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Technical Note

Preliminary Investigations into Using Eugenol to Recover Erased Characters on Polymers

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Abstract: In this paper we present our preliminary studies on the efficacy of eugenol for the recovery of erased characters on polymers. We have found that eugenol rapidly recovers erased characters when applied to the surface of polymer substrates. By applying eugenol to the surface to be treated using a cotton bud or paintbrush, we were able to revitalize erased characters from a range of polymers, including acrylonitrile butadiene styrene, acrylic, high-impact polystyrene, and polystyrene. Eugenol is a safe, nonhazardous, and easily sourced reagent for this purpose.

Background

The recovery of erased characters on metal surfaces via chemical and physical treatments for forensic purposes is well established. However, polymer surfaces represent a particular challenge for forensic scientists, and procedures are less well known. Approaches that have been reported include heat treat-
ment of the polymer object using a heat gun, various visible and infrared light sources [1-3], solvents, swelling agents, and relief polishing [3]. The results from these approaches are highly variable, depending on the identity of the polymer. They use potentially hazardous reagents and may be destructive to the evidence [2, 3]. There is also a commercial kit (RESTO-PLAS, Sirchie Fingerprint Laboratories Inc.) available that involves treatment with a proprietary compound with subsequent heat treatment. This reagent was not tested in these studies.

In 2004, Katterwe reported the serendipitous discovery that clove powder reacted with the surface of acrylonitrile butadiene styrene (ABS - an acrylonitrile-styrene co-polymer containing finely dispersed particles of butadiene rubber), revealing previously concealed damage to the polymer surface [4]. He then demonstrated that a poultice of damp clove powder could be used to recover erased characters on ABS [4].

Cloves are the dried flower buds of the clove tree (*Syzygium aromaticum*) [5, 6] and have been used since ancient times for medical and culinary purposes. Oil of clove or clove oil is the essential oil obtained from cloves and makes up 15 to 18% by weight of the dried cloves [5, 6]. In addition to its use for medical and culinary purposes, it is used for microscopy [5]. Clove oil is a pungent smelling, colorless to yellowish liquid when fresh. The main component is the compound eugenol (4-allyl-2-methoxyphenol), which is present at levels of 82 to 87% in the oil [5]. Other minor constituents include eugenol acetate; caryophyllene; and low concentrations of furfural, vanillin, and methyl amy1 ketone [5, 6]. Katterwe suggested that eugenol was the active ingredient in the clove powder but did not present any data to support this postulate [4].

In this article, we present our preliminary studies on whether eugenol is indeed the active component and the effect of eugenol on erased characters on a range of polymers.

**Materials and Methods**

**Materials**

Preliminary trials were carried out on polystyrene samples obtained from cut-up compact disc (CD) cases (identified by infrared spectroscopy). Acrylonitrile butadiene styrene, acrylic
(ACR), high-impact polystyrene (HIPS), poly(propylene) (PP), and poly(vinyl chloride) (PVC) were all supplied by EFM Plastics (North Geelong, Australia).

Eugenol (99%, Aldrich) was used as supplied. Clove powder was obtained either preground (McCormick Foods, Australia) or was freshly ground from whole cloves.

**Preparation of Stamped Samples**

Die stamping was carried out using 6 mm stamps and a spring-loaded handle (Sontax automatic punch and letter set, Bunnings Warehouse, Australia) with adjustable tension. The stamping was conducted at room temperature either with the handle set at the highest tension or with the tension spring removed and the handle hit with a hammer. Each sample was stamped with three randomly chosen characters, which were manually erased with sandpaper (#80 grit aluminium oxide, Pragar Tools painters roll, Bunnings Warehouse, Australia). Erasure continued until the characters were no longer visible to the naked eye.

After stamping and erasure, the samples were set aside for at least 48 hours before treatment to allow for stress relaxation within the sample [4].

**Recovery of Characters**

Treatment with clove powder followed the method of Katterwe [4]. Briefly, clove powder was applied dry or as a damp poultice (with varying levels of water content) to the sample surface to give a layer approximately 2 mm in thickness. To produce the poultice, clove powder was mixed with water in a grinding bowl until the appropriate paste was created. Samples were left in a glass container to exclude drafts for a minimum of ten hours before removal of the clove powder and inspection.

Eugenol was applied either with a cotton bud (Farmland brand, Coles Supermarket, Australia) or with a paint brush (577 Francheville bristle size 0 flatbrush, Bunnings Warehouse, Australia). For treatment with the cotton bud, the bud was soaked in varying amounts of eugenol and applied over the obliterated region. Treatment was similar using the paintbrush. Samples were inspected after varying times of development.
Observations and Interpretation

Recovered characters were observed and assessed with the naked eye. Images were recorded using a Video Spectral Comparator VSC-1 (Foster and Freeman, XTEK, Melbourne, Australia) and oblique lighting using a Crime Lite forensic light source (Blue: 430-470 nm, Foster and Freeman, XTEK, Melbourne, Australia).

Visual observations of recovered characters were assessed according to the classification system outlined in Table 1.

