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EVALUATION OF CLEANING TREATMENTS FOR TARNISHED SILVER: THE CONSERVATOR´S PERSPECTIVE

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Abstract

Several treatments appear in the bibliography to clean the tarnished silver. However, the information about the aspect and efficiency of the cleanings is spread or not completely detailed in these works. With this purpose, a worldwide survey has been carried out to identify of the most common treatments for cleaning tarnished silver and the personal experience of professionals about the frequency of use, the cleaning efficiency, the surface appearance, the application speed, the difficulty and the applicability. This paper contains the results of this survey.

***Keywords**: Silver; Silver Sulfide; Tarnishing; Cleaning treatments; Conservation*

Introduction

Silver is a noble material, which has a good corrosion resistance, however, any slight corrosion on a silver surface has a high visual impact, as it turns a clear shining surface into a dark and dull one. The main responsible for silver alteration in atmospheric environments in museums and collections is sulfur [1]. Gaseous sulfur compounds such as hydrogen sulfide (H_2S), carbonyl sulfide (OCS) and other organic compounds come from external sources, anthropogenic emissions or materials commonly found in exhibition or storage areas such as natural or synthetic rubber (in stoppers, O-rings, and latex gloves), certain paints, and some textiles (e.g. wool or felt) [2]. These compounds attack silver covering its surface with a thin but visible layer of silver sulfide (Ag_2S). Silver sulfide is a very stable compound, which does not compromise the material integrity, but turns surface into a yellowish to black color [3, 4]. For this reason, silver collections need periodical attention by conservators. Renowned institutions such as the Canadian Conservation Institute (CCI) and the Victoria and Albert Museum even give some recommendations for the conservation and cleaning of silver objects [2, 5-10].

There are different methods for cleaning silver available for conservation professionals, mainly mechanical, chemical and electrochemical, plus other more advanced methods such as laser, plasma, etc. which are more restricted [11-13]. Some of these methods have been reviewed and compared in literature [1, 14-20], with a scientific approach. Nevertheless, some aspects relevant for the conservator´s experience have not been fully addressed. On the other hand, conservator´s practices and concerns are of major relevance when designing a scientific

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study on conservation methodologies. For this reason, a worldwide survey on silver cleaning was carried out, to serve as the basis from an evaluation on cleaning procedures. Results of this survey are now published as they may result of interest both for conservators and conservation scientists, and discussed according with the results of scientific studies on cleaning methods

Methodology

The identification of the most used treatments for cleaning tarnished silver was carried out with an online survey to worldwide conservators. The link to the survey was sent by mail, and posted in social media such as LinkedIn or Facebook, in conservation-related groups and webpages. This survey included sections to describe each treatment and their personal experience as well as queries about the frequency of use, the cleaning efficiency, the surface appearance, the application speed, the difficulty and the applicability. The complete questionnaire is presented in Appendix A.

A total of 40 conservators from 17 countries proposed 154 treatments. The majority of the conservators were from USA (23%) and Spain (20%); nevertheless, conservators from Canada, New Zealand, Australia, UK, Belgium, Croatia, Israel, Mexico, Netherlands, Norway, Peru, Portugal, Qatar and Switzerland also participated in the study.

Survey results

Results from the survey were classified into four groups: mechanical, chemical, electrochemical and others. The most popular treatments for cleaning tarnished silver were mechanical treatments (49%), followed by chemical (38%), electrochemical (12%) and other methodologies (1%) (Fig. 1). It is worth mentioning the relatively high use of commercial products, which were applied in ~26% of the treatments, especially for mechanical and chemical cleanings. Commercial products are widely used, mainly for convenience, as they are effective and require no previous preparation. The main disadvantage of using commercial products is that their precise composition is unknown, and may be unexpectedly modified by the manufacturer, changing therefore their effectiveness and aggressiveness. Even, the CCI warns about the application of commercial metal cleaners and coin dips (cloths, wadding, liquids or pastes) to clean historical coins because they can scratch their surface damaging the stamp [7].

