Rhodamine B, a fluorescent substance, was used as a tracer to investigate in vitro early microleakage from around amalgam restorations in machinable mica glass-ceramic after thermal stress. Five types of amalgam, i.e., low-copper spherical, low-copper lathe-cut, high-copper admixture, high-copper lathe-cut, and high-copper spherical, were examined in the present study. The results indicated that early microleakage from alloys of lathe-cut particles was lower than that from alloys of spherical particles in both low-copper and high-copper amalgam restorations. A high-copper amalgam with a mixture of lathe-cut and spherical particles tended to exhibit the lowest early microleakage.

Key words: Amalgam, Microleakage, Rhodamine B

INTRODUCTION

Dental amalgams are widely used in the restoration of decayed teeth because of their easy manipulation and of their passable mechanical properties. However, the difference in coefficient of thermal expansion, the lack of chemical bonding, and the shrinkage during setting produce gaps between the cavity walls and the amalgam restoration. The subsequent penetration of fluids, debris, and microorganisms around the margins of the restoration may cause hypersensitivity and secondary caries. Therefore, various test methods have been designed to study marginal leakage around amalgam restorations.

Christen et al.\(^1\) used fluorescent dye penetration method in their tests, because fluorescent dye materials successfully penetrate flaws or crevices in test objects. Fluorescent dyes are useful as tracers, both topically and systemically, since they are detectable in extremely dilute concentrations. In another recent study, Leinfelder et al.\(^2\) proposed the use of calcium hydroxide as an agent for monitoring microleakage and demonstrated that it can be used to detect microleakage in vivo. Isenberg et al.\(^3\) applied this method in vivo and reported a major difference from previous studies in that the detecting agent leaks from within to the outer environment. These studies gave rise to a new laboratory method for the testing of microleakage in amalgam restorations.

The purpose of the present investigation is to describe a new in vitro method for measuring microleakage around amalgam restorations and report the results of its use in experiments on low-copper and high-copper amalgam alloys under thermal cycle conditions.
MATERIALS AND METHODS

Materials

Human teeth are generally considered to be the best material for the sort of the experiment described in this report. When human teeth were used in the preliminary experiment, however, rhodamine B within the filter paper which was placed in the cavity base infiltrated the pulp cavity through dentinal tubules. This made it difficult to measure the exchange of fluid in the gap between the tooth and the restoration. We therefore examined the properties of a machinable mica glass-ceramic* as a potential replacement for human teeth. The chemical composition of this material is shown in Table 1.

First, we observed the cut surface of this material using an X-ray microanalyzer@. A secondary electron image of this cut surface is shown in Photo 1. There were no porosities on the surface of the material. Secondly, we measured its coefficient of thermal expansion using a dilatometer@@. The average value obtained was \(10.4 \times 10^{-6}/^\circ\text{C}\) within the range from 20 to 120\(^{\circ}\text{C}\). This value was similar to that of the human teeth reported by Xu et al.\(^4\). Finally, we examined the cutting property of this material and found that it was cut easily without breakdown. Moreover, with respect to this property of the machinable mica glass-ceramic, Taira et al.\(^5\) have demonstrated that the material can be used for typodont teeth. From these preliminary experiments and reports, we decided to use machinable mica glass-ceramic instead of human teeth in the present study. Three low-copper and six high-copper amalgam alloys were used in this study. Table 2 lists the amalgam alloys used in this study. These alloys were selected not only for their chemical composition, but also for the shape of their particles (spherical, lathe-cut and admixed).

Preparation of test specimen

Cylindrical cavities, 4.5 mm wide and 3 mm deep were prepared in machinable mica glass-ceramic plates measuring 10 mm \(\times\) 10 mm \(\times\) 5 mm. Filter papers** (diameter 4 mm, thickness 0.25 mm) were immersed in a 1% aqueous solution of rhodamine B*** and then dried. These were placed at the base of each cavity before it was filled with amalgam.

