AN INNOVATIVE METHOD TO REMOVE PRESSURE-SENSITIVE TAPE FROM CONTEMPORARY FELT-TIP PEN AND BALLPOINT PEN DRAWINGS ON PAPER. THE CASE STUDIES OF FEDERICO FELLINI FROM RIMINI FILM LIBRARY

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The removal of aged Pressure-Sensitive Tape (PST) is one of the most common issues during the restoration of drawings, as aged PST on paper can damage or negatively affect the artwork.
The removal of these materials, especially when applied on contemporary inks such as felt-tip pen, has to be considered carefully as it can present challenges that might not be successfully resolved using established approaches and materials. Conservators are familiar with many tape-removal methods including: mechanical action, immersion, poultices; each method however presents some associated risks, which may result in undesirable changes of the artwork (e.g.media bleeding, tidelines).
The purpose of this study is to develop a safe method to remove PSTs from works of art with felt-tip pen and ballpoint pen technique.
The study commenced with the preparation of test samples, exposed to accelerated aging treatment, choosing PSTs and techniques similar to those of the drawings by Fellini examined as case studies in this research.
Organogels based on the crosslinking of poly(methyl methacrylate) and poly(ethyl methacrylate) loaded with diethyl carbonate have been tested as a new removal system, made of gel and a non-toxic solvent. Applied directly on the PST, they led to removal of the backing and the adhesive of the PST.
The positive results obtained on test samples have been applied on Fellini's works, representing the first case of removability of PST from felt-tip pen and ballpoint pen artworks.

Key-words: Pressure-sensitive tape (PST), Tape stain removal, Felt-tip pen, Ballpoint pen, Organogel

1. Introduction

Pressure-sensitive tapes (PSTs) consist of four component layers: the backing, the adhesive mass, the release coat and the primer coat.

The development of the pressure-sensitive tape industry started around 1845 in the medical field with the production of adhesives and sticking plasters [1]. Starting from 1920, PSTs were mainly used in the auto industry; in 1925 Richard Gurley Drew invented masking tape, a PST with a paper backing and a rubber based adhesive. Gradually the natural rubber adhesive component was replaced with synthetic based mixtures, and with a new transparent backing, cellophane.

Originated in the context of the economic crisis that hit America at the end of the 20s, PSTs soon became a highly successful product.

PSTs have been erroneously used to repair tears or for mounting drawings on paper supports. Applied on paper they can damage and negatively influence the perception of the artwork: over time in fact they deteriorate differently according to the kind of backing and adhesive.

The chemical and physical damages that PSTs produce over time on paper supports are various, and can increase considerably if the PSTs are applied on paper and contemporary techniques, such as felt-tip pen and ballpoint. The physical damages include deformations and undulations, which generally occur in the case of PSTs with a backing-film; for example polyethylene, whose morphology is different from paper, under even a minimal thermo-hygrometric variation tends to move in a different way, creating tensions which can originate undulations (Fig. 1).

Chemical damages are due to the oxidation of the adhesive which, partially penetrating within the paper support, causes a visible alteration, resulting in a dark stain as can be seen for example in one of the case studies, partly above the drawing medium (Fig. 2).

Fig.1

In the case of PSTs with an acrylic adhesive, the adhesive's solvent can interact with inks, such as those of ballpoint pens, causing alterations that can be under the form of chromatic variations and colour migration (bleeding) (Fig. 3).

Fig.2

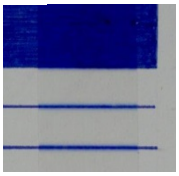

Fig.3

The most common removal methods of PSTs including both dry techniques (use of heat, silicone paper, spatulas and erasers) and wet cleaning, mainly using solvents [2].

Heat induced onto the PST's surface allows its detaching from the paper support, softens the adhesive and thus facilitates the removal with tweezers. This technique allows to remove the backing and part of the PST's adhesive. The simultaneous use

of heating tools and tweezers does not however allow a perfect control of the artwork being treated, making the intervention difficult.
Teflon spatulas and erasers, such as the crepe rubber eraser squares, based on natural latex, are generally used for the removal of residues of active (sticky) adhesive, but the friction can often cause tears or creases especially on low weight papers. Solvents that can be used for the removal of PSTs are: water, ketones (acetone), esters (ethyl acetate), alcohols, cyclic ether (tetrahydrofuran), and aliphatic hydrocarbons (cyclohexane). Many of these products can however solubilize some contemporary artistic techniques, such as felt-tip pen and ballpoint pen. The only solvent stable towards these techniques is cyclohexane.
In this study a new removal method will be presented - "Organogels in DEC" - to remove PSTs from works of art with felt-tip pen and ballpoint pen technique.
The organogels in DEC were applied on both model and on Fellini's drawings.