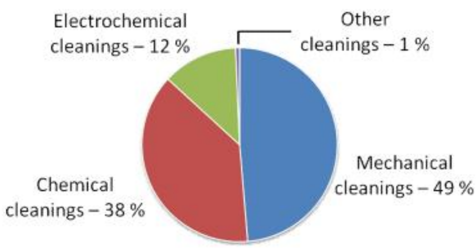

Fig. 1. Nature of cleaning treatments according to the survey results.

Mechanical treatments

According to the survey results, the 50% of the mechanical treatments are based on commercial products (Fig. 2). These products can be liquids, foams, pastes, waddings or cloths and they are usually produced by mixing an inorganic abrasive as SiO_2, Al_2O_3, TiO_2 or $CaCO_3$, with an organic substance (surfactants, soaps, fatty acid…), which keeps the abrasive in suspension and carries away the abraded material [15, 17]. It frequently contains a detergent with surface active chemicals to wet the surface and remove the grease; and tarnishing inhibitors, which are sulfur-containing organic compounds with hydrophobic character that act as a barrier to tarnishing gases slowing the re-tarnishing process. However, it has been observed in a recent study, that remains of these compounds can induce a fast re-tarnishing process after several applications [20]. In less quantity, commercial silver polishes present dyes to help detecting the remains of the product on the silver object; and perfume which cover up the smell of sulfide, result of the cleaning [1, 16, 17].

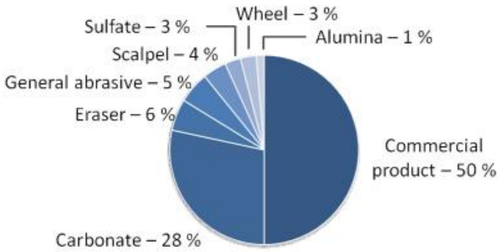

Fig. 2. Mechanical cleaning treatments according to the survey results.

The cleaning efficiency of commercial products is considered good by users in the majority of the cases and they leave an optimal surface appearance, although sometimes could be too shiny. The principal advantages of these products, according to conservator's responses, are the ease of use, and the applicability to small and large objects, even at several objects at the same time. Nevertheless, over a half of the conservators consider that the cleaning speed is medium, while the 22% of the survey responses considered the mechanical cleaning with commercial products as a fast treatment (Fig. 3). Regarding the commercial products, Pre-lim surface cleaner (Neuburg silica chalks in a water/white spirit emulsion produced by Picreator Enterprises Ltd.) is the preferred one (~23% of the commercial products).

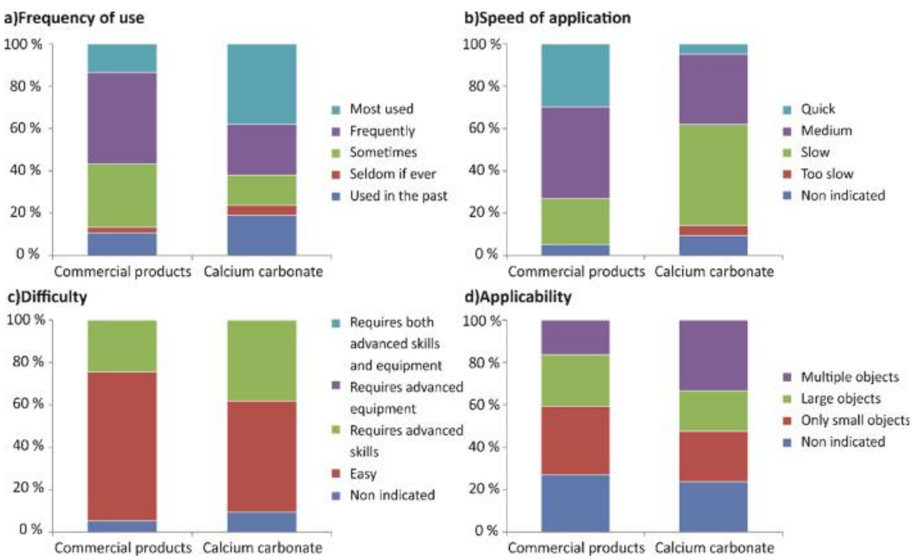

Fig. 3. Personal experience of professionals with mechanical cleaning treatments.

