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STUDIES ON THE PENETRATION OF MERCURY THROUGH THE DENTAL HARD TISSUES, USING HG^{203} IN SILVER AMALGAM FILLINGS

by

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A number of histological (*Applebaum* 1929) and spectrographic studies (*Fields* and *Charles* 1950, *Massler* and *Barber* 1953, *Schoonover* and *Souder* 1941) have been made of the problem of resorption of mercury from dental silver amalgam fillings. In view of the somewhat contradictory results, the authors have studied this problem using radioactive mercury.

MATERIAL

Radioactive mercury was obtained by neutron bombardment of mercury metal in an atomic pile (A.E.R.E., Harwell, England). Hg^{203} with a half-life of about 45 days was obtained. The radiations emitted from this isotope are β of a maximum energy of 0.21 MeV and γ of 0.28 MeV. The specific activity was originally about 1.5 mC/g metallic mercury. The chemical state of the radioactive mercury was not known with certainty but since it followed the liquid mercury fraction and was measurably volatilized above 300° C, it was believed that a great part of the radioactive mercury was in metallic form.

The radioactive mercury was mixed with an equal part (w/w) "silver" alloy "68" (Guldsmedsbolaget, Stockholm) in the usual way to form amalgam, which was then introduced into prepared human tooth cavities (*in vitro* and, in two cases, *in vivo*) and in prepared tooth cavities on two monkeys (a total of about 40 fillings *in vivo*) and on one dog (twelve fillings *in vivo*).

ASSAY METHODS

After some preliminary studies, including the use of stripping film autoradiography (*Berggren* and *Cederberg* 1952), the authors decided to use apposition autoradiography, Geiger-Müller countings and scintillation countings.

1. Apposition autoradiography

Several types of sections were used. In some cases teeth were cut longitudinally without touching the radioactive filling, which loosened spontaneously. In other cases transverse sections were used. In still other cases the teeth were examined *in situ* after sectioning the whole jaw.

The autoradiographic exposures were undertaken under moderate pressure, which did not cause pressure artefacts of the film. The exposure times varied from 5 days up to 30 days. Gevaert Dentus Rapid film was usually employed.

A schematic outline of one type of autoradiographic exposure is indicated in fig 1. T is the tooth, with pulp, P, partly retained, and the prepared cavity, C. Ph is the photographic emulsion. The direct exposure in the cavity is denoted E_c and the total exposure in the central pulp region is denoted E_p. The latter is mainly composed of two parts, the direct exposure from Hg^{203} in the pulp, E_d, and the indirect exposure from secondary electrons excited by gamma rays from the cavity, C. The indirect exposure is denoted E_i. Some calculations are essential to understand the contribution of E_i to the total exposure E_p.

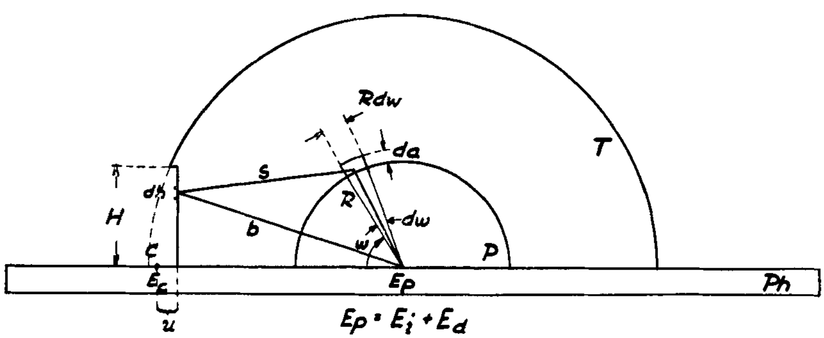

Fig. 1. Schematic outline of one type of exposure in apposition autoradiography. T is the tooth (cross section), P the pulp cavity, C the artificial cavity and Ph the photographic emulsion. Other notations stand for distances, angles and points.

