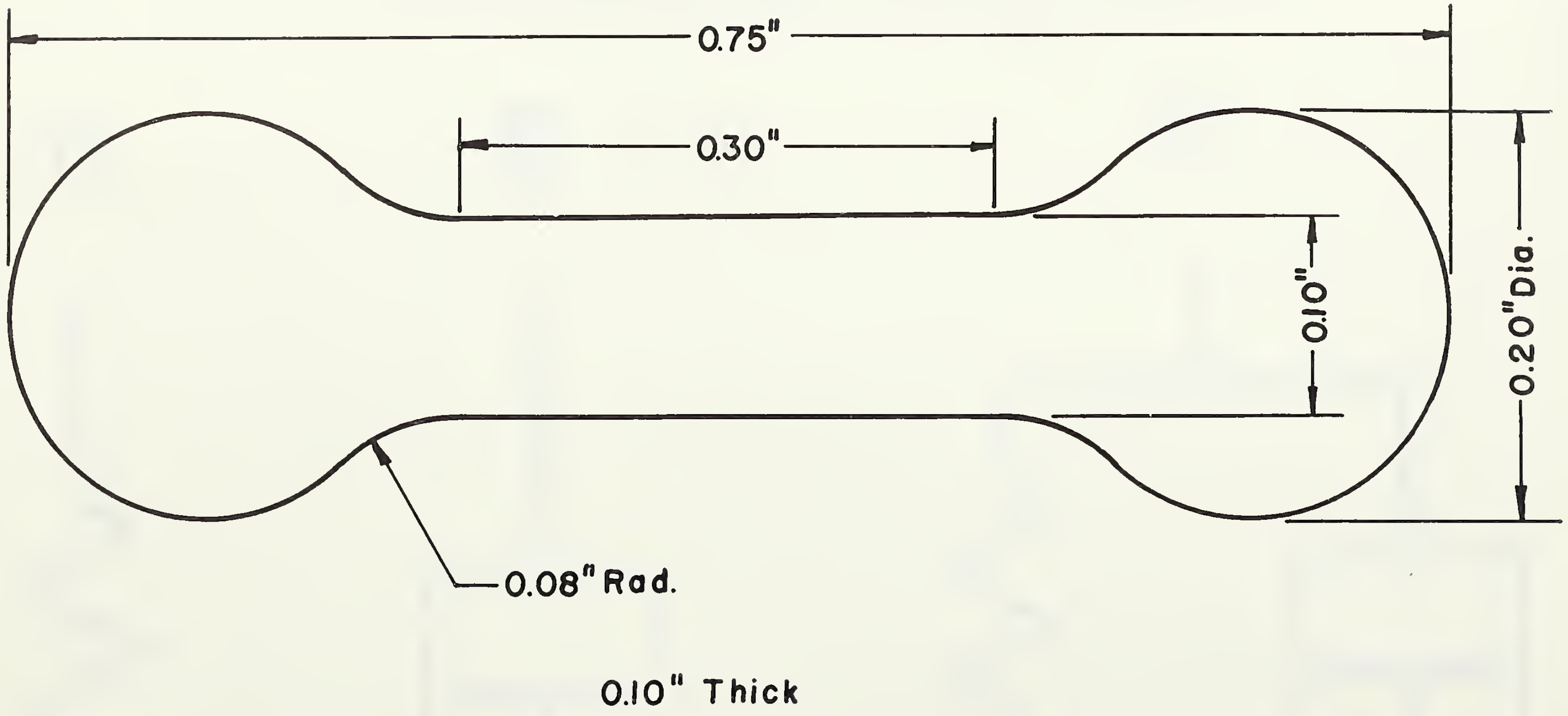

Figure 2. Dimensions of the dumbbell-shaped tensile specimen of amalgam.

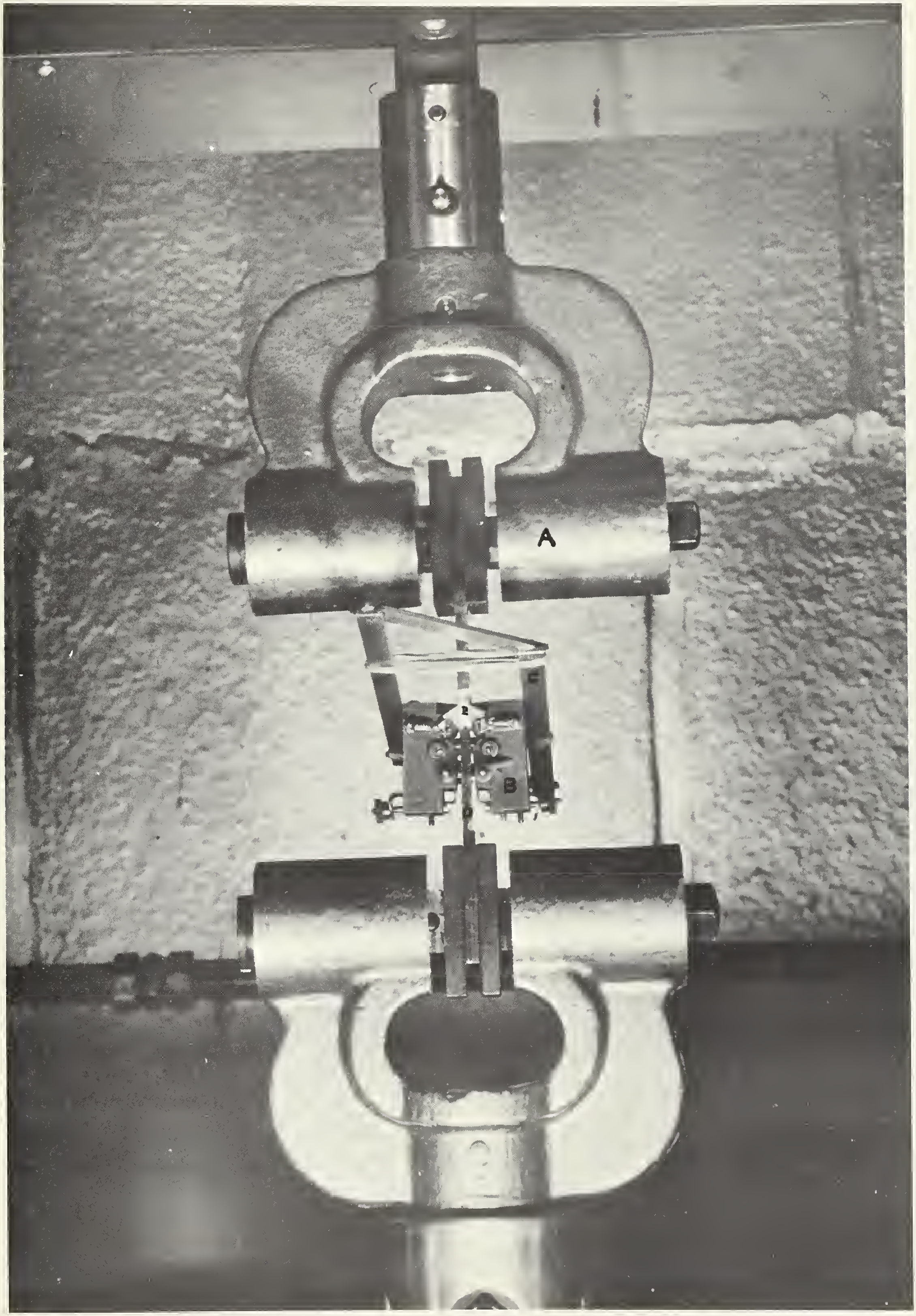

Figure 3. Tensile specimen in position for load application with optical strain gages mounted on opposite sides of the specimen.