2. New methods for the removal of PSTs: Organogels in Diethyl carbonate

Organogels are a chemical kind of gel, capable of loading solvents or mixtures of solvents in their polymeric network and releasing them gradually onto the surface of the artwork. Their confinement avoids an uncontrolled release of the solvent, which can cause problems such as the migration of the artistic technique; moreover, confining the solvent within the gel guarantees the decrease of the solvent's evaporation and therefore also reduces harmful risks for the operators' health.
This new method for PST removal from contemporary drawings on paper with felt-tip pens and ball-point pen has been developed and experimented, in the framework of the NANORESTART project (H2020-NMP-21-2014/646063), at the Research Centre CSGI, Florence University Chemistry Department, in close collaboration with Antonio Mirabile. It is based on the use of organogels loaded with diethyl carbonate (DEC), considered to be a "green" solvent, never used before in the field of restoration.
The synthetized organogels can be considered more or less retentive according to their graduality of releasing the solvent; the more they are retentive, more the solvent is released gradually and in less quantity.
The organogel used for the removal of PSTs are based on poly(methylmetacrylate) (PMMA) [3] and poly(ethylmethacrylate) (PEMA) [4].
Three gels, containing DEC and at different retention capacity, were synthetized: 1) PMMA E2: ESC=85%, 2) PMMA E2.5: ESC=84%, 3) PMMA E3: ESC=75%. Amongst these the third formulation (PMMA E3) is the most retentive. Two organogels made of PEMA were also tested: ED50 with ESC=82% and EEA50 (containing ethyl acetate) with ESC=73%. ED50 is more retentive than EEA50.
The Equilibrium Solvent Content (ESC) indicates the mass of solvent in the gel at equilibrium (after a prolonged immersion in the solvent) in relation to the total mass of the gel.

Organogels present good transparency optical properties and good mechanical properties due to the formation of a three-dimensional network between the polymeric chains. These properties allow an easy removal without leaving residues on the artwork's surface. The absence of residues has been previously investigated by means of IR Spectroscopy (ATR-FTIR).

DEC: A new green solvent

Diethyl carbonate (DEC) is a green solvent; its constitutional formula is $O=C(OCH_2CH_3)_2$ [5]. The idea of ''green'' solvents expresses the goal to minimize the environmental impact resulting from the use of solvents in chemical production. [6]. DEC is part of the family of alkyl carbonates, and is a polar aprotic solvent, it is also called ethyl carbonate or carbonic acid diethyl ester [7]. It can soften natural and synthetic polymeric substances and be considered a valid green alternative to esters and ketones, which have been used for the removal of PSTs [8]. Amongst green solvents, DEC is the one that has shown the best results in terms of solubility on PST adhesives and stability towards all the artistic media used in the samples prepared for this study, reason why the organogels used for this experimentation were loaded mainly with this solvent.

2.1. Removal tests with Organogels in DEC on the samples

Choice and realization of the samples

The materials used in the realization of the samples were chosen in order to create samples similar in their paper support, artistic technique and PSTs to those of several drawings by Federico Fellini realized with felt-tip pens and ballpoint pen from the Film Library of Rimini, the case studies of an OPD diploma dissertation in which organogels were applied and experimented [9].

Thanks to the collaboration of Roberto Mannoni, a close collaborator of Federico Fellini, it was ascertained that Fellini used the following brands of felt-tip pens: Tombow AB and Swiss made Caran D'Ache.

Comparing Tombow felt-tip pens used by Fellini with those currently on the market, the first ones are called "Tombow AB", the latter "Tombow ABT Acid free". This leads to hypothesize a different formulation, which needs to be further understood, analysing and comparing the components of the original drawing lines with those realized with the current Tombow felt-tip pens.

Another important aspect taken into account in our research on felt-tip pen medium is the solubility of their ink, in water or in organic solvents, based on which felt tip pens are divided into two classes: water soluble felt-tip pens and solvent soluble felt-tip pens.