Let us assume that an element dh in the bottom of the shallow cavity emits Ndh beta particles resp. gamma rays during the time of exposure. The exposure is here defined as the number of incident electrons or beta particles per cm^2 of emulsion surface. The exposure at a point u cm from the bottom of the cavity is then

$$E_c = \int_0^H \frac{N(h)\, dh}{4\pi(h^2 + u^2)} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

In calculating the secondary radiation emitted by the gamma rays, let us consider a small element in the dentin of the volume dadcRdw (see fig. 1). μ is the effective absorption coefficient for gamma rays from Hg^{203}. s is the distance from dh to the scattering element dadcRdw. We integrate over the whole prepared cavity and the whole "effective layer" of dentin:

$$E_i = \int_0^H \int_0^{w_2} \int_R^{R+a_0} \int_{-c}^{+c} \frac{N(h)\, dh}{4\pi s^2} \exp[-\mu(s)\, s] \frac{da\, dc\, (R+a)\, dw}{4\pi(R+a)^2} \cdot f \quad (2)$$

In this equation

$$s^2 = (R+a)^2 + b^2 - 2b(R+a)\cos\left(w - \arcsin \frac{h}{b}\right) \quad . \quad . \quad . \quad . \quad (3)$$

and

$$f = \mu \cdot \gamma_e / \gamma_{tot} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

In eq. (4) the quotient γ_e / γ_{tot} expresses the fraction of interacting gamma quanta giving rise to "effective" secondary electrons.

The formulas given above are only approximate, such factors as the angular distribution of the emitted secondary electrons, their energy distribution, the longitudinal extension of the pulp cavity, the variation in film sensivity with the angle of incidence, etc., have not been accounted for sufficiently.

In order to obtain reasonable numerical values, measurements of the geometrical factors were performed on exposed teeth and autoradiographs and absorption coefficients were obtained according to the work of *Leicester* (1949) on the elementary composition of dentin and that of *Bethe* and *Ashkin* (1953) on the gamma ray absorption coefficients.

If the numerical values $b_0 = 0.5$ cm, $a_0 = 0.02$ cm, $H = 0.1$ cm, $f = 0.1\ \text{cm}^{-1}$, $R = 0.1$ cm, $w_2 = \pi$ and $\mu(s) < 0.3\ \text{cm}^{-1}$ N(h), $\pm C = \pm\ 0.1$ cm, and expresses an even distribution of Hg^{203} on the bottom of the cavity, we obtain by approximate evaluation of the integrals

$$E_i/E_c \approx 10^{-3} \qquad (5)$$

Taking the exponential relation between density and exposure

$$D = D_{max}[1 - \exp(-qE)] \qquad (6)$$

the obtained slight exposure over the pulp (see fig. 2) which is obtained experimentally, may be partly or wholly explained by the presence of secondary photo- and Compton electrons from the dental walls of the pulp cavity, since the experimentally found value of E_ρ/E_c = about 10^{-3}.

Neither the integrals in eq. (1) and (2) nor the exposures obtained from eq. (6) can be obtained with sufficient accuracy to permit any reliable quantitative conclusions. For a more detailed interpretation of quantitative geometrical factors in autoradiography, see the paper by *Odeblad* (1952).

Fig. 2.

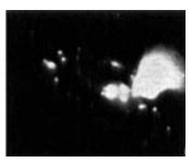

Fig. 3.

Fig. 2. Autoradiograph of cross section of the root of a tooth. White parts indicate radioactivity. A faint exposure is seen corresponding to the root canal and the periodontal tissue.

Fig. 3. Autoradiograph of longitudinal section of a tooth made with the geometrical relationships indicated in fig. 1. White parts indicate radioactivity. The very intense exposure from the artificial cavity is seen and the faint image corresponding to the pulp. Small spots of exposure correspond to radioactive particles, probably dust, containing Hg^{203}. One finding in support of this is the presence of such spots also in regions corresponding to dentine and enamel.

Another difficult technical and theoretical problem arises from the presence of radioactive particles (exemplified in fig. 3) which are probably accidentally introduced as dust from the cutting and grinding operations. The beta particles emitted from such radioactive particles, when present in the pulp cavity, undergo inelastic or elastic (nuclear or electronic) scattering and cause a more or less diffuse exposure of the whole pulp region of the film. This scattering problem is extremely difficult to treat quantitatively. The scattering may be theoretically estimated to occur in between 10 and 20 per cent of beta particles incident on dentine tissue. Experimental measurements using a scatterer of Al (which may represent a mean of the dentin atomic numbers) give figures of about 30 per cent for Hg^{203} beta radiation.

Autoradiographs of disc-formed cross sections of roots were more useful because of the absence of the very active artificial cavity. On these autoradiographs faint exposure was often present over the pulp region, which may indicate the presence of small amounts of Hg^{203} in the pulp. Careful interpretation of the autoradiographs reveals, however, signs of the presence of radioactive dust, making quantitative interpretation somewhat questionable.