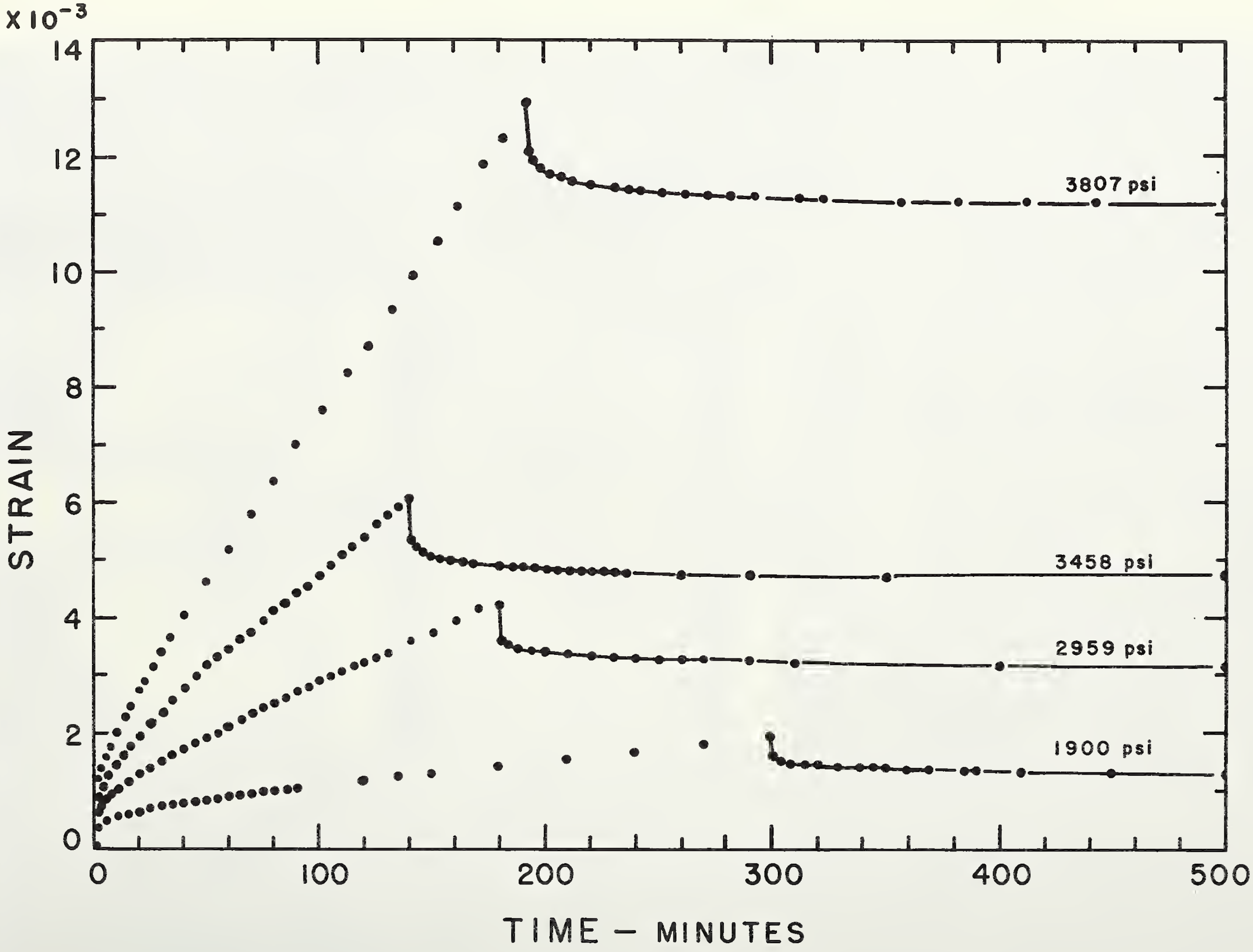

Figure 4. Creep and recovery of amalgam loaded in tension.

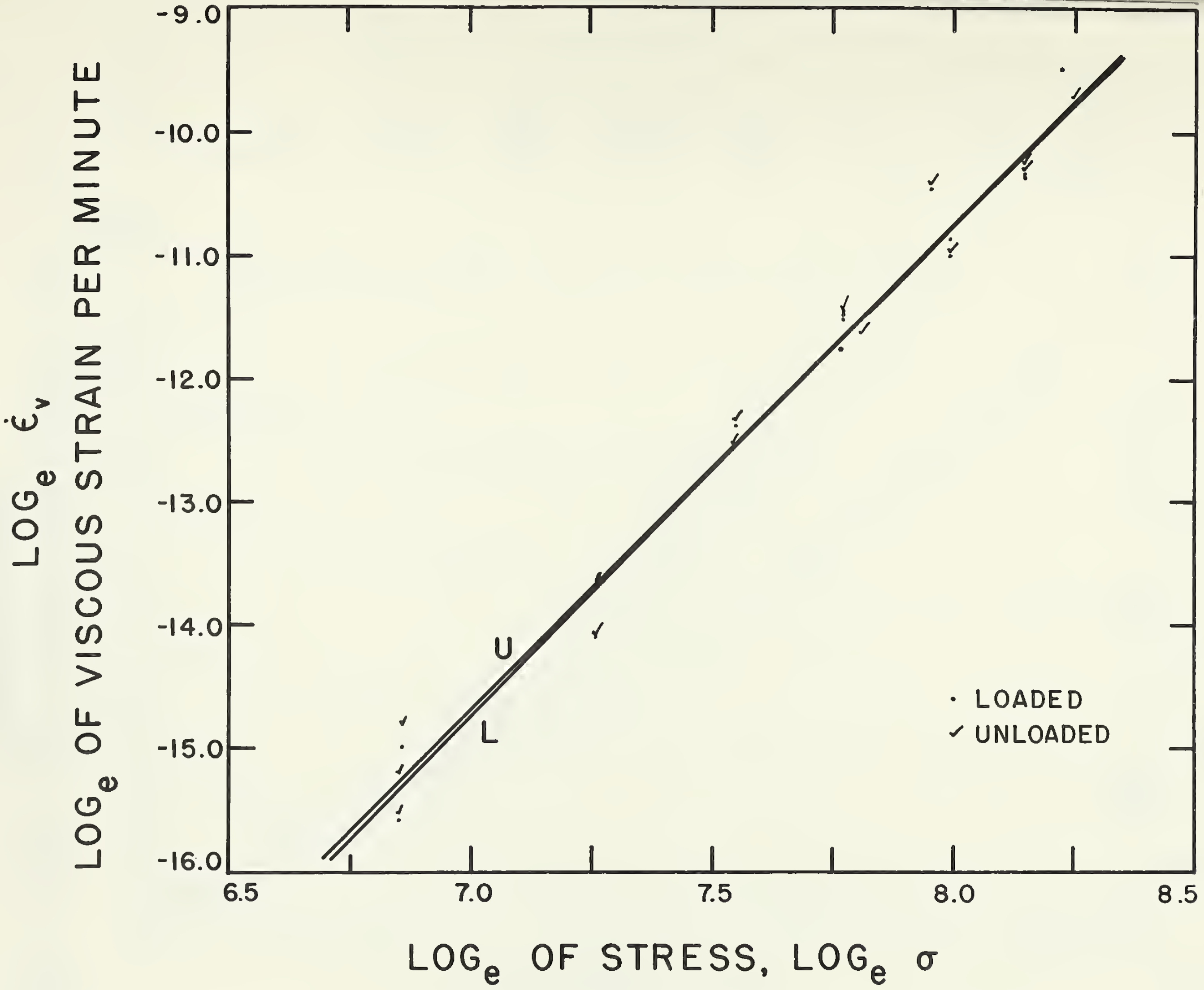

Figure 5. Relationships between viscous strain rate and stress. Straight lines are least-squares fits of equation $\log \dot{\epsilon}_v = \log K + m \log \sigma$ to the data. Loaded values are from straight portion of creep curve; unloaded values are from strain remaining in specimen after unloading and recovery.