<table>
<thead>
<tr>
<th>Chemical composition of a machinable mica glass-ceramic, as specified by the manufacturer (wt %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{SiO}_2)</td>
</tr>
<tr>
<td>(\text{MgO})</td>
</tr>
<tr>
<td>(\text{Al}_2\text{O}_3)</td>
</tr>
<tr>
<td>(\text{K}_2\text{O})</td>
</tr>
<tr>
<td>(\text{B}_2\text{O}_3)</td>
</tr>
<tr>
<td>(\text{F})</td>
</tr>
</tbody>
</table>

---

* MACOR, Ishihara Chem. Ltd., Tokyo, Japan
** Toyo Roshi Co. Ltd., Tokyo, Japan
*** Wako Pure Chem. Ind. Ltd., Osaka Japan
@ JCMA-733, Jeol Co. Ltd., Tokyo, Japan
@@ TMA-8140, Rigaku Co. Ltd., Tokyo, Japan
MICROLEAKAGE OF AMALGAM

Photo 1 Photomicrograph of cut surface of machinable mica glass-ceramic.

Table 2 Materials used in this study

<table>
<thead>
<tr>
<th>Code</th>
<th>Batch No.</th>
<th>Classification</th>
</tr>
</thead>
<tbody>
<tr>
<td>HAM</td>
<td>131151</td>
<td>Low-copper</td>
</tr>
<tr>
<td>LUNA</td>
<td>PX-1</td>
<td>Low-copper</td>
</tr>
<tr>
<td>DM</td>
<td>08712</td>
<td>Low-copper</td>
</tr>
<tr>
<td>DIS</td>
<td>58044L</td>
<td>High-copper</td>
</tr>
<tr>
<td>LUMI</td>
<td>070771</td>
<td>High-copper</td>
</tr>
<tr>
<td>HV</td>
<td>02208714</td>
<td>High-copper</td>
</tr>
<tr>
<td>SP-D</td>
<td>128298</td>
<td>High-copper</td>
</tr>
<tr>
<td>TY</td>
<td>8928602X</td>
<td>High-copper</td>
</tr>
<tr>
<td>SY</td>
<td>011684</td>
<td>High-copper</td>
</tr>
</tbody>
</table>

© Hi-Atomic M, GC Corporation., Tokyo, Japan
© Non Zinc Luna Alloy, GC Corporation., Tokyo, Japan
© Dong Myung, Dong Myung Dental Material Industrial Co., Kyung-KiDo, Korea
© Dispersalloy, Johnson & Johnson Dental Products Company, East Windsor, NJ, USA
© Lumi Alloy, GC Corporation., Tokyo, Japan
© Hi-Veraloy, Hankuk Engelhard Corp., Kyung-KiDo, Korea
© Spherical-D, Shofu Dental Mfg. Co., Kyoto, Japan
© Tytin, The S S White Company, South St., Holmdel, NJ, USA
© Sybraloy, Kerr Manufacturing Company, Roumulus, MI, USA
Table 3 Methods and equipment used in this study

<table>
<thead>
<tr>
<th>Code</th>
<th>Mercury to Alloy ratio</th>
<th>Trituration time (sec)</th>
<th>Amalgamator used</th>
<th>pestle</th>
</tr>
</thead>
<tbody>
<tr>
<td>HAM</td>
<td>0.75</td>
<td>10</td>
<td>$</td>
<td>without</td>
</tr>
<tr>
<td>LUNA</td>
<td>1.6</td>
<td>10</td>
<td>$</td>
<td>with</td>
</tr>
<tr>
<td>DM</td>
<td>1.0</td>
<td>10</td>
<td>$ $</td>
<td>with</td>
</tr>
<tr>
<td>DIS</td>
<td>1.0</td>
<td>15</td>
<td>$ $</td>
<td>with</td>
</tr>
<tr>
<td>LUMI</td>
<td>0.83</td>
<td>15</td>
<td>$</td>
<td>without</td>
</tr>
<tr>
<td>HV</td>
<td>1.0</td>
<td>11</td>
<td>$ $</td>
<td>with</td>
</tr>
<tr>
<td>SP-D</td>
<td>0.84</td>
<td>7</td>
<td>$ $ $</td>
<td>without</td>
</tr>
<tr>
<td>TY</td>
<td>0.74</td>
<td>6</td>
<td>$ $ $</td>
<td>without</td>
</tr>
<tr>
<td>SY</td>
<td>1.0</td>
<td>20</td>
<td>$ $</td>
<td>with</td>
</tr>
</tbody>
</table>