Regarding non-commercial products, the most usual abrasive is calcium carbonate (~28% of the mechanical results, 14% of the total results) (Fig. 2). It is a soft abrasive with similar hardness to silver (Mohs' Scale of silver: 2.5 – 4.0 and $CaCO_3$: 3.0) [16, 21]. Previous studies confirmed that abrasives such as alumina, quartz or hematite can leave scratches on the silver surface due to their higher hardness [15, 17]. To abrade the surface, calcium carbonate is usually dispersed in distilled water. Most of the conservators believe that it is a medium-slow treatment although it is easy to apply (Fig. 3). The CCI has also published a note about the polishing on tarnished silver with $CaCO_3$ [9].

One of the added values of mechanical treatments is the sense of control in the effect of the cleaning material, as one can modulate the intensity and stop when considered, while it is not possible to control the rate and to stop a chemical process with the same immediacy to avoid the damage of the object. But several studies have demonstrated that mechanical cleanings, despite their apparent mildness, can induce significant and uncontrolled mass losses, mainly with rotary tools and hard abrasives, such as multi-purpose metal polishes [8, 17, 19, 20, 22]. In addition, successive cleanings can leave scratches on the surface and, even, alter and loss the surface motifs of the object, such as in coins or medals [7]. The most suitable treatment are abrasive particles with small size to produce scratches which can scatter the visible light and, hence, not be visible to human eye [16]. Wharton et al. suggested to polish with a less abrasive material for a longer period of time than a more abrasive material for a shorter period of time to avoid the damage of the object [15], they also recommended to use deionized water with a small amount of non-ionic surfactant as carrier fluid, instead of ethanol because it can evaporate or spread into the cloth increasing the aggressiveness of the abrasive [15]. The product application is extremely important since the same polish, applied by two different people using different pressures, may produce different results [17, 23]. The CCI also recommends to test the polishes on an acrylic surface before being used on silver to evaluate the depth and pattern of scratching [1, 8].

Finally, abrasive remains are considered to be easier to clean, as they are visible to the eye, while chemicals are not. However, this sense is misleading because not visible to the eye residues from the multiple chemical products used in the formulation of commercial products might remain adsorbed on the metal surface.

Chemical treatments

The second group of treatments applied by professionals for cleaning tarnished silver is chemical treatments (38%) (Fig. 1). Here the number of different products is much higher, although commercial products are again the most popular ones (~18% of the chemical results, 7% of the total results) (Fig. 4) because they are quick and easy to apply principally on small objects, although can be applied on multiple objects at the same time (Fig. 5). However, conservation professionals also indicated that they were “sometimes” and “seldom if ever used”.

It is important to recall that the two major ingredients in silver dip cleaners are a mineral acid, sulfuric or formic acid; and a sequestering agent, normally thiourea, which can have an undesired effect on the object [16, 24-29]. They often include acid-stable detergents to remove the grease, a colorant to distinguish the liquid from water and a water-soluble perfume to mask the smell of the hydrogen sulfide produced during the cleaning [16]. The evaluation of different commercial products showed that the pieces cleaned with abrasive polishes presented dark tarnish in the deep grooves of the pattern in comparison with the objects cleaned with dip products; however, the latter was more tarnished after six months [17]. This behavior is due to the remains of the cleaning product in the surface which can accelerate the re-tarnishing process leaving the silver with a yellowish color [18-20] due to the decomposition of the complex thiourea-Ag and precipitation of silver sulfide [30].