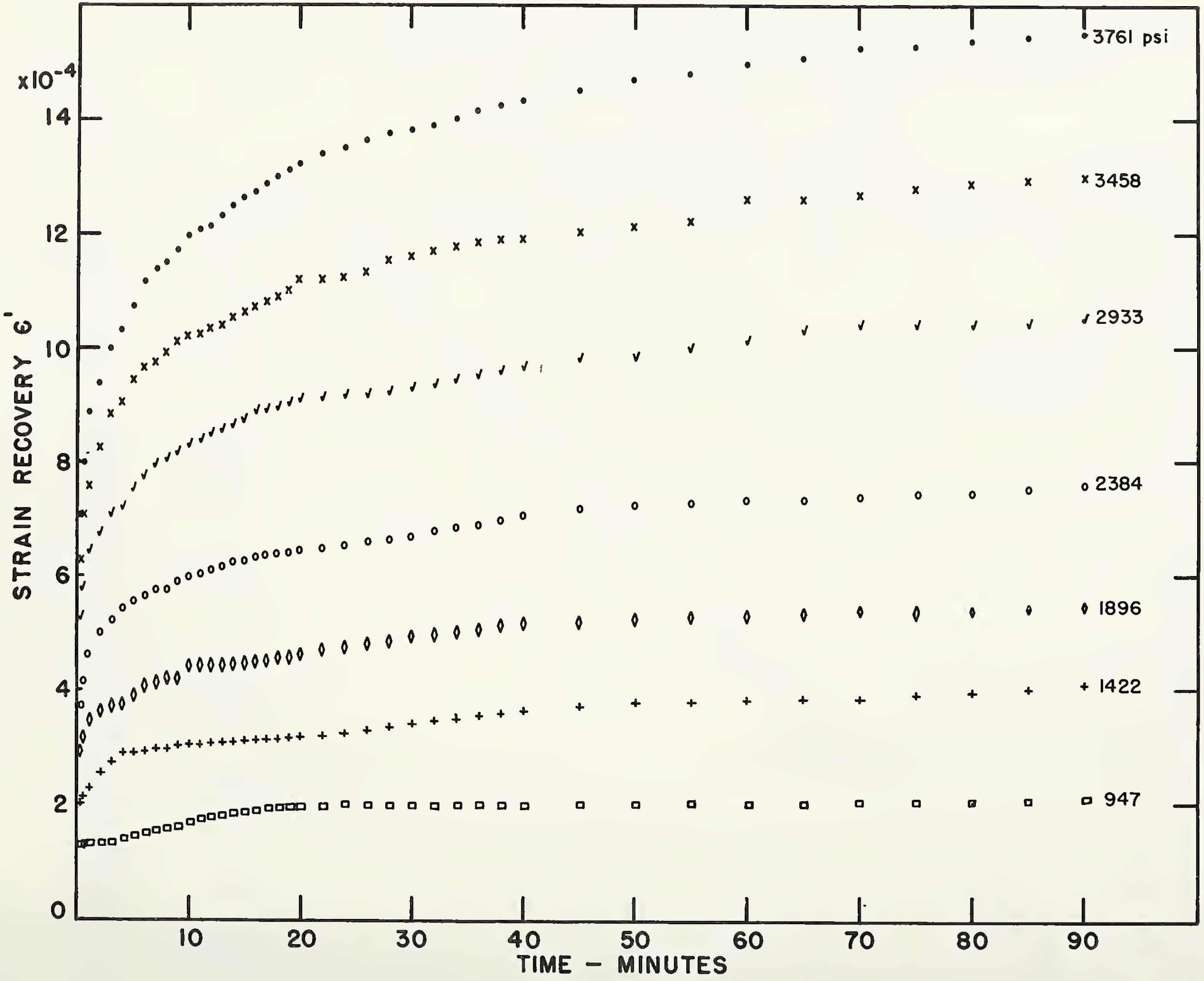

Figure 6. Strain recovery after release of tensile stress. Each curve is an average of 2 to 7 runs.