Amalgam pastes were mixed according to the manufacturers’ instructions (Table 3). Amalgam specimens were then condensed into the cylindrical cavities using a hand condenser (diam. 2 mm) with a force of 1.5 to 3.5 kg. Measurement of the condensation pressure was based on the method described by Lussi et al. After condensation, excess amalgam paste was removed using a razor blade. The samples were then stored in an incubator for 24 hours at 37°C with a relative humidity of 100%. Following this, the specimens were immersed in test tubes containing 5 ml of distilled water.

In common amalgam restorations, the space between the restoration and cavity wall is known to be filled with debris from the polishing process. This debris, consisting of amorphous materials, is supposed to improve the marginal seal. Since the present study was to determine pure microleakage, polishing was not performed on the surface of the amalgams.

**Thermal cycling**

Thermal cycling conditions for the evaluation of microleakage from amalgam restorations have varied from study to study. The majority of investigators have used temperatures of 4 and 58 or 60°C. The immersion times used vary from a few seconds to two minutes, and the number of temperature cycles employed ranges from 15 to 2500.

We referred to these studies in deciding the conditions to be used for the current experiment, settling on three daily cyclings using temperatures of 4 and 60°C. However, we discovered through preliminary experimentation that an interval of at least three minutes is necessary to increase the temperature of the test tube solution to 60°C using an indirect heating method. Each thermal cycling thus consisted of ten repetitions of three minutes immersion in a water bath at 4°C, followed by three minutes immersion in a water bath at 60°C. Between cycling, the test tubes were stored in an incubator at 37°C.

**Measurement of rhodamine B**

The amount of rhodamine B was measured using a spectrofluorophotometer with an excitation wave length of 525 nm, an emission wave length of 575 nm and a spectrum band

---

$ HIMIX VS-II, GC Corporation, Tokyo, Japan
$ $ Van-Mix II-M, L. D. Caulk Company, Milford, DE, USA
$ $$$ CAPMASTER, S.S. White Company, Philadelphia, PA, USA
### RF-540, Shimazu Co. Ltd., Kyoto, Japan
width of 5 nm.

As a control, samples of the machinable mica glass-ceramic and set amalgams alone were subjected to the same thermal cycling process as the test specimens to determine whether either contained other fluorescent material. No other spectrum in this region was detected.

Measurement of rhodamine B was performed after one, three, five, seven, and ten days of the immersion period. Five samples of each alloy were tested. Averages and standard deviations were calculated.

**Observation of interface between restoration and cavity walls**

We observed the interface between the amalgam fillings and cavity walls in the machinable mica glass-ceramic by scanning electron microscopy to examine the relationship between the concentration of released rhodamine B and the size of the gap. The specimens which were employed to measure the concentration of rhodamine B were fixed to the machinable mica glass-ceramic with a small amount of adhesive and then sectioned longitudinally. The specimens were ground on 600 and 1000 grid silicon carbide papers under cold water. After polishing, the specimens were washed with distilled water in an ultrasonic cleaning machine for five minutes.

**RESULTS**

The cumulative amounts of rhodamine B found dissolved in the distilled water from the immersion of various amalgams subjected to thermal stress are shown in Figs. 1 to 3.

The results for low-copper amalgams are presented in Fig. 1. HAM showed marked microleakage on the first day, which levelled off thereafter. Although the microleakage from DM was smaller, this alloy showed a similar pattern to NAM. On the other hand, while microleakage from LUNA was slight on the first day, but increased at a steady pace.

---

**Fig. 1** Cumulative rhodamine B dissolution from low-copper amalgams after thermal cycling immersion test. Figures show averages and standard deviations.

**Fig. 2** Cumulative rhodamine B dissolution from high-copper (admixed type) amalgams after thermal cycling immersion test. Figures show averages and standard deviations.
thereafter.