The most known non-commercial products are the thiourea-acid solutions with mineral acids such as chloride or nitric acid (16% of the chemical results, 6% of the total results) (Fig. 4). Thiourea is a chelating agent of silver which dissolves the tarnished layer on the surface by the Reaction 1 [16, 18].

$$Ag_2S + 2H^+ + 2n[CS(NH_2)_2] \rightarrow 2Ag[CS(NH_2)_2]_n^+ + H_2S \qquad (1)$$

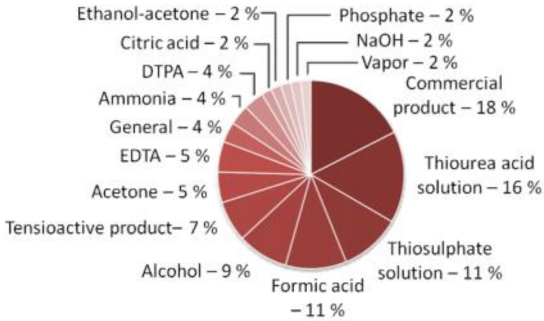

Fig. 4. Chemical cleaning treatments according to the survey results.

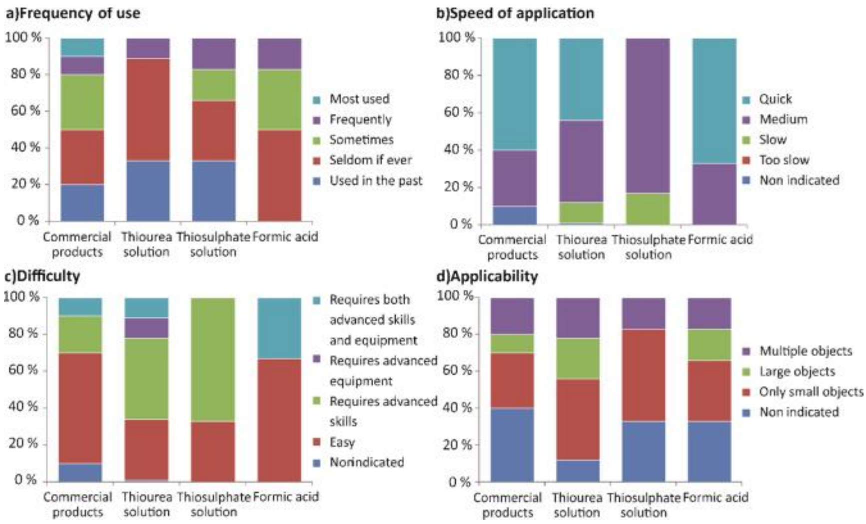

Fig. 5. Personal experience of professionals with chemical cleaning treatments.

It is a fast method, which leaves a bright finish (Fig. 5). It is easy to use, though conservators warned that it needs some skills to avoid damage the objects. In fact, thiourea acid solutions were popular in the past, but now they are scarcely used due to their side effects. Scientific studies have proved that successive treatments can damage the object inducing a dull and etched surface which can severely alter the original surface motifs, engravings, filigrees and gildings [18, 20, 29]. In sterling silver, a preferential attack can occur in the grain boundaries of the alloy and a dark layer of CuS can be formed [22]. Additionally to the effects on the silver objects, thiourea is a suspected carcinogen which can induce dermatitis and increases the sensitivity to UV light due to their good bonding to the skin components [31, 32].