For our study the following brands were chosen: Giotto Turbo color, Carioca Doodles Italy, Tombow ABT Acid free, Swiss made Caran D'Ache, Permanent

Stabilo, OHPen universal. These are all water soluble, with the exception of Permanent Stabilo OHPen, soluble in organic solvents.
For the realization of the samples with ballpoint medium (the original typology was not ascertained) three different ballpoint pen brands were selected, in use between the 70s and 80s of the 20th century: Bic148, Staedtler noris stick 434 M, both with oily inks, and Pilot G-2 07 with fluid so-called "gel" inks.
The choice of PSTs fell upon the two main kinds of adhesives and backings generally applied on artworks to repair tears or for mounting systems: rubber and acrylic for the adhesives, cellulose and polypropylene for the backings. The chosen PSTs are: masking tape (paper backing and natural rubber adhesive) and ordinary tape (polypropylene backing and acrylic adhesive).
The following samples on paper (similar to office paper sheets used by Federico Fellini, produced by Cartiere Miliani Fabriano) were therefore prepared, with PSTs both on the rear and the front:
- Sample 1: Fabriano paper, Permanent Stabilo, Giotto and Carioca, masking tape (MKT) and ordinary tape (OT)
-Sample2: Fabriano paper, Tombow ABT, MKT, OT
-Sample3: Fabriano paper, Swiss made Caran D'Ache, MKT, OT
-Sample4: Fabriano paper, Bic, Staedtler noris stick 434M, PilotG-2 07, MKT, OT.

Removal tests – Application method
The PST removal tests were realized with organogels in DEC, cut to the size of the PST to remove; the gel was applied onto the PST and covered with a melinex sheet and a glass plate in order to create a confined environment and avoid the evaporation of the solvent. The gels were applied (for a maximum of 30 minutes) and subsequently removed without leaving any residues (Fig. 4). The action of the gel allows a softening-swelling of the PST and the following mechanical removal with a scalpel.

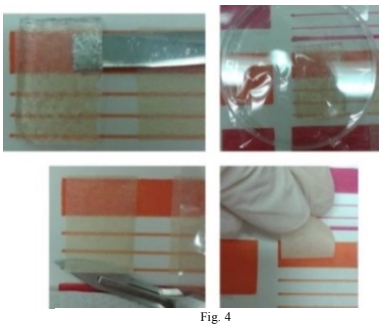
Fig. 4

2.2 Results and discussion
The removal tests on unaged samples revealed that the organogels in DEC work well in the removal of both kinds of PSTs used on the samples. On the treated samples, in the case of both masking tape and ordinary tape, it is possible to observe that there are no residues of adhesive and the paper supports and the artistic media are stable.
The organogels that gave the best results for the removal of masking tape were PMMA E3 and PEMA ED50 in DEC, and on ordinary tape PMMAE2 in DEC.

On the artificially aged samples the adoption of these new systems gave good results for the removal of ordinary tape. The best result was obtained with PMMA E3 EA 5% v/v in DEC, with the removal of the backing and of most of the adhesive.
The macroscopic observation of the aged masking tapes reveals that the backing appears rigid and tends to detach with simple mechanical action. A layer of deteriorated adhesive remains on the surface, covering the graphical strokes and it has partially penetrated into the paper matrix.
In the tests carried out, the main problem was the spreading of the deteriorated adhesive, which produced a visible stain over the artistic media. The best result in the adhesive removal was obtained with the most retentive gel, PMMA DEC E3%.

3. Case studies: the drawings by Federico Fellini of the Film Library of Rimini
The drawings by Federico Fellini subject of this study were realized on various kinds of paper, with felt-tip pens and ballpoint pen as artistic media.
Coming from different graphical collections, the drawings were acquired by the Municipal Film Library of Rimini between 1995 and 1997. A few of these drawings have been restored by the Department of Conservation and Restoration of Paper and Membrane materials of the Opificio delle Pietre Dure (OPD), Florence.
The works are: *Self portrait with the Oscar. "To Rinaldo, friend forever"*, 1993; *Anniversary: "14 May 1957 – 14 May 1987"*, 1987; *Titta and Federico in front of the remains of frescoes in the Church of Sant'Agostino, Rimini*, 1989. The drawings present damages due to the application of various kinds of PSTs, applied both on the back and front of the artworks, some above the artistic media and some only on the paper support, to fix some tears or to mount the drawings onto secondary supports.

3.1 Diagnostic Analyses on the PSTs
In order to analyse the typologies of PSTs present on the artworks, as well as photographs in visible light and observations under the stereo-microscope, useful for monitoring the conservation conditions and phases of the intervention on the PSTs, attenuated total reflection infrared spectroscopy in Fourier transform (ATR-FTIR) was used to identify the chemical nature of the PSTs (backing and adhesive).

Federico Fellini*, Self portrait with the Oscar. "To Rinaldo, friend forever"*, 1993, felt-tip pens on paper, Giuliano Geleng Collection (2nd)

The verso of the drawing is fixed with PSTs along its four corners to a secondary support in cardboard (fig. 5), applied after the realization of the artwork.