With regard to the sensitivity of the film used, the detection limit may be estimated at about 10^{-8} mC/mm^3 of wet tissue, corresponding to about 0.01 μg Hg element/mg of wet pulp, provided that no secondary or scattered radiation or other artefacts interfere.

2. *Geiger-Müller measurements*

Geiger-Müller countings were undertaken with a TCG-2 (Tracerlab) tube with an efficiency of 13 ± 2 per cent for a UX_2 standard. The mica window thickness was about 2 mg/cm^2.

Dried squash preparations of dental pulp wih a thickness < 3 mg/cm^2 were made, giving an estimated self-absorption of Hg^{203} beta rays of < 15 per cent. The detection limit of the method was about 12 imp./min. above background (about 35 imp./min.). This corresponds to a detection limit of about 0.05 μg Hg element.

3. *Scintillation countings*

The scintillation counter was a Tracerlab P20 scintillation detector operated at 1 050 volts, feeding a scaler. The set-up was capable of recording gamma impulses of > 0.15 MeV. The efficiency for Hg^{203} gamma radiation under the prevailing conditions was estimated at 12 ± 4 per cent. With a detection limit of 35 imp./min. above background (about 180 imp./min.) about 0.1 μg Hg element could be detected. The organs examined for Hg^{203} gamma radiation were dried and put in a sample holder in front of the crystal.

RESULTS

1. Autoradiographic results

As discussed above, the film exposure in the pulp region may in many cases be partly or wholly explained by secondary or scattered radiation. In a few teeth the autoradiographs may indicate the presence of about 0.02 μg Hg/mg of wet pulp tissue. In most cases no reliable conclusions could be drawn.

2. Results of Geiger-Müller countings

The results on a dog, receiving several radioactive buccal fillings and on which the pulp tissues were extracted from the lingual side, are collected in fig. 4. The pulp tissue of the teeth

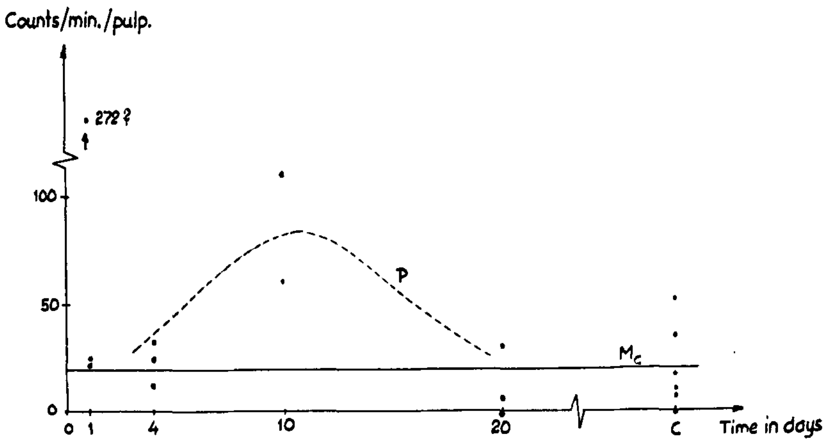

Fig. 4. Relation between the amount of Hg^{203} in dental pulp and time after introducing the radiomercury in the teeth of a dog. Ordinate: counts/min./pulp. Abscissa: time in days. C denotes control values from teeth not receiving radioactive fillings. Mc is the level of the mean of control values. The dotted line P represents the suggested uptake of Hg^{203} in the pulp. The one-day value 272 is probably accidentally high.

with fillings generally contained small amounts of Hg^{203}. The presence of radioactivity in control preparations indicates, however, the risk of contamination. It seems safe to conclude that there is some probability of amounts of Hg well above the detection limit and control level 10 days after introducing the radioactive amalgam fillings.

3. Scintillation counting results

As indicated in Table I, the scintillation countings did not reveal Hg^{203} in some distant organs examined.

Table I

Results of scintillation countings on some distant organs in a dog

Organ	Impulses/minute	μg Hg per organ
Spleen	−5 ± 13	0
Heart	17 ± 12	probably 0
Liver..............	0 ± 12	0
Kidney	−4 ± 13	0
Brain..............	2 ± 13	0

SUMMARY AND CONCLUSIONS

The penetration of mercury from dental fillings was studied with the aid of radioactive mercury (Hg^{203}), using apposition autoradiography, Geiger-Müller countings and scintillation countings.

