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STUDIES ON THE PENETRATION OF ADDED WATER IN HYGROSCOPIC SETTING EXPANSION TECHNIQUE

SOME EXPERIMENTS USING TRITIATED WATER

by

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When additional water or certain other liquids are brought into contact with dental casting investments during setting, an expansion termed "hygroscopic setting expansion" will be observed. This type of expansion is more pronounced than that known as "normal setting expansion" which appears when dental casting investment is allowed to set in air.

Hygroscopic expansion has been the subject of determinations and explanations by a number of authors since *Scheu* (1932) first called this phenomenon to the attention of the dental profession. Recently, *Mahler & Ady* (1960) and *Skinner* (1960) presented reviews of the many investigations and theories concerning the explanation and nature of the hygroscopic setting expansion.

Degni (1946) proposed that additional water causes a swelling of the gypsum gel. *Lyon, Dickson, & Schoonover* (1955), on the basis of their experimental data, believed that hygroscopic expansion is a continuation of the normal setting expansion during the period when the investment is in a semisolid state, and the additional water permits further hydration of calcium sulphate. *Ryge & Fairhurst* (1956) found that hygroscopic expansion can be produced by certain liquids other than water. They did not agree, however, with the theory of further hydration mentioned above. They explained the hygroscopic expansion on the basis of physical characteristics; thus, additional water or other liquids are absorbed by the setting investment because of a reduction in true volume and increase the surface film thickness on inert particles and gypsum crystals, thereby forcing them apart. *Fusayama* (1957) considered that additional water or other liquids are pulled in to fill spaces produced by initial expansion. This liquid supports the space in the manner of a filler and helps the crystal ends grow further. *Mahler & Ady* (1960) proposed that the setting expansion of dental investments is physically contained in the liquid-phase volume by virtue of surface-tension forces. Sufficient water brought into contact with the setting material allows for an outward growth of gypsum crystals into the additional volume provided by this added water, with no build-up of surface-tension forces. In accordance with *Mahler & Ady* (1960), *Jørgensen* (1960) demonstrated a clear relationship between setting expansion and surface-tension of the liquid-phase. With increased surface tension the setting expansion is lowered.

A problem of special interest in this connection is how deep the added water penetrates into the investment mass. The theories of *Degni* (1946), *Lyon, Dickson, & Schoonover* (1955), *Rygge & Fairhurst* (1956), and *Fusayama* (1957) seem to be based on the hypothesis that the added water penetrates the mass uniformly. *Mahler & Ady* (1960), on the other hand, concluded in their proposed explanation that the added water need not penetrate any deeper than the surface of the setting investment. Using dye materials in water solution penetration studies have been published by *Luster* (1957) and *Anske* (1958). *Luster* found just surface penetration of the added liquid into the mass. *Anske* reported that the dye after 30 minutes had penetrated about 6—7 mm into the mass.

The contradictory reports in the literature have encouraged us to a closer study of the penetration of added water in hygroscopic setting expansion technique. It seemed to us that scintillation counting technique and autoradiography using tritium-labelled water (HTO) would, at present, be the most adequate way to demonstrate the penetration of added water into setting dental casting investment. It was decided to study the water penetration under pronounced hygroscopic setting expansion conditions.

MATERIALS AND METHODS

Isotope used

The tritiated water (HTO) was supplied by the Radiochemical Centre, Amersham.

The radioactive isotope tritium disintegrates with the emission of negatrons of 0.019 megaelectronvolt. Its half-life is 12.5 years.

Investing equipment and procedure

The investment used throughout the investigation was Kerr's Cristobalite Investment.

In these studies flexible rubber casting rings were used. The inner dimensions of the rings were 50 · 30 mm. When preparing investment cylinders for autoradiography a glass slide was placed in the rubber cone (see Fig. 1) before the investment was poured into the ring to facilitate a rapid splitting of the set investment into halves. For the autoradiographic study it was necessary to have a plane smooth investment surface in contact with the nuclear