The results for admixed high-copper amalgams are presented in Fig. 2. LUMI showed a pattern of microleakage similar to that of HAM, although the amount was smaller. On the other hand, DIS showed a small amount of microleakage in a pattern similar to that of LUNA. The microleakage from this amalgam after 10 days was the smallest of all the amalgam alloys used in this experiment.

The results for single-composition high-copper amalgams are presented in Fig. 3. TY and SP-D showed almost the same pattern, and the amount of microleakage from these amalgams on the first day was marked. SY showed a pattern similar to those of TY and SP-D, but with a smaller amount. Although the microleakage from HV was slight on the first day, it increased after three days and leveled off thereafter. In Fig. 4, the total amount of microleakage from each amalgam after immersion in distilled water for ten days has been arranged according to copper content and particle shape.

Table 4 shows a comparison of the total amount of microleakage from each amalgam alloy using the two-tailed t-test. Of the low-copper amalgams, HAM, which is composed of spherical particles, showed a greater microleakage than did alloys of lathe-cut particles (LUNA and DM); a highly significant difference was observed between HAM and both LUNA and DM (p<0.01). On the other hand, LUNA and DM showed almost the same level of microleakage, and no significant difference was observed between them. As for the single-composition high-copper amalgams, SP-D, TY, and SY, which are composed of spherical particles, showed greater microleakage than HV, which is composed of lathe-cut particles. A highly significant difference (p<0.01) was observed between the former alloys and HV. The admixed high-copper amalgam LUMI, which is composed of a mixture of spherical particles of different compositions23), showed almost the same level of microleakage as the single-composition high-copper amalgams having spherical particles; and no signifi-
The amount of rhodamine B dissolution from spherical, lathe-cut, and admixed (L+S, S+S) type amalgams after 10 days thermal cycling immersion test. Figures show averages and standard deviations.

Table 4 Comparison of amounts of rhodamine B microleakage from various amalgams at ten days

<table>
<thead>
<tr>
<th></th>
<th>LUNA</th>
<th>DM</th>
<th>DIS</th>
<th>LUMI</th>
<th>HV</th>
<th>SP-D</th>
<th>TY</th>
<th>SY</th>
<th>HAM</th>
</tr>
</thead>
<tbody>
<tr>
<td>N.S.</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>N.S.</td>
<td>**</td>
<td>N.S.</td>
<td>N.S.</td>
<td>N.S.</td>
<td>LUMI</td>
</tr>
<tr>
<td>**</td>
<td>*</td>
<td>*</td>
<td>N.S.</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>DM</td>
</tr>
<tr>
<td>**</td>
<td>**</td>
<td>N.S.</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>N.S.</td>
<td>DIS</td>
</tr>
<tr>
<td>**</td>
<td>N.S.</td>
<td>N.S.</td>
<td>N.S.</td>
<td>N.S.</td>
<td>**</td>
<td>**</td>
<td>**</td>
<td>N.S.</td>
<td>LUMI</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*p<0.05  
**p<0.01
N.S. = not significant (p>0.05)

Significant difference was observed between LUMI and SP-D, TY, and SY. On the other hand, DIS, which is composed of a mixture of lathe-cut and spherical particles, showed little microleakage compared with SP-D, TY, SY, HV, and LUMI, this difference being highly significant (p<0.01).

The amounts of microleakage from alloys of lathe-cut particles (LUNA, DM, and HV) were less than those from alloys of spherical particles (HAM, SP-D, TY, SY, and LUMI) in both high-copper and low-copper amalgams. The differences in the amounts of microleakage between the spherical particle and lathe-cut particle amalgams were less significant (p<0.05),
Photo 2  Photomicrograph of margins between the low-copper amalgams and machinable mica glass-ceramics. A = amalgam, G = machinable mica glass-ceramic.
Photo 3  Photomicrograph of margins between the high-copper (admixed and single types) amalgams and machinable mica glass-ceramics. A=amalgam, G=machinable mica glass-ceramic.
Photo 4  Photomicrograph of margins between the high-copper (single types) amalgams and machinable mica glass-ceramics. A=amalgam, G=machinable mica glass-ceramic.
MICROLEAKAGE OF AMALGAM

or more highly significant (p<0.01). Furthermore, DIS showed significantly less microleakage (p<0.01) than all the other amalgams used in this experiment.