The thiosulfate solution has been applied to remove incrustations of silver chloride and thick corrosion layers on silver objects due to its complexation of silver ions (Reaction 2) [1, 22]. The most common compositions, according to conservator's responses, are solutions of sodium or ammonium thiosulfate 15-30% w/w in distilled water. These procedures are medium-slow ones which require advanced skills from the conservator to avoid the damage of the objects. It is frequently applied to small objects, although it can also apply to multiple objects at the same time. In long-term treatments, a stable thin purple layer can be formed onto the surface due to the dismutation of $Ag_2S_2O_3$ and formation of Ag_2S (Reaction 3) [22, 33].

$$Na_2S_2O_3 + 2\ Ag^+ \rightarrow Ag_2S_2O_3 + 2\ Na^+ \qquad (2)$$

$$Ag_2S_2O_3 + H_2O \rightarrow Ag_2S + SO_4^{2-} + 2 H^+ \quad (3)$$

Formic acid diluted in water has been frequently applied in archaeological silver, mainly on corrosion products such as silver oxides and chlorides [1, 34]. It is an easy and quick procedure, and it can be applied to large objects. However, the final aspect of the object is still slightly tarnished. The cleaning just with organic acids is not effective in eliminating the sulfide tarnishing [19, 20] because Ag_2S is insoluble in water ($Ks = 6 \cdot 10^{-30}$) and just soluble in nitric and sulfuric acids [21]. Formic acid is good for removing copper corrosion, although some damage is observed on the metal surface after being immersed during 24 h [22].

The cleaning treatment may be applied by immersion to avoid leaving marks on the surface. However, if the objects have a complex morphology or present crevices or other materials, the most convenient procedure is to clean with a swab or gel [1].

Electrochemical treatments

Regarding electrochemical methods, the galvanic reduction with aluminum and an alkaline electrolyte is the most common one (48% of the electrochemical results, 6 % of the total results) (Fig. 6).

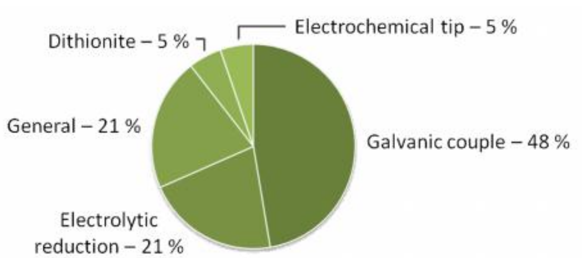

Fig. 6. Electrochemical cleaning treatments according to the survey results.

In this treatment, the less noble metal, aluminum or zinc, becomes the anode and corrodes; while the silver, the noblest metal, remains uncorroded. The tarnishing cleaning is a secondary reaction due to hydrogen formation, which reduces silver sulfide forming H_2S gas according to the reaction 4 [16].

$$H_2 + Ag_2S \rightarrow H_2S + 2Ag \quad (4)$$

This type of cleaning was used sometimes because it is an easy treatment with medium application speed (Fig. 7). The judgment of the final aspect of the object differed between conservators, some thought that it was irregular while other thought it was optimum. This different aspect could depend on the type of silver, the initial state of degradation of the silver object and the time of cleaning. In sterling silver, the electrochemical reduction of the silver sulfide is accompanied by a similar reduction of any copper compounds present. The result is the deposition of metallic copper along with silver on the surface, changing the object color to a copper-reddish hue [16].

The second chosen electrochemical cleaning procedure is electrolytic reduction**.** This type of electrochemical cleaning uses an external source of energy such as a battery, a generator or a potentiostat/galvanostat to reduce the corrosion products on the silver. The most common electrolytes employed by professionals are sodium sesquicarbonate, NaOH and $NaNO_3$. The electrolytic reductions are only reported to be applied "sometimes" because they require advanced equipment and skills (Fig. 7). However, when the assembly is done, the cleaning of small objects is quick. Usual electrolytic cleanings are carried out by immersion in the electrolytic baths; for this reason, ~80% of the survey responses indicated that the method is applied to small objects (Fig. 7). Nevertheless, the development of the electrolytic pencil allows cleaning small areas avoiding the immersion of the objects and allowing the cleaning of bigger objects without disassembling [35].