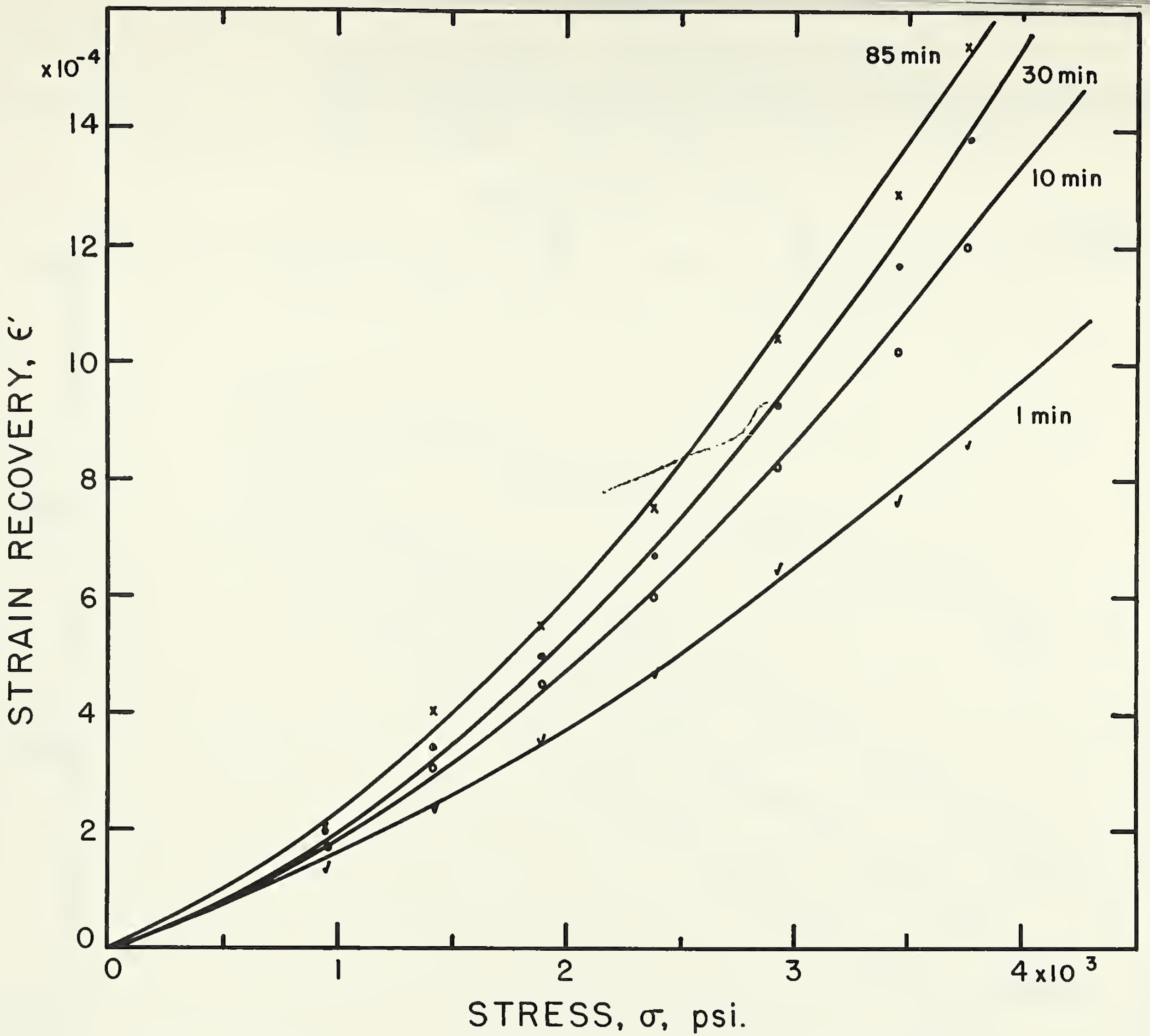

Figure 7. Relationships between recovery strain, stress and time. Plotted points are averages of 2 to 7 determinations. Curves are least-squares fits of the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$ to the data.

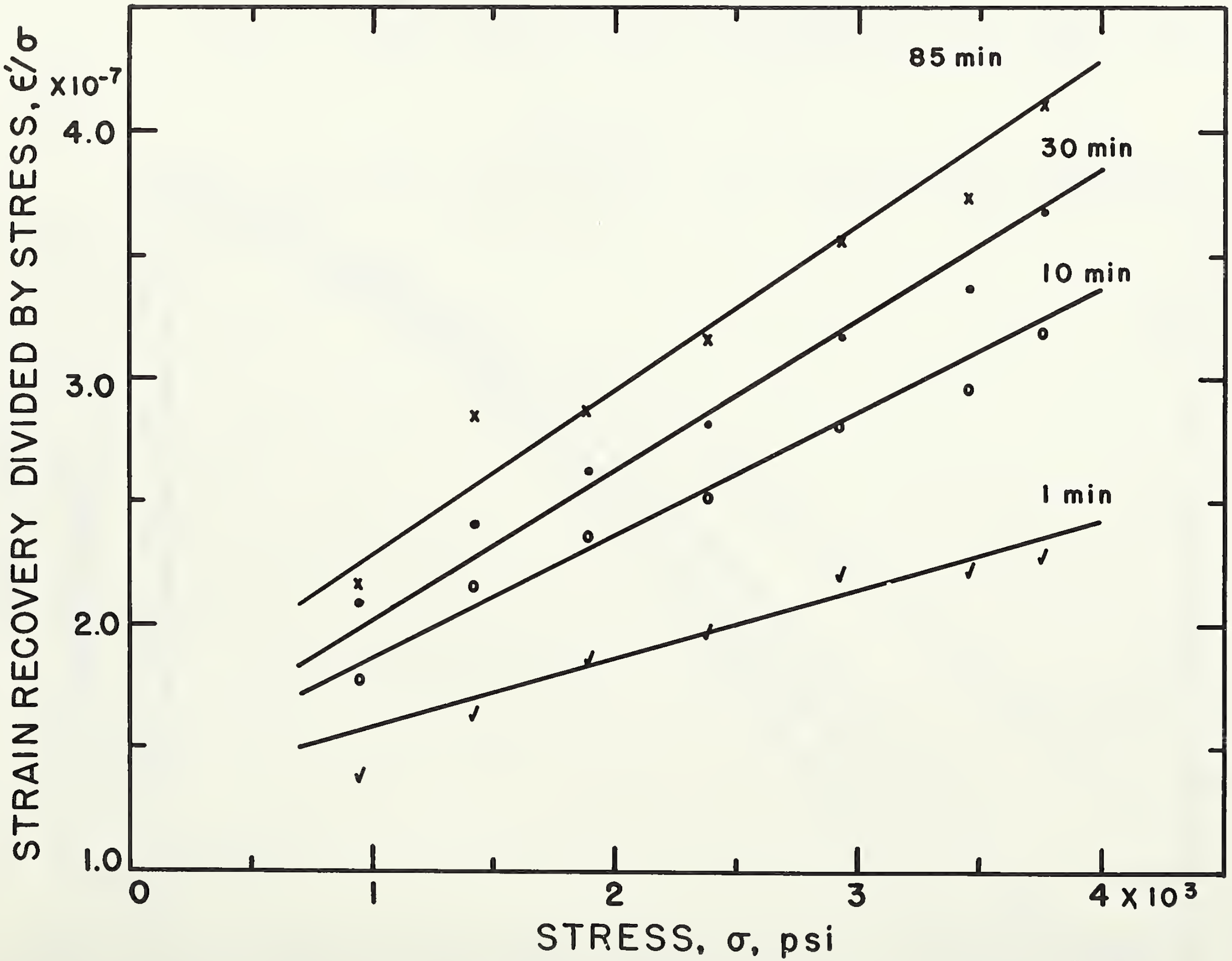

Figure 8. Relationships between recovery strain, stress and time. Plotted points are averages of 2 to 7 determinations. Straight lines represent the constants obtained by least-squares fit of the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$ to the data.