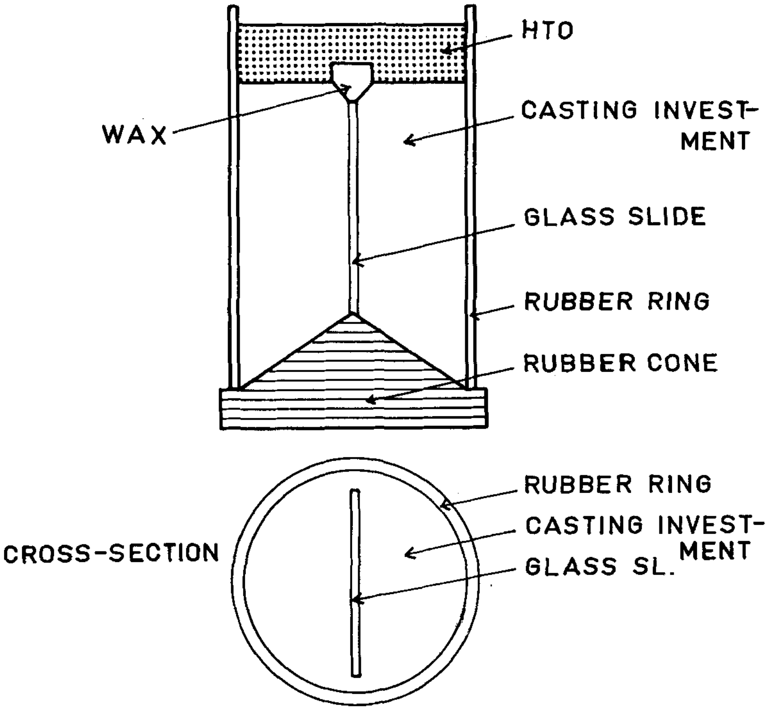

Fig. 1. Test arrangements.

emulsion. The dimensions of the glass slide were in millimeters: 1.5 · 26 · 30. To avoid having the added HTO come into direct contact with the glass slide when it was administered at the top of the investment, the top of the slide was covered with wax (Fig. 1).

The investment powder, 25 g, was mixed with 10 cm³ distilled water (20 °C) in a rubber bowl under vibration, and then the mass was thoroughly spatulated for one minute. The casting ring was then filled with the investment under vibration. The investment was poured along one side into the ring so that it was filled from the bottom up to a level about 10 mm from the top. The casting ring was thereafter placed under vacuum and vibrated for one minute to eliminate air bubbles in the investment mass, consequently reducing the porosities (*Phillips*, 1947).

Five minutes after the beginning of the mixing 2.5 ml HTO (20 °C), corresponding to 500 mC tritium, was added at the top of the investment. After 60 minutes the excess HTO at the top of the set investment was removed and the investment cylinder was gently pushed out of the ring. The cylinders for autoradiography were split into halves along the glass slide.

Autoradiography

In one series of ten investment cylinders the two halves of every cylinder were immediately placed in contact with Ilford G5 nuclear research emulsion plates for one week of exposing. In order to prevent evaporation of HTO during the exposing period the gypsum piece was pressed to the emulsion plate by Scotch tape and embedded in paraffin *ad modum Ullberg* (1961, personal communication). After exposure the emulsion was cut free from the plate, and the gypsum piece with the adhering emulsion was immersed in developing and fixing solutions.

Quantitative measurements

Four gypsum cylinders were cut in one-cm-thick cross sections which were placed in glass bottles with 10 ml toluol. Each gypsum section was then split into very small pieces, and the weight of the section was determined. Scintillation phosphor, 2-5-difenyloxazol (1 g per 300 ml toluol), was added to the bottles, and the amount of tritium in the sections was measured in a tricarb liquid scintillation spectrometer of the type manufactured by *Packard*. The activity was expressed as counts. min. $^{-1}$ · g^{-1} · section^{-1}.

RESULTS

Tritiated water, added at the top of setting dental casting investment five minutes after the beginning of the mixing of the investment mass and allowed to be in contact for 60 minutes, penetrated