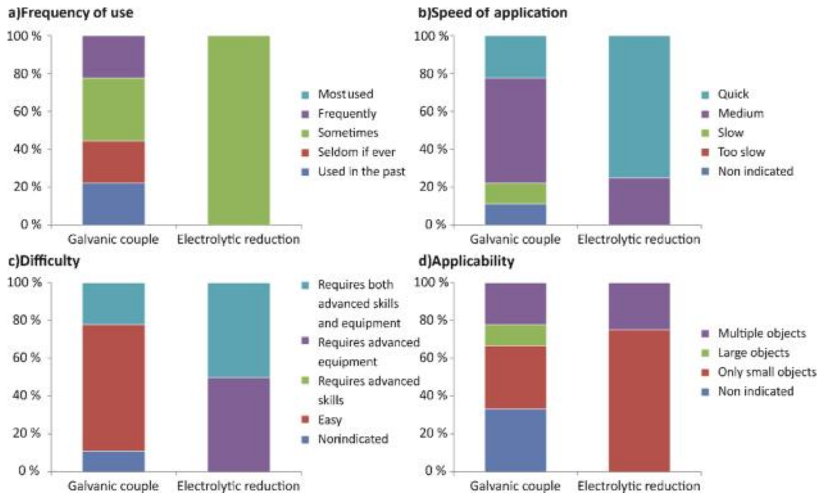

Fig. 7. Personal experience of professionals with electrochemical cleaning treatments.

Previous studies confirmed that electrochemical cleanings are effective on pure silver objects because they present a good efficiency removing sulfides and an almost negligible mass loss after successive cleanings [20, 36]. However, in sterling silver the re-deposition of copper corrosion products on the silver surface is favored by the lower reduction potential of the former metal ($E°_{Ag}$ = 0.7996V, $E°_{Cu}$ = 0.521V [21]), leaving a copper-reddish surface and making the method unacceptable unless it was combined with other procedures such as sodium persulfate [20, 22].

Conclusions

The cleaning procedures applied by professionals depend on the type of object and the skills and equipment of the conservator. Response to the survey has shown that mechanical methods are preferred by conservators, being those based on commercial products (mainly Prelim) and calcium carbonate the most popular ones, although they present a medium speed of application.

The application of commercial products is comparatively high, both for mechanical and chemical cleanings, despite the unknown composition of these products. Regarding chemical cleaning, besides commercial solutions, the thiourea-acid solution has been the most applied one, mainly due to be a fast and easy procedure, and the shiny final aspect of the object. Nevertheless, scientific studies confirmed that this treatment can be too aggressive if it is not applied correctly or if it is applied several times successively, so its use has decayed at present times.

The electrochemical procedures are the less used ones because they normally require advanced skills and specific equipment. As a final remark, it is important to know the type of silver (pure or alloyed), the type of corrosion products and the possible effects on the silver object before to carry out a cleaning treatment because the result could be even more harmful than the original situation.

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Appendix A

Silver cleaning survey

1. Could you please tell us your country and position (conservator, researcher...)?

2. Please describe as detailed as possible up to five cleaning methods for silver you have experience with.

Cleaning method 1

Cleaning method 2

Cleaning method 3

Cleaning method 4

Cleaning method 5

E.g. Cleaning method 1: mechanical cleaning with Pre-lim© using cotton swabs. Rinsing off with ethanol-deionised water 50% v-v. Drying with tissue paper

3. If none has been included in previous question, have you ever used any electrochemical cleaning methods? If yes, please describe the method. If not, please, describe why.

4. Regarding the methods described in previous questions, please describe your experience

	Frequency of use	Cleaning efficiency	Surface appareance	Speed of application	Difficulty	Applicability
Cleaning method 1						
Cleaning method 2						
Cleaning method 3						
Cleaning method 4						
Cleaning method 5						
Electrochemical method 1						
Electrochemical method 2						

7. Any additional comments?

8. (Optional) Name /Affiliation/ Contact Details

Done

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