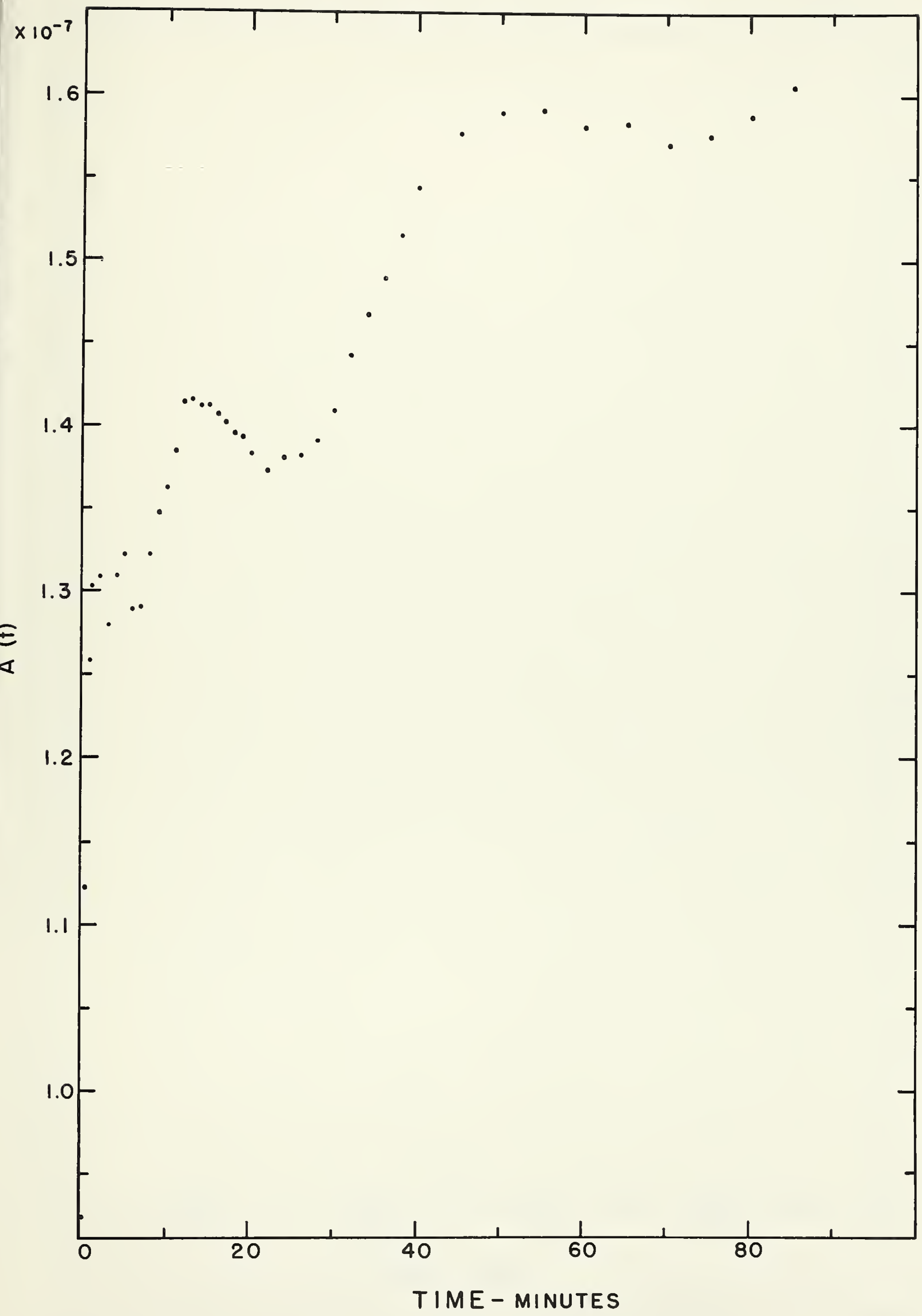

Figure 9. Variation with time of the linear creep compliance term, A(t), in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$.

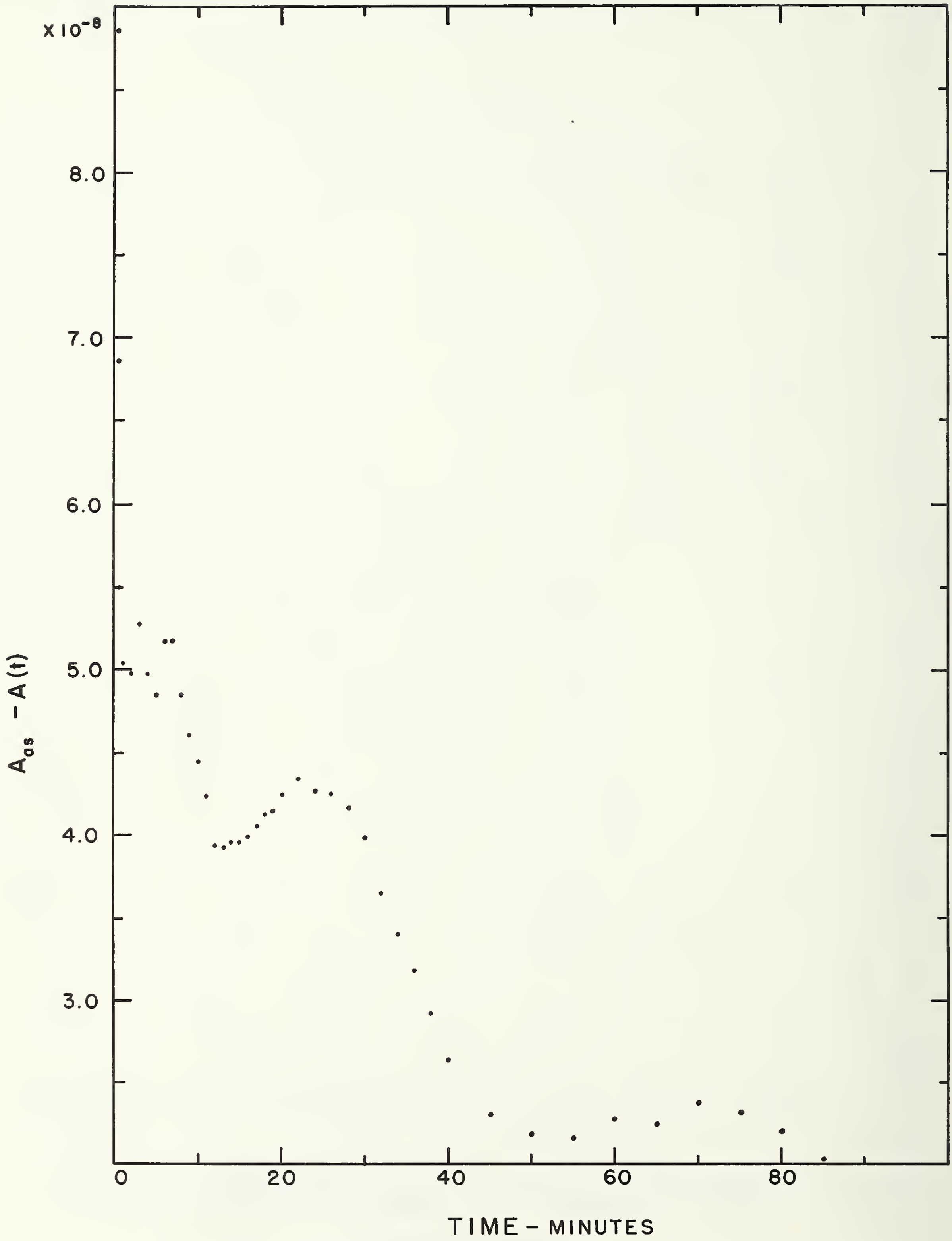

Figure 10. Variation with time of $(A_{as}-A(t))$ where $A(t)$ is the linear creep compliance term in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$ and $A_{as}$ is the asymptote value of $A(t)$ as t becomes large.

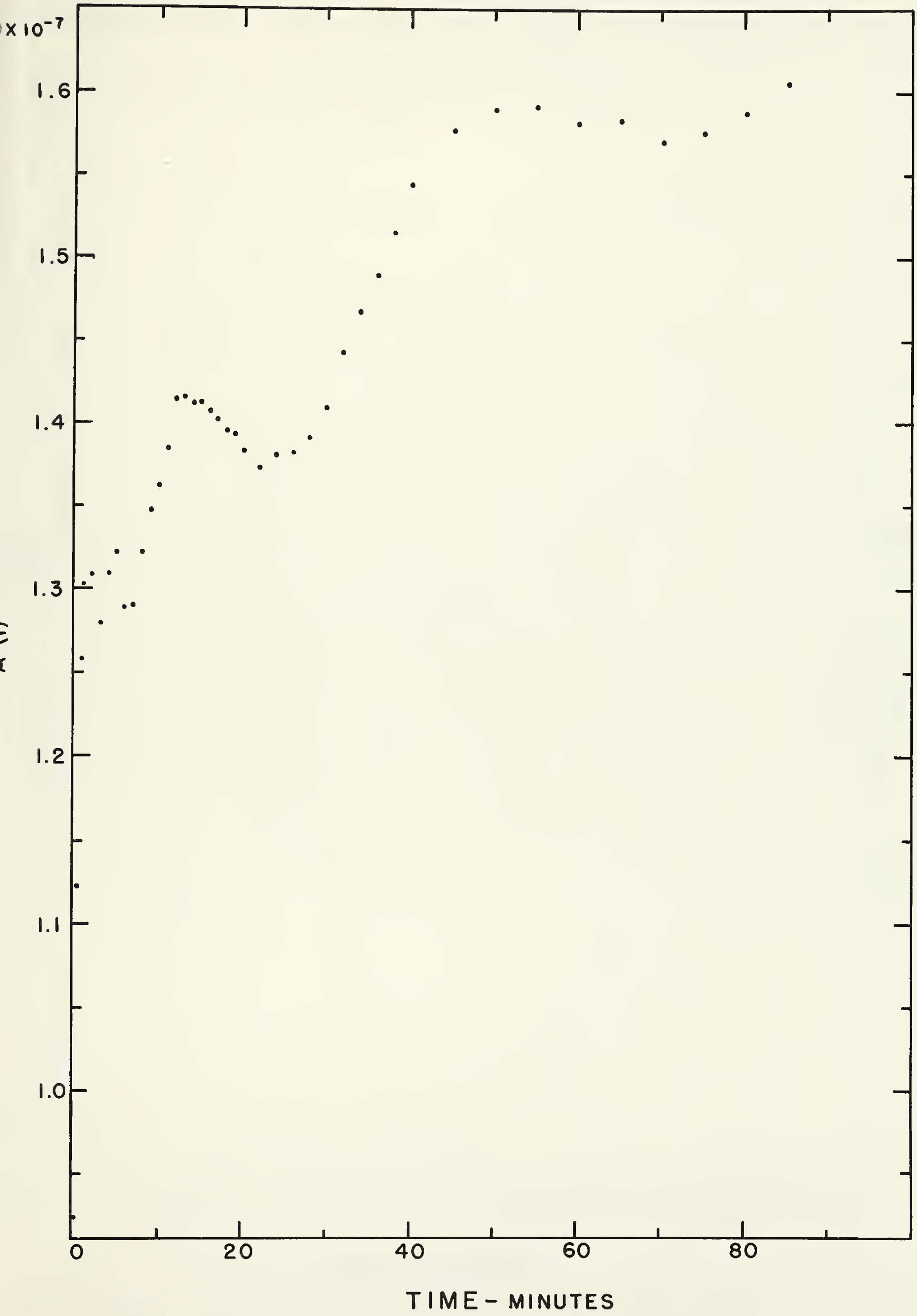

Figure 9. Variation with time of the linear creep compliance term, A(t), in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$.

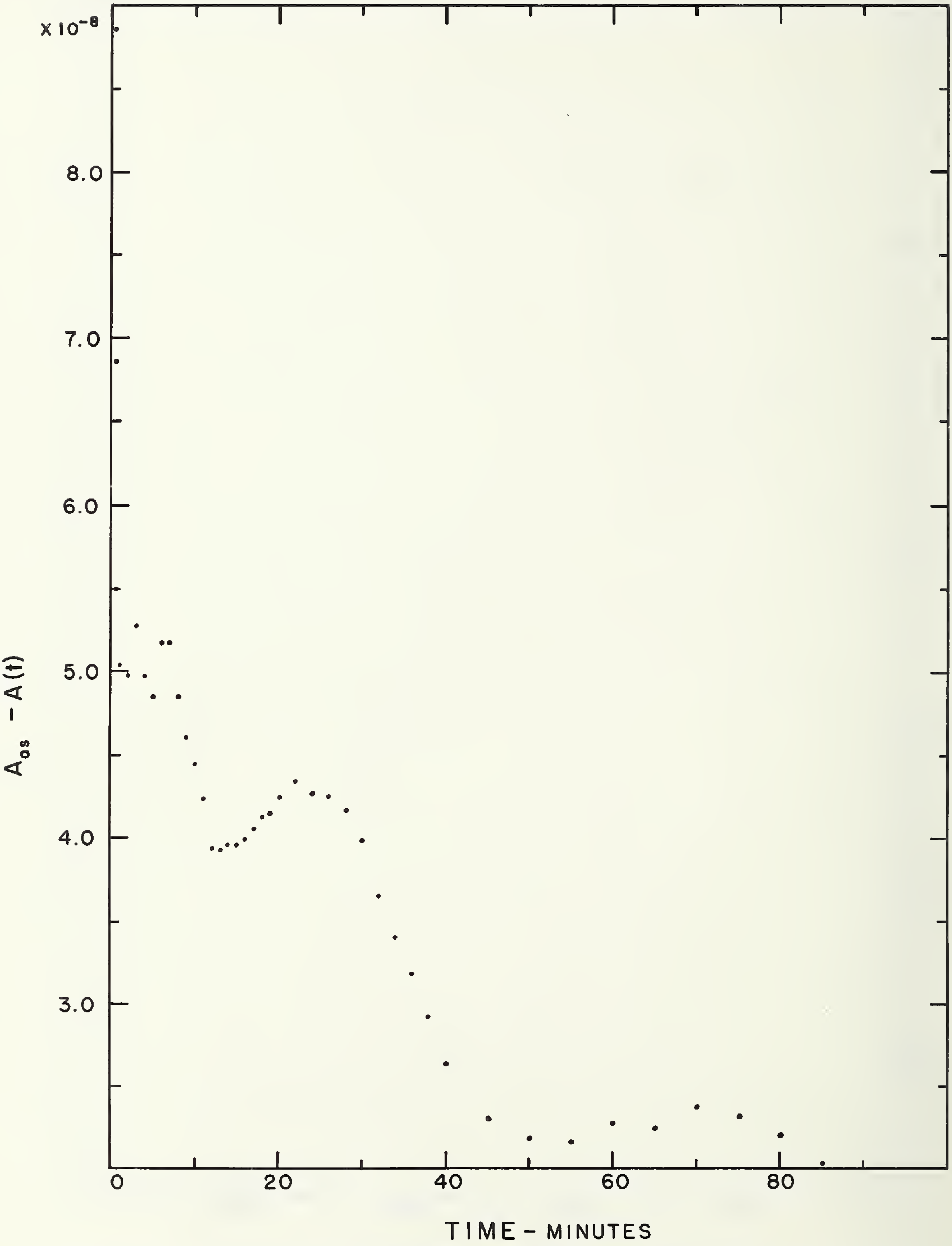

Figure 10. Variation with time of $(A_{as}-A(t))$ where A(t) is the linear creep compliance term in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$ and $A_{as}$ is the asymptote value of A(t) as t becomes large.