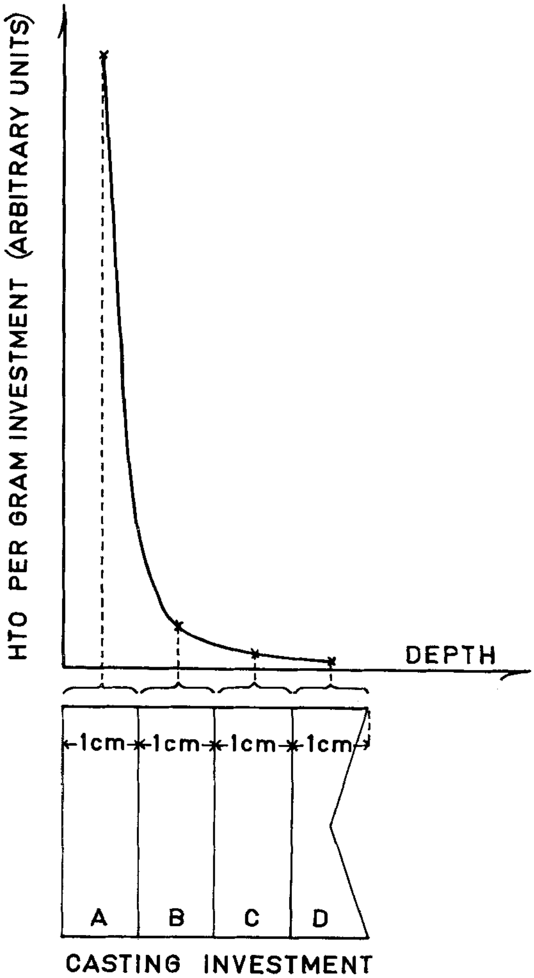

Fig. 2. The concentration of HTO in the set investment cylinder (mean of four cyl.). The concentration was measured with a liquid scintillation spectrometer. HTO was added at the top of the investment five minutes after start of investment mixing. The concentration rapidly decreases from the top to the bottom.

the whole investment cylinder. This was found by means of autoradiography as well as by quantitative measurements in a liquid scintillation spectrometer. The results of these quantitative measurements are plotted in Fig. 2. It was observed that the concentration of HTO per length unit of the investment cylinder rapidly decreased from the top to the bottom. The average concentration of HTO in the first cm at the top (section A in Fig. 2) was about 15 times greater than in the next cm (section B). The autoradiograms showed that the concentration of penetrated HTO in the first few millimeters was very high and then gradually decreased throughout the investment cylinder. The concentration was very low in the layer adjacent to the rubber cone. At the various levels the concentration within each level was the same, i.e. the concentration was the same in the regions close to the ring as in the centre of the investment cylinder.

DISCUSSION

Suffert & Frajndlich (1956) reported that normal as well as hygroscopic setting expansion is greater in an expandable ring than in a rigid ring. Furthermore, the hygroscopic expansion is greater when there is a short time interval between mixing the investment and placing it in contact with water. The expansion was found from the experimental data obtained by *Landgren & Peyton* (1950) to be greatest if the investment was placed in contact with water 2—12 minutes after mixing. To get an idea of the distribution of HTO during pronounced hygroscopic expansion conditions the interval of 5 minutes and flexible rubber casting rings were used in the present investigation. It should be mentioned that after five minutes the top surface of the investment was still wet.

The results obtained in the present study are contradictory to those reported by *Luster* (1957) and *Anske* (1958). Using dye-methods these two authors found just surface penetration (*Luster*), or a penetration about 6—7 mm into the mass (*Anske*) of the added liquid. The divergency between their results and ours is probably due to the difference of the added liquids used. Dye molecules are much larger than water molecules and thus dye materials are not so well suited for penetration studies of this kind.

The theories on hygroscopic setting expansion presented by *Degni* (1946), *Lyon, Dickson, & Schoonover,* (1955), *Ryge & Fairhurst* (1956), and *Fusayama* (1957) seem to be associated with the hypothesis that the added liquid penetrated the investment mass uniformly. The results of the present study do not support such a hypothesis either, as it was found that HTO certainly penetrated throughout the investment mass, but that the concentration of HTO rapidly decreased from the top to the bottom of the investment cylinder.

A theory that is compatible with the results obtained in the present investigation is that proposed by *Mahler & Ady* (1960). According to the latter authors, the mechanism of expansion, under both normal setting and hygroscopic setting conditions, is a result of the same phenomenon, viz. the outward growth of gypsum crystals. The liquid phase of the setting investment is continuous during expansion and adjusts to the addition of water, so that increased volume is available at all points and not just in the area of the added water. As a conclusion of their proposed explanation *Mahler & Ady* (1960) presume that the added water need not and does not penetrate any deeper than the surface of the setting investment material. As mentioned it was, however, shown in the present study that tritiated water added 5 minutes after investment mixing penetrated throughout the investment mass, although it must be emphasized that the concentration of HTO rapidly decreased from the top to the bottom of the investment cylinder. In the deepest layer adjacent to the rubber cone the concentration accordingly was very low.