Photo 2, 3 and 4 show secondary electron images of the largest spaces found between the amalgam fillings and the cavity walls on either side of each specimen. Of the low-copper amalgams, HAM, which is composed of spherical particles, showed a fairly large gap between the amalgam fillings and cavity walls. On the other hand, the width of the gaps formed by LUNA and DM, which are composed of lathe-cut particles were smaller than those of HAM. In the high copper amalgams, the width of gaps formed by HV which is composed of lathe-cut particles, and DIS, which is composed a mixture of lathe-cut and spherical particles, were smaller than those formed by SP-D, TY, SY, and LUMI, which are composed spherical particles. In addition, alloys of spherical particles (HAM, LUMI, SP-D and TY) showed many voids within the body of amalgams compared with lathe-cut particles (LUNA, DM, and HV) or of a mixture of lathe-cut and spherical particles (DIS).

DISCUSSION

In the present study, the amalgams labeled HAM, TY, SP-D, SY, and LUMI, which are composed of spherical particles, all showed a similar marked microleakage on the first day. On the other hand, the amalgams labeled LUNA, DM, and HV, which are composed of lathe-cut particles, showed little microleakage; and DIS, which is composed of a mixture of lathe-cut and spherical particles, showed the least microleakage.

The pattern of early microleakage obtained in the present study from the amalgams consisting of lathe-cut particles agrees with the report of Ousley et al.8). Using a radioactive isotope after thermal stress, they reported that the mean cumulative microleakage of low-copper amalgam composed of lathe-cut particles increased for the first seven days and then decreased thereafter. Likewise, Lyell et al.22) using an isotope dye combination method, reported that marginal penetration around a low-copper lathe-cut amalgam by dye and isotope did not decrease at all after 2 week's storage in distilled water or moist air.

In the present experiments, the total amount of early microleakage after thermal stress was highest from alloys with spherical particles, moderate from alloys with lathe-cut particles, and lowest from the alloy with a mixture of lathe-cut and spherical particles. These results are fully or partially consistent with the following earlier reports: Ben-Amar et al.9), using dyes combined with thermal cycling, reported that the least microleakage was observed from an admixed high-copper amalgam, in contrast to spherical high-copper or lathe-cut low-copper amalgams; Ben-Amar et al.10), again using the same method, indicated that a high-copper spherical particle alloy tended to exhibit greater microleakage than did a high-copper admixed particle alloy; Fayyed et al.11), using a dye penetration method with thermal stress, reported that the sealing ability of an amalgam made from low-copper lathe-cut particles was superior to that made from high-copper spherical or blended particles. On the other hand, the findings of Andrew et al.12), using a radio isotope after thermal stress, indicated that the leakage around an amalgam of low-copper lathe-cut particles after 24 hours was almost the same as that around high-copper amalgams. The results of the present study, however, did not support the findings of Smith et al.13), who used dyes combined with
thermal cycling, and reported that the marginal adaptation of an alloy with low-copper spherical particles was greater than that of alloys with low-copper lathe-cut or high-copper spherical particles.

In the analysis of early microleakage of the amalgam alloys in this study, we observed the interface between the amalgam fillings and cavity walls by scanning electron microscopy. The width of the gaps formed by alloys of lathe-cut particles (LUNA, DM, and HV) or of a mixture of lathe-cut and spherical particles (DIS) were smaller than those formed by alloys of spherical particles (HAM, SP-D, TY, SY, and LUMI) in both high-copper and low-copper amalgams. Furthermore, alloys of spherical particles except SY showed many voids within the body of amalgams compared with either lathe-cut particles (LUNA, DM, and HV) or a mixture of lathe-cut and spherical particles (DIS). The interface gaps appear to be enough to permit microleakage to some degree in all specimens examined. These results agree with the findings of Wing et al., who found that a well-condensed spherical particle amalgam was separated from the tooth by a wider space than a similarly-condensed lathe-cut amalgam.