The macroscopic analysis of the front of the drawing reveals the presence, in correspondence of the four corners, of rectangular shaped dark brown stains (fig. 6), indicating an oxidation layer of the PSTs' adhesive, penetrated within the paper matrix. After the removal of the secondary support, it could be observed that on the rear of the drawing the PSTs' adhesive was still active (sticky) (fig. 7): this confirms the presence of double sided PSTs.


Fig. 5

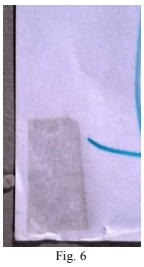
Fig. 6

From the analyses in IR Spectroscopy (ATR-FTIR) performed on a fragment of PST (Sample GR_13461/1(1)), it was ascertained that the adhesive is compatible with a synthetic rubber Styrene-Butadiene-Styrene (SBS) segment butadiene: 1449, 964, 2916, 2844 cm-1; segment styrene: 697, 1601 cm-1 (fig. 8).

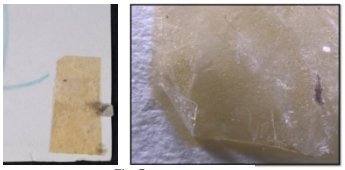
Fig. 7.

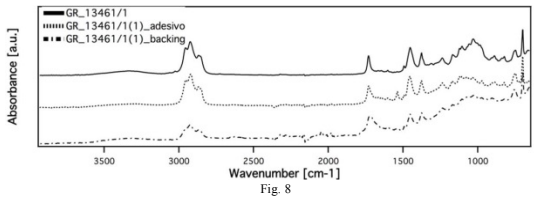

Fig. 8

Federico Fellini, *Anniversary: "14 May 1957 – 14 May 1987"*, 1987, felt-tip pens and ballpoint pen on paper, Anna Giovannini Collection

This artwork presents PSTs of the same typology, some also above the artistic media and some only on the paper support (Fig. 9). On the front three PSTs are visible: one on the upper right edge above the artistic medium, on the red number seven and on some of the light blue speech bubble strokes; another, in the central part of the same edge, covers red exclamation marks and is folded onto the back of the drawing. The third is on the left hand side of the lower edge, it covers pencil strokes and is folded onto the back; the PSTs were probably applied to repair tears visible on the artwork.

Fig. 9.

From the analyses in IR Spectroscopy performed on a fragment of PST (Sample GR_13461/6), it was ascertained that the adhesive is compatible with an acrylic adhesive (ca. 1730 cm-1, 1160 with a shoulder at 1240 cm-1) and the backing is compatible with a cellulose based compound (Fig. 10).

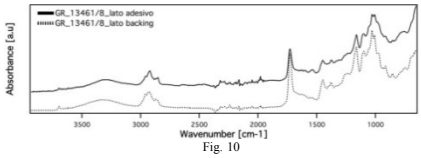

Fig. 10

Federico Fellini, *Titta and Federico in front of the remains of the frescoes of the Church of Sant'Agostino, Rimini*, ballpoint pen on paper, 1989, Benzi Collection

On the back of the drawing, above the paper support and the artistic medium are two PSTs, whose adhesive has penetrated within the paper matrix (Fig. 11).
The analyses in IR spectroscopy carried out on a fragment of PST (Sample GR_13461/9) reveal that the backing consists of polypropylene (2950, 2916, 2865, 2838 cm-1; 1450 and 1375 cm-1) whilst the adhesive is a synthetic rubber SBS (Fig. 12).

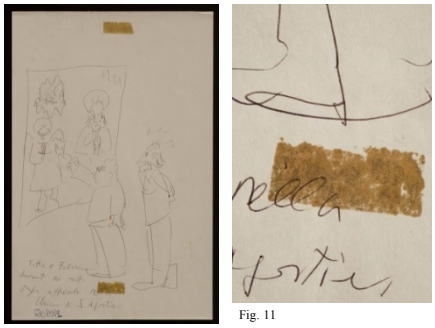
Fig. 11

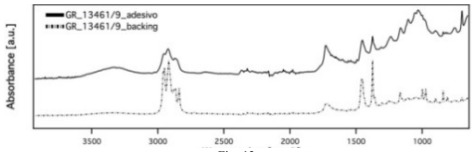

Fig. 12

3.2 Removal of the Pressure-sensitive tapes with organogels in DEC
After the positive results of the experimentation on the samples we proceeded to the PST removal from Federico Fellini's drawings.
Before this step, some preliminary operations were necessary: the detaching of the drawings from the secondary support, cleaning with dry techniