FORGIA INSTITUTE OF TECHNOLOGY OFFICE OF CONTRACT ADMINISTRATION X. ORIGINAL PROJECT ADMINISTRATION DATA SHEET REVISION NO. 7/22/81 Project No. E-19-B06* DATE: School/kak Project Director: Dr. Miroslav Marek Chemical Engineering والمعدية والمعادية المتعادية Sponsor: DHHS/PHS National Institute of Dental Research Type Agreement: ____Grant No. 5-R01-DE03601-09 Award Period: From 7/1/81 To 6/30/82 (Performance) -(Reports) Contracted through: Sponsor Amount: \$44,387 Cost Sharing: \$3,824-4 (E-19-360) Title: Effect of Corrosion on Dental Amalgams OCA CONTACT Duane Hutchison x4820 ADMINISTRATIVE DATA 1) Sponsor Technical Contact: Thomas M. Valega, Ph.D.; Chief, Restorative Materials Program Branch; Extramural Programs, NIDR; Public Health Service; Dept. of Health and Human Services; Bethesda, MD 20205 (301)496-7491 2) Sponsor Admin./Contractual Contact: Robert Ginsburg; Grants Management Officer Extramural Programs, NIDR; Public Health Service; Dept. of Health and Human Services; Bethesda, MD 20205 301-496-7437 Reports: See Deliverable Schedule Security Classification: none Defense Priority Rating: RESTRICTIONS See Attached NIH Supplemental Information Sheet for Additional Requiremen Travel: Foreign travel must have prior approval - Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of \$500 or 125% of approved proposal budget category. GIT; however, we are accountable for all equipment Title vests with Equipment: purchased. and the second second second second second COMMENTS: * Continuation of E-19-B05.

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EFFECT OF CORROSION ON DENTAL AMALGAMS

1. List all publications, not previously reported, resulting from work supported by this grant (author(s), title, page numbers, year, journal or book). List manuscripts separately as submitted for publication or accepted for publication.

2. Provide two reprints of publications not previously submitted to the awarding unit. 3. Progress Report. (See instructions)

Publications

D. B. Mahler, J. D. Adey, and M. Marek. "Creep and Corrosion of Amalgam." J. Dent. Res. 61: 33-35 (1982)

Conference Presentations:

- D. F. Averette and M. Marek: "The Effect of Creep Deformation on Corrosion of Dental Dental Amalgam;
- M. Marek, T. Okabe, M. B. Butts, and C. W. Fairhurst: "Corrosion of the η' (Cu-Sn) Phase in Dental Amalgam";

Presented at the 1982 meeting of the IADR, March 17-21, 1982, New Orleans, LA.

Progress Report

- 1. OBJECTIVES
 - A. <u>Overall Objectives</u>. To improve the understanding of the role of corrosion in the deterioration of dental amalgam restorations by investigating the mechanism of corrosion, the effects of corrosion on the properties, and by developing suitable testing methods.
 - B. Goals for the Current Year.
 - a. To examine the breakdown of the Cu₆Sn₅ phase in mixtures with other phases.
 - b. To examine the effects of aging on the corrosion of high copper amalgam.
 - c. To determine the effect of tensile creep deformation on the corrosion behavior of various amalgams.
 - d. To examine the mechanism of corrosion in the pores in dental amalgams.
- 2. STUDIES AND RESULTS
 - A. A Study of the Breakdown of the Cu $_{6}$ Sn $_{5}$ Phase in Mixtures with Other Phases. Previous studies have shown that when high-copper amalgams corrode the Cu $_{6}$ Sn $_{5}$ reaction phase is primarily attacked. Corrosion tests have shown, however, that the metallurgically prepared, pure Cu $_{6}$ Sn $_{5}$ is highly corrosion resistant. In this study samples were prepared in which crystals of Cu $_{6}$ Sn $_{5}$ were dispersed in various matrices, which included the γ_{1} phase (Ag-Hg and Ag-Hg-Sn), pure Ag, and a mixture of γ_{1} + unreacted alloy (commercial high-copper dental amalgam alloy). The susceptibility to corrosion was evaluated using constant potential coulometry; the electrodes were exposed to 1% NaC1 at 37°C and maintained potentiostatically at 0.0 V (SCE), the current was integrated for 24 hours. Some of the electrodes were examined in the SEM before and after the test.

Section IV (Continued)

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The results confirmed the high corrosion resistance of pure, isolated Cu_6Sn_5 . There was no increase in susceptibility in the matrix of Ag. Susceptibility sharply increased in the matrix of Ag-Hg and Ag-Hg-Sn. The SEM examination showed that corrosion initiated at the interphase. The increase in corrosion susceptibility was attributed to the presence of Hg. This was confirmed in tests in which small amounts of mercury were plated on pure Cu_6Sn_5 ; a significant increase in the corrosion current density was observed.

The study was performed in cooperation with the Medical College of Georgia, where most of the specimens were prepared. All corrosion tests were performed at Georgia Tech. The results were reported at the 1982 Session of the IADR.

B. A Study of the Effects of Aging on Corrosion of High Copper Amalgam. It has been reported in the liberature that the dispersed-phase highoppper amalgam (Dispersalloy) exhibits an increase in corrosion resistance as a function of aging (Sarkar and Greener, J. Oral Rehab. 2, 139 (1975)) This effect was attributed to small initial amounts of the γ_2 phase which is consumed as the amalgam ages. In this study the corrosion behavior as a function of the aging time was examined both for the dispersed phase amalgam, which contains about 12% Cu in the alloy, and for a very high-copper amalgam (Sybraloy), which contains about 30%Cu in the alloy and in which no γ_2 phase can be detected even initially.

The corrosion tests were made using the constant potential coulometry. The aging times ranged from one day to two months. The results have confirmed the previously reported change in the corrosion susceptibility of Dispersalloy with aging; however, the very high-copper amalgam showed even stronger influence of aging on corrosion. The corrosion index (total anodic charge in 24 hours) for Dispersalloy changed from 9.9C to 8.0C (mean values) for aging time one day and two months. For Sybraloy the mean values were 12.3C and 6.4C. A metallographic and X-ray examination of the structure of Sybraloy as a function of aging so far has not revealed the cause of the change in corrosion susceptibility, and the study is continuing.

C. A Study of the Effect of Tensile Creep on Corrosion of Various Amalgams. Both creep and corrosion have been correlated with deterioration of dental amalgam restorations. In this study the relationship between creep and corrosion was examined by subjecting amalgam specimens to tensile loads sufficient to induce creep strain and measuring the corrosion parameters. A constant load pneumatic tensile machine was adapted for the tests; dumbbell-shaped amalgam specimens were placed in a temperature-controlled cell which also contained a reference and counter electrodes. The electrolyte was synthetic saliva at 37°C. Tests were made at various tensile loads. Five commercial amalgams, representing all the major types, were included in the study. The polarization resistance, which is inversely proportional to the corrosion rate, was determined using a transient polarization technique. The results showed that for γ_2 -containing amalgams the creep deformation had a marked effect on polarization resistance, indicating that deformation of restorations enhances the corrosion attack, especially in the regions of margins. This effect has been attributed mainly to the destruction of the passive film on the γ_1 and γ_2 phases by the strain.

Section IV (Continued)

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High-copper amalgams, which exhibit much lower creep rate and do not contain the soft γ_2 phase also showed much smaller effect of tensile load on corrosion.² Anodic polarization curves for loaded and unloaded specimens confirmed the effect of creep; the passive current density increased for each type of amalgam.

The results of the study were presented at the 1982 Session of the IADR.

D. A Study of Corrosion in the Pores in Dental Amalgam.

A study of the effect of porosity on corrosion of a high-copper dental amalgam, performed in the previous year, has shown a significant relationship between porosity and susceptibility to corrosion. In a continuation of this study, the structure of the amalgam in the vicinity of the pores was examined using scanning electron microscopy and energy-dispersive X-ray analysis. The phases observed on the internal surfaces of the pores were then metallurgically duplicated, assembled in galvanic cells simulating the cells in the pores, and the corrosion potentials and currents were measured.

The results have shown that the acidification of the solution in the pores, which results from the corrosion processes, makes the Cu_6Sn_5 more susceptible to the attack; the γ_1 phase containing tin also showed anodic activity. This study was performed before the results of the study on the effect of mercury on the corrosion of Cu_6Sn_5 were known; therefore, the effect of mercury was not taken into account.

3. SIGNIFICANCE

Understanding of the causes of deterioration of the restorations is necessary for the improvement of their performance.

4. RESEARCH GOALS FOR THE COMING YEAR

A. Determination of the Effect of Mercury on Corrosion Susceptibility of Phases in Dental Amalgam:

The effect of Hg on corrosion of $Cu_5 Sn_5$, found in the past year, raises the question of a similar effect on other phases, specifically $Cu_3 Sn$ and $Ag_3 Sn$. The data in literature were obtained without regard to the possible effect of Hg.

B. Quantitative Determination of the Relationship between Strain and Corrosion Parameters:

The data obtained to date related corrosion parameters to load. In the follow-up the strain will be measured and the relationship between strain and corrosion determined.

C. Investigation of the Effect of Aging on Corrosion of High-Copper Amalgams: Further effort will be made to determine the microstructural changes which cause the change of corrosion resistance with aging. A more detailed analysis of the corrosion behavior of aged specimens will be made.