Some evidence is presented in favour of the importance of secondary and scattered radiation and dust contamination as sources of error in autoradiography. This technique might have revealed the presence of a few hundredths of a μg Hg per mg of pulp tissue in some cases.

Geiger-Müller countings indicated the presence of some tenths of μg Hg in whole pulp 10 days after introducing amalgam fillings.

Scintillation countings did not reveal mercury in kidney and some other distant organs examined.

RÉSUMÉ

ÉTUDES SUR LA PÉNÉTRATION DU MERCURE À TRAVERS LES TISSUS DURS DES DENTS, À L'AIDE DE HG^{203} MIS DANS LES PLOMBS D'AMALGAME D'ARGENT

La pénétration du mercure des plombs d'amalgame dans les dents a été examinée à l'aide de mercure radioactif (Hg^{203}), l'autoradiographie par apposition ainsi que des compteurs Geiger-Müller et de scintillation étant utilisés.

Des preuves furent données en ce qui concerne l'importance du rayonnement secondaire et des impuretés radioactives portées par les poussières, comme sources de fautes lors de l'autoradiographie. Avec cette technique il a été possible de démontrer l'existence de quelque centaine de μg de mercure par mg de tissue de pulpe dentaire dans certains cas.

Le compteur Geiger-Müller montre la présence de quelques dizièmes de μg de mercure sur toute la pulpe dentaire, 10 jours après l'exécution du remplissage d'amalgame.

Le compteur de scintillation ne montra aucune trace de mercure dans les reins ou les autres organes éloignés qui furent examinés.

ZUSAMMENFASSUNG

STUDIEN ÜBER DIE PENETRATION DES QUECKSILBERS DURCH HARTE ZAHNGEWEBE BEIM BENÜTZEN VON HG^{203} IN SILBERAMALGAMFÜLLUNGEN

Die Penetration von Quecksilber aus Amalgamfüllungen in den Zähnen wurde mit Hilfe von radioaktivem Quecksilber (Hg^{203}) untersucht, wobei die Appositionsautoradiographie sowie der Geiger-Müller-Zähler und der Szintillationszähler zur Anwendung kamen.

Nachweis wurde erbracht hinsichtlich der Bedeutung sekundärer Strahlung und durch Staub mitgeführter radioaktiver Verunreinigungen als Fehlerquellen bei der Autoradiographie. Mit Hilfe dieser Technik ist wahrscheinlich das Vorkommen von einigen Hunderten μg Hg per mg Pulpagewebe in bestimmten Fällen nachgewiesen worden.

Die Geiger-Müller-Zählung zeigt das Vorkommen von einigen Zehnteln μg Hg im ganzen Pulpagewebe 10 Tage nach Ausführung der Amalgamfüllung.

Die Szintillationsrechnung wies kein Vorkommen von Quecksilber in den Nieren oder anderen abgelegenen Organen, die Gegenstand der Untersuchung waren, auf.

REFERENCES

Applebaum, E., 1929: Lymph channels in dentine and enamel stained by amalgam. Journ. Dent. Res. *9*: 487.

Bartelstone, H. J., 1954: Radioactive isotopes in dental science. Internat. Dent. Journ., *4*: 629.

Berggren, H., and *I. Cederberg*, 1952: The translucent zone of reaction in the dentine. A radioautographic study. Arkiv för Fysik, *4*: 9.

Bethe, H. A., and *J. Ashkin*, 1952: Passage of Radiation through Matter. In *Segré, E.:* Experimental Nuclear Physics, Vol. I. Wiley & Sons, New York.

Fields, L. B., and *G. W. Charles*, 1950: A spectrographic investigation of trace elements in human teeth. Proc. Okla. Acad. Sci., *31*: 47 (Chem. Abstr. 6715, h).

Leicester, H. M., 1949: Biochemistry of the teeth. Mosby, St. Louis.

Massler, M., and *T. Barber*, 1953: Action of amalgam on dentin. Journ. Amer. Dent. Assoc. *47*: 415.

Odeblad, E., 1952: Contributions to the theory and technique of quantitative autoradiography. Acta Radiologica, Suppl. 93.

Schoonover, I. C., and *W. Souder*, 1941: Corrosion of dental alloys. Journ. Amer. Dent. Assoc. *28*: 1278.

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