In this connection it will be noted that under normal setting expansion conditions, i.e. setting in air without added liquids, there is an evaporation of water from the mass (Fig. 3). This evaporation was prevented when oil was added at the top of the investment. It therefore seems logical that the hygroscopic expansion noted when oil is added to a certain extent may be due to prevention of evaporation of water from the investment mass. The hygroscopic expansion observed when oil is added is smaller than that obtained with added water (*Ryge & Fairhurst,* 1956). It was also observed that the evaporation of water from the investment in casting rings increased when the ring was lined with wet asbestos

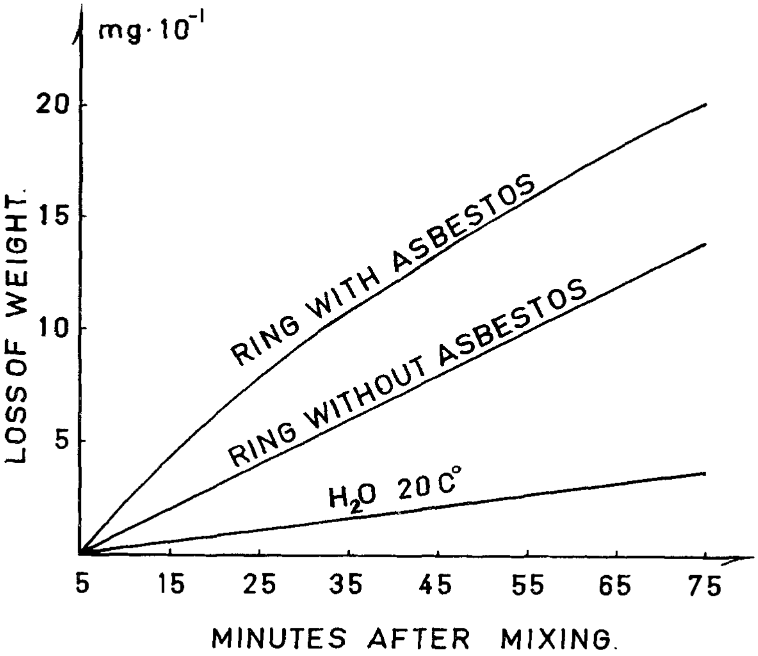

Fig. 3. Evaporation of water from investment during setting under various conditions. The lowest curve illustrates the evaporation of water from a casting ring without investment.

(Fig. 3). It is probable that this greater evaporation was provided by water absorption from the asbestos into the investment mass. In a series of three investment cylinders using casting rings lined with asbestos wet with Na^{22}-labelled saline solution, we found that the radiosodium in the asbestos penetrated into the investment mass when the investment was allowed to set in air.

Some additional experiments were made using a saline solution labelled with the radioactive isotope Na^{22}. In these experiments the radiosodium solution was added at the top of the investment. Autoradiograms of three investment cylinders and scintillation measurements of four investment cylinders gave results in accordance with those obtained using HTO as added liquid.

In Fig. 4 a typical autoradiogram is presented showing the distribution of Na^{22}-labelled saline solution. It can be seen that the concentration of Na^{22} rapidly decreases from the top to the bottom of the investment cylinder. In the layer adjacent to the rubber cone the concentration is very low. Inside the wax pattern the concentration seems to be somewhat higher than outside. White areas correspond to high concentration of radiosodium.

These additional experiments confirm the conclusion to be drawn from the present investigation, viz. that water added at the top of the investment, five minutes after mixing, can penetrate throughout the mass, but that the penetration is not uniform.