In addition, these results were also supported by the findings of Symer et al., who reported that, in general, lathe-cut alloys adapt better to cavity walls than do spherical alloys. Furthermore, Cunningham found significant differences between the adaptability of lathe-cut and spherical alloys, the former being the more favorable.

It is known that adaptation depends on the plasticity or condensability of the amalgams. The condensability of spherical amalgams is not as good as that of lathe-cut amalgams because spherical particles tend to roll away from the condenser, resulting in gaps or voids and lack of adaptation to cavity walls. The lack of adaptation may result in the ingress of oral fluids into the gaps between the amalgam and the cavity walls. Although we only observed the gaps between the cavity walls and the amalgam fillings by scanning electron microscopy, these observations seem to coincide with the results of the measuring method for microleakage employed in this study, since the size of the gaps appears to be proportionally related to the amount of microleakage.

Fanian et al. published results of an experiment regarding marginal leakage of dental amalgam concluding that marginal leakage of dental amalgams should not be judged solely on the basis of the shape of the particle or composition. However, it appears from the results of the present study that the shape of the alloy particle may affect the marginal leakage of dental amalgams.

CONCLUSIONS

Rhodamine B was found to be useful as a tracer in this study, as it successfully passed through the crevices around amalgam fillings producing leakage that was detectable in extremely dilute concentrations. The sealing ability of amalgams consisting of lathe-cut particles was found to be superior to that of those having spherical particles. An amalgam having a mixture of lathe-cut and spherical particles tended to exhibit the lowest early microleakage.
REFERENCES


FGP 法の記録と適合した歯冠を比較することで、咬合面の修正を行った。

分光蛍光光応用によるアマルガム修復物の初期辺縁漏洩の定量的分析について

金 芝娟、高橋好文、紀藤政司、
森本凱也、長谷川二郎
愛知学院大学歯学部歯科理工学教室

アマルガム修復物の充填初期における辺縁漏洩を定量的に観察するために、分光蛍光光応用ローダミン B をトレーサーとして、また被修復材料としてヒト歯の代わりにマシナブルセラミックを用い、熱サイクルを加えた浸漬条件下で検討を行った。ローダミン B は容易に歯歯とアマルガムの隙間を通過するため、トレーサーとして有用であるとともに、きわめて低濃度の測定が可能です。アマルガム修復物からの充填初期における辺縁漏洩は、低銅型球状、単一組成高銅型球状および混合組成高銅型球状では浸漬 1 日後において著しい辺縁漏洩が観察されたが、低銅型球状、単一組成高銅型球状および混合組成高銅型球状は球状と削片状の混合型は少なかった。浸漬 10 日後の辺縁漏洩はアマルガム合金粒子の形状により異なり、低銅型および高銅型とも削片状合金は球状合金よりも辺縁漏洩は少なく、特に高銅型で球状と削片状の混合組成を有する合金が最も少ない辺縁漏洩を示した。

高周波ろう付け用埋没材の開発

西村文夫*，中村英雄*，高橋英和**，高本敏政*
*東京医科歯科大学歯学部第一理工学教室
**昭和大学歯学部第一補綴学教室

埋没材に導電性を付与し、高周波誘導加熱によって発熱する埋没材の開発を試み、高周波ろう付け用埋没材としての適性を検討した。

マグネシアクリンカーを耐熱材とし、結合材として硬質石膏 5 mass%を配合したものを基本組成とし、これに Co, Fe, Ni の金属粉末を 10～20%添加し、誘導加熱を行ったときの埋没材と被ろう付け金属の時間-温度曲線、埋没材の温度-膨張曲線などを検討した結果、10 mass% Co 添加が最も有効であり、埋没試片の予熱なしで埋没材温度は約 40 秒の誘導加熱により 900 °C に達した。熱膨張曲線は直線的に上昇し、真空加熱時に 1000 °C で 1.25%の膨張率を示した。このろう付け用埋没材を用い、洋銀を銀ろうで真空誘導ろう付けしたときの寸法変化を検討した結果、金属粉末未添加の埋没材に比し、寸法変化の少ないことが認められ、高周波ろう付け用埋没材として有効であると判定された。