<table>
<thead>
<tr>
<th>Observable results of three characters</th>
<th>Score out of ten</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 perfectly recovered numbers</td>
<td>10</td>
</tr>
<tr>
<td>3 very clear letters</td>
<td>9</td>
</tr>
<tr>
<td>3 clear letters</td>
<td>8</td>
</tr>
<tr>
<td>3 legible letters</td>
<td>7</td>
</tr>
<tr>
<td>3 hard to read or 2 legible and 1 difficult to read letter</td>
<td>6</td>
</tr>
<tr>
<td>3 difficult to read letters</td>
<td>5</td>
</tr>
<tr>
<td>2 difficult and 1 very difficult to read</td>
<td>4</td>
</tr>
<tr>
<td>3 very difficult to read</td>
<td>3</td>
</tr>
<tr>
<td>2 difficult and 1 not visible/legible or 1 legible, 1 very difficult and 1 not visible/illegible</td>
<td>2</td>
</tr>
<tr>
<td>1 difficult and 2 not visible/illegible</td>
<td>1</td>
</tr>
<tr>
<td>No result or 3 not visible/illegible</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 1

Visual interpretation of recovered characters.

Scanning electron microscopy was carried out with a Phillips XL30 scanning electron microscope equipped with a secondary electron and backscatter electron detector. Samples were viewed using an accelerating voltage of 25 keV with a spot size of 4 and a working distance of 10.2 mm. Samples were sputter gold coated, enabling secondary electron images to be taken.

Results and Discussion

Preliminary experiments were performed to investigate the effect of clove powder (damp or dry) on erased characters on a range of polymer substrates. Katterwe had reported the effect of clove powder on ABS, but had not applied it to other polymers. Our results showed that the phenomenon observed by Katterwe also occurred with other polymers and that, as observed in his work, damp clove powder gave the best results.
Acting on the hypothesis that it is eugenol that is the active ingredient responsible for the recovery of the erased characters, experiments were performed using polystyrene samples sourced from CD cases. Eugenol was applied using a cotton bud, without any excess being removed. Characters became visible within minutes of application. Eugenol was then applied to erased characters on a range of polymers and the results are presented in Table 2. These results are a summary of replicate measurements. Examples of characters recovered from HIPS are presented in Figure 1.

<table>
<thead>
<tr>
<th>Polymer type</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>HIPS</td>
<td>7</td>
</tr>
<tr>
<td>ABS</td>
<td>8</td>
</tr>
<tr>
<td>PVC</td>
<td>0</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>0</td>
</tr>
<tr>
<td>Acrylic</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 2
Recovery of characters using eugenol.

(a)  (b)  (c)

Figure 1
Recovery of characters on HIPS using eugenol. Images taken using VSC-I with object illuminated obliquely using a forensic light source (Crime Lite, Blue: 430-470 nm). (a) HIPS after erasure of characters; (b) HIPS after recovery of characters with eugenol; (c) HIPS control sample to show characters before erasure.

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Investigations were carried out into the method of application of the eugenol comparing a cotton bud versus a paint brush. The results are presented in Table 3. Application with a paintbrush appears to lead to improved visualization; this may be due to more control over the quantity of eugenol being applied to the surface, with treatments with excess eugenol leading to some distortion of the characters (Figure 2). Studies into the best method of application are still underway. However, at this stage, the application of a thin film seems to be the best approach. If this does not produce sufficient recovery, further thin films may be applied to produce the desired results.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Sample</th>
<th>ABS</th>
<th>Acrylic</th>
<th>HIPS</th>
<th>Polypropylene</th>
<th>PVC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cotton bud</td>
<td>a</td>
<td>4</td>
<td>0</td>
<td>5</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>b</td>
<td>3</td>
<td>0</td>
<td>5</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>c</td>
<td>2</td>
<td>0</td>
<td>4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Paint brush</td>
<td>d</td>
<td>5</td>
<td>0</td>
<td>7</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>e</td>
<td>8</td>
<td>6</td>
<td>8</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>f</td>
<td>8</td>
<td>0</td>
<td>8</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 3
Comparison of application method.

Figure 2

Effect of excess eugenol on recovered characters from polystyrene. Images taken using VSC-1 with object illuminated obliquely using a forensic light source (Crime Lite. Blue: 430-470 nm). (a) Undistorted recovered character; (b) Distortion due to excess eugenol.
The mechanism of this phenomenon is unknown at this stage. Katterwe has suggested that revisualization by swelling agents is due to underlying damage to the polymer microstructure. It is possible that the recovery of characters is due to differential adsorption of the eugenol into the polymer matrix. What is clear is that there is a physical effect, not an optical effect, as can be seen from the SEM photomicrograph presented in Figure 3.

Studies are currently underway to determine the nature of this phenomenon and to validate this approach on a wider range of polymer types, character formations, and erasures.

Figure 3

SEM photomicrograph of eugenol-recovered character from HIPS. Viewed using an accelerating voltage of 25 keV with a spot size of 4 and a working distance of 10.2 mm. Sample sputter gold coated.

Conclusions

Katterwe has previously reported that clove powder reacts with the surface of ABS co-polymer, revealing marks that had been stamped on the polymer surface and subsequently erased. The preliminary investigations reported here confirm that eugenol, the major component of clove oil, is the active ingredient. Eugenol is a safe, nonhazardous, and easily sourced reagent for this purpose. Furthermore, eugenol is also effective for the recovery of erased characters on a wider range of polymers, including polystyrene, HIPS, and acrylic polymers, in addition to ABS.
Acknowledgments

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