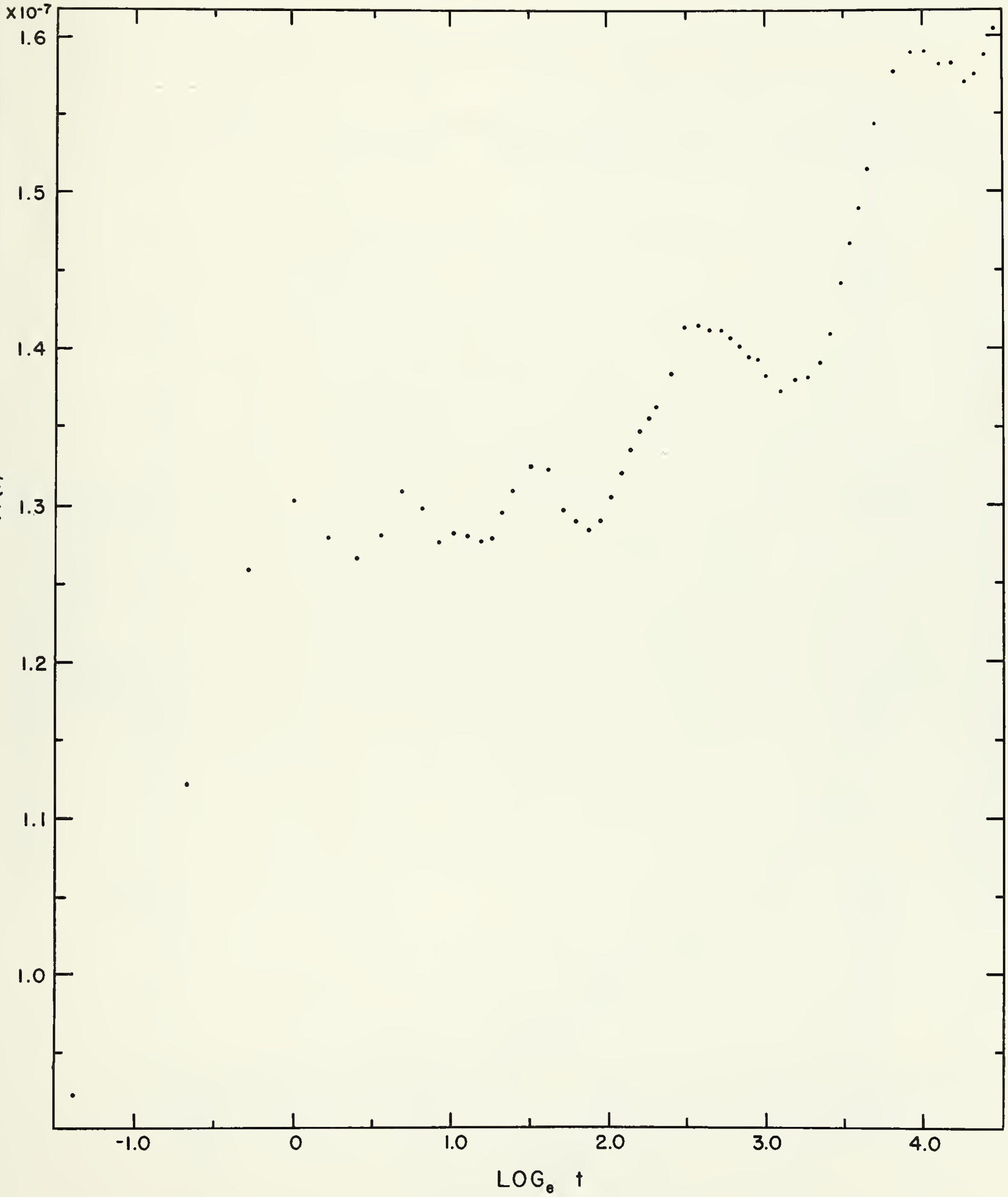

Figure 11. Variation with log of time of the linear creep compliance term, A(t), in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$.

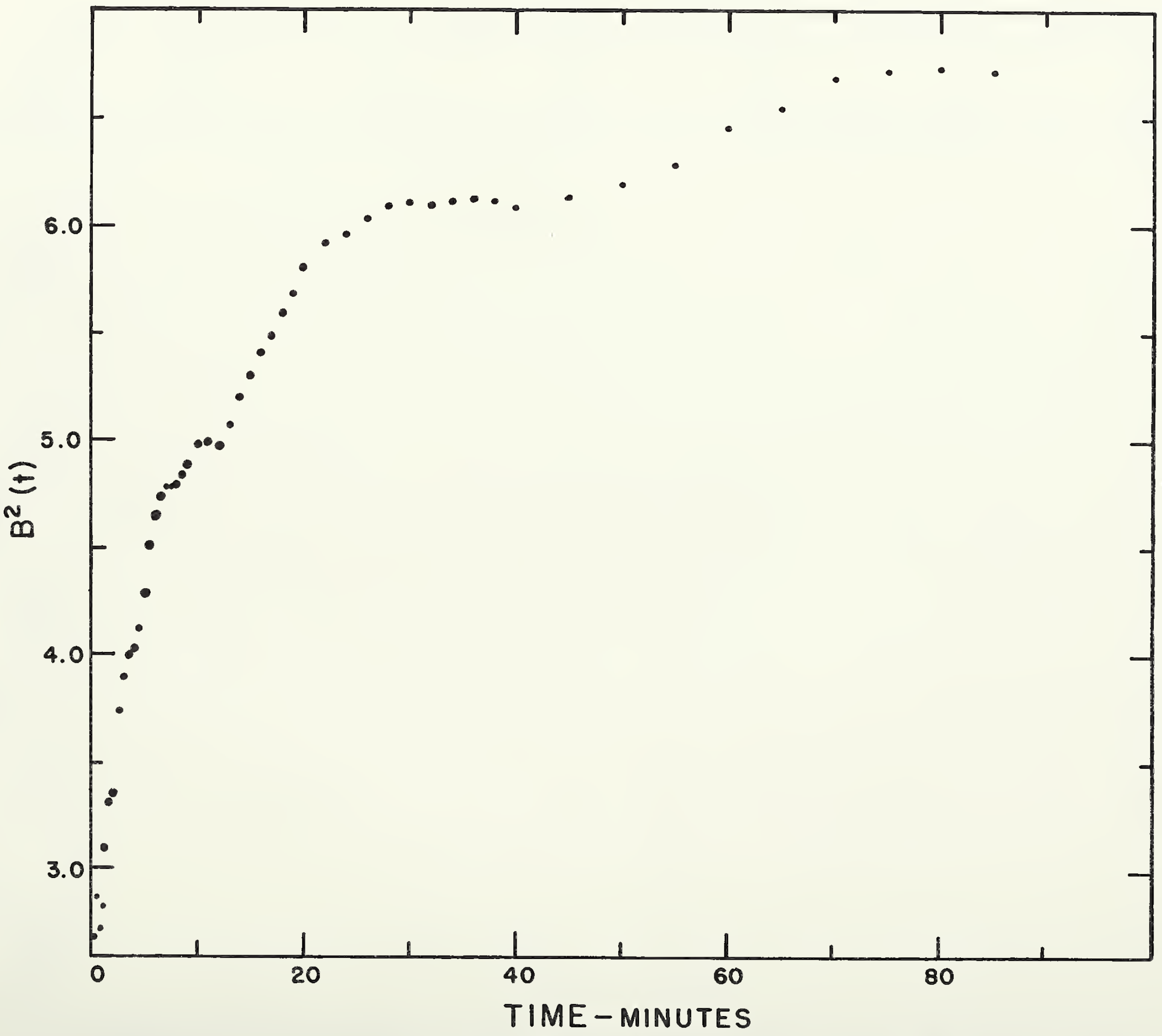

Figure 12. Variation with time of nonlinear creep compliance term, $B^2(t)$, in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$.