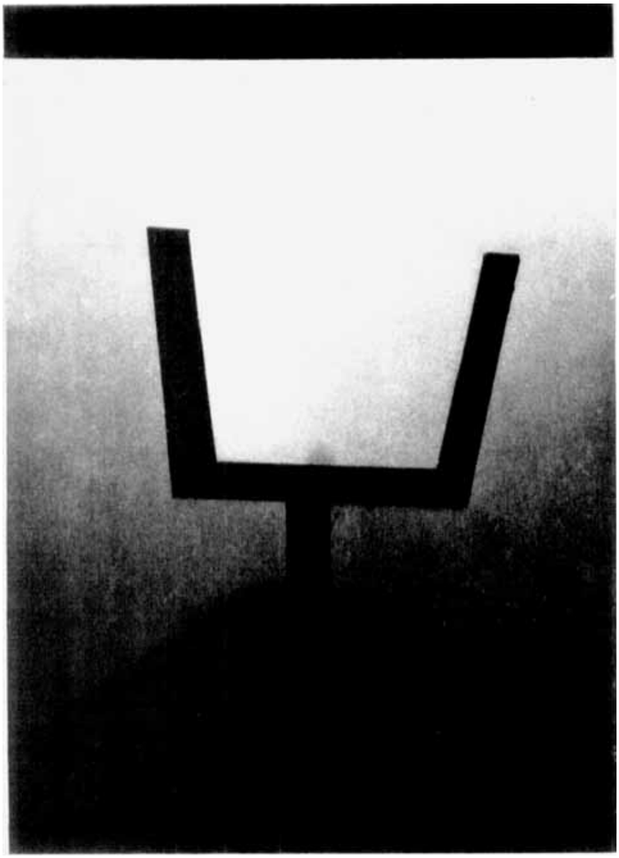

Fig. 4. Autoradiogram showing the distribution of Na^{22}-labelled saline solution in the centre of the investment cylinder. The solution was added five minutes after start of investment mixing. The concentration rapidly decreases from the top to the bottom. Note also the high concentration inside the wax pattern. White areas correspond to high concentration of Na^{22}.

It would be interesting to study with the technique used in this study the penetration of other liquids that have been reported to increase the normal setting expansion, e.g. oils, glycerine and alcohol (*Ryge & Fairhurst*, 1956). It would also be of interest to obtain closer information of the magnitude of hygroscopic expansion on various levels in the investment cylinder. This could perhaps be studied with a method similar to that described by *Asgar, Lawrence, & Peyton* (1958).

SUMMARY

The penetration of added tritiated water (HTO) in hygroscopic setting expansion technique was studied. 2.5 ml HTO was added at the top of the investment cylinder five minutes after the beginning of the mixing of the investment and was allowed to be in contact for sixty minutes. Autoradiograms and quantitative measurements with a liquid scintillation spectrometer showed that the added HTO penetrated throughout the investment mass but the concentration decreased rapidly from the top to the bottom.

Additional experiments with Na^{22}-labelled saline solution as the added liquid gave results in accordance with those obtained when using HTO. The mechanism of hygroscopic setting expansion is discussed.

RÉSUMÉ

ÉTUDES DE LA PÉNÉTRATION D'EAU AJOUTÉE DANS LA TECHNIQUE D'EXPANSION HYGROSCOPIQUE DE PRISE. QUELQUES EXPÉRIENCES AU MOYEN D'EAU MARQUÉE AU H^3

La pénétration d'eau marquée au tritium (H^3) ajouté dans la technique d'expansion hygroscopique de prise a été étudiée. 2.5 ml de tritium ont été ajoutés au sommet du cylindre de revêtement cinq minutes après le commencement du mélange du revêtement et ont été laissés en contact 60 minutes. Des autoradiogrammes et des mensurations quantitatives par un spectromètre de scintillation-liquide ont montré que le tritium (H^3) ajouté a pénétré le plâtre mais la concentration a rapidement diminuée du haut en bas.

Des expériences supplémentaires en employant une solution saline physiologique marquée au Na^{22} comme liquide d'addition ont donné des résultats conformes à ceux obtenus par H^3.

Le mécanisme de l'expansion hygroscopique de prise a été discuté.

ZUSAMMENFASSUNG

UNTERSUCHUNGEN ÜBER DAS EINDRINGEN VON ZUGESETZTEM WASSER BEI DER HYGROSKOPISCHEN EXPANSIONSMETHODE. EINIGE VERSUCHE UNTER BENUTZUNG VON MIT TRITIUM (H^3) GEMEKRTEM WASSER

Bei der hygroskopischen Expansionsmethode wurde das zugesetzte Wasser mit Tritium gemerkt und das Eindringen desselben untersucht:

5 Minuten nach Beginn des Anrührens der Einbettungsmasse wurden 2,5 ccm mit Tritium gemerkten Wassers in den oberen Teil des Einbettungszylinders gegeben und 60 Minuten lang in Kontakt belassen. Autoradiogramme und quantitative Messungen mit einem Flüssigkeits-Szintillationsspektrometer zeigten, dass das zugesetzte Tritium-gemerkte Wasser die Einbettungsmasse durchdrang, wobei jedoch die Konzentration von der Spitze zum Boden schnell abnahm.

Zusätzliche Experimente, bei denen mit Na^{22} gemerkte physiologische Kochsalzlösung als zugegebene Flüssigkeit verwendet wurde, gaben Ergebnisse in Übereinstimmung zu denen mit Tritium gemerktem Wasser.

Der Mechanismus der hygroskopischen Expansionsmethode wird besprochen.

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