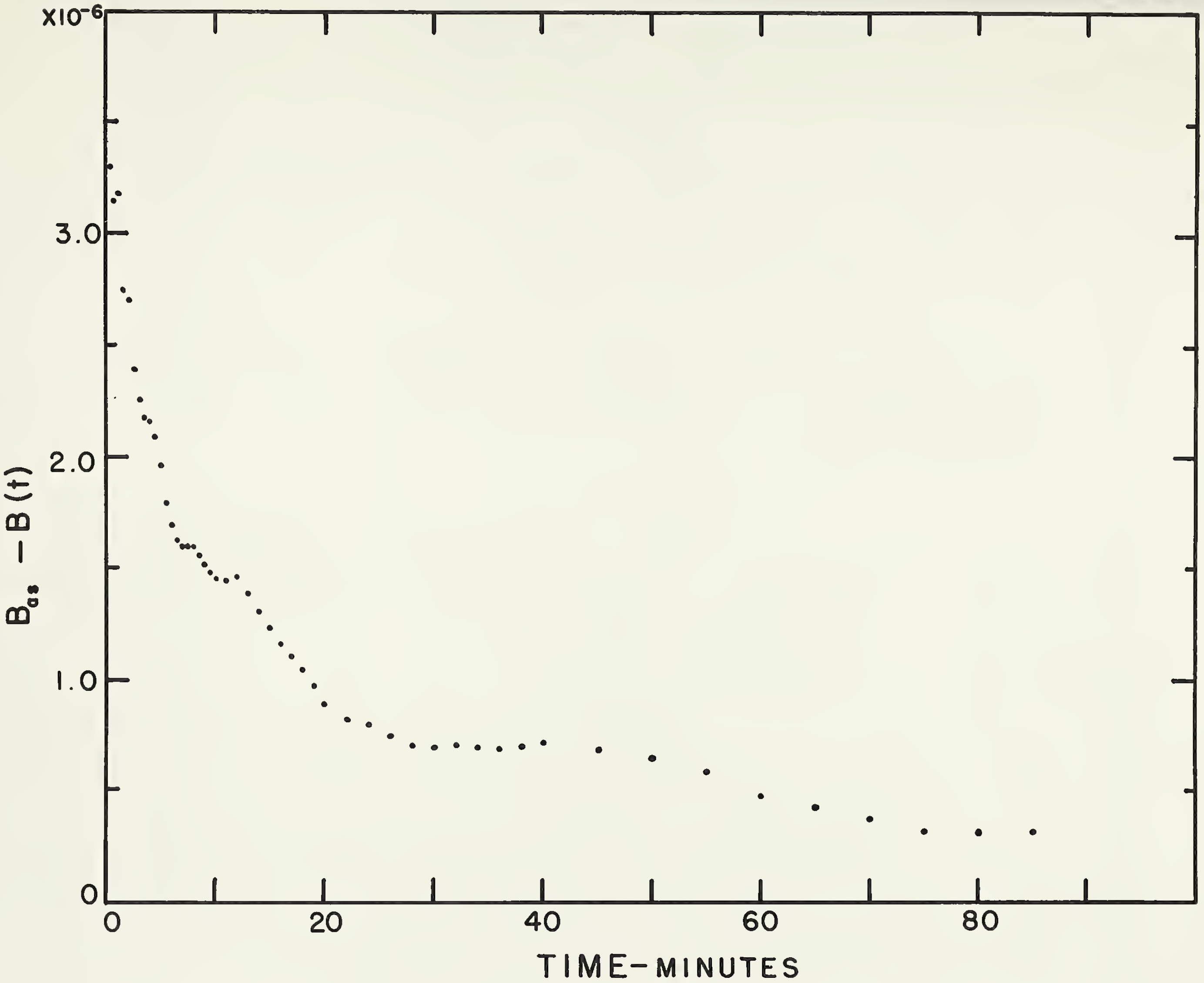

Figure 13. Variation with time of $\left(B_{as} - B(t)\right)$ where $B^2(t)$ is the nonlinear creep compliance term in the equation $\epsilon' = A(t)\sigma + B^2(t)\sigma^2$ and $B_{as}$ is the asymptote value of $B(t)$ as $t$ becomes large.

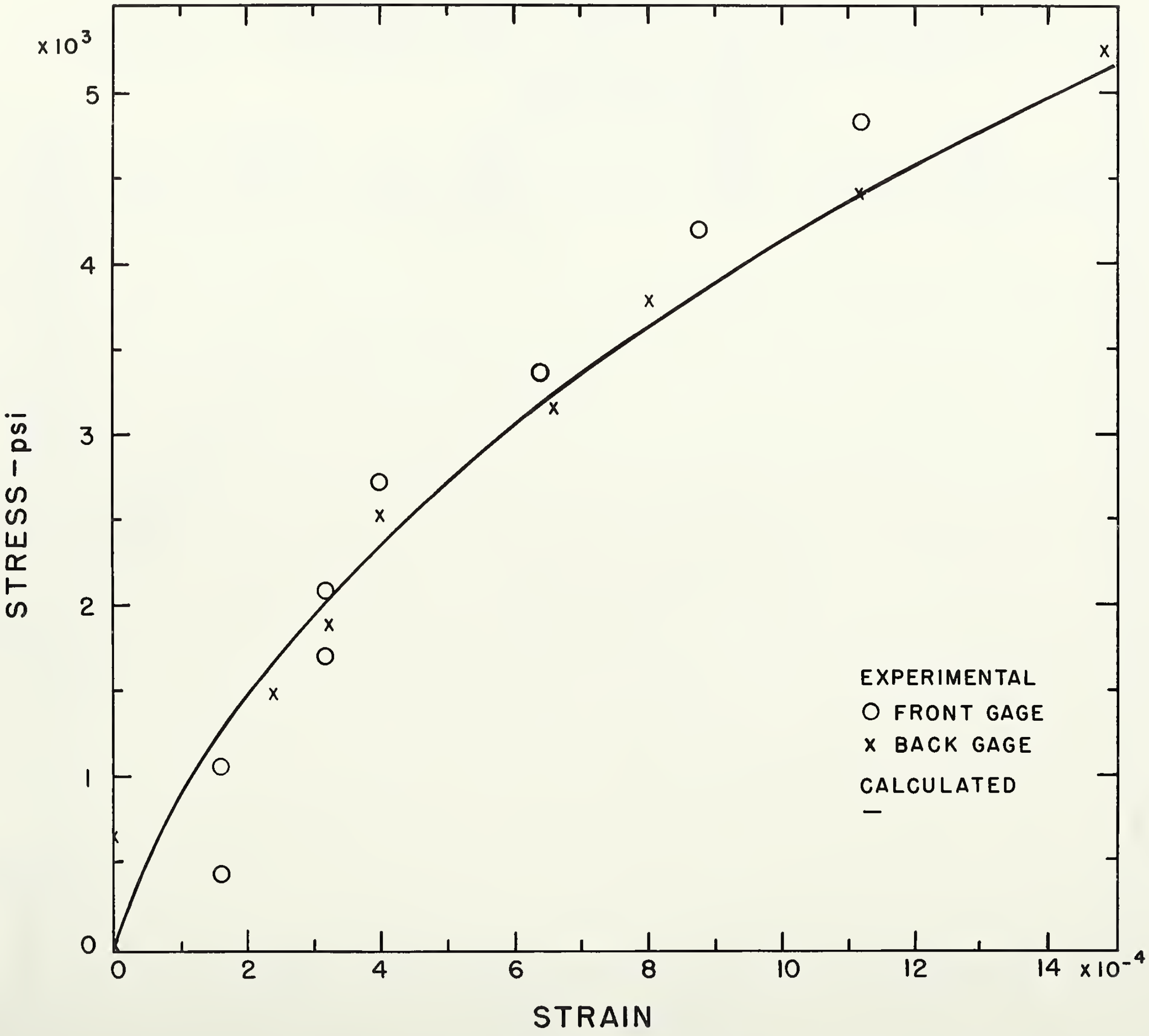

Figure 14. Comparison of calculated and experimental stress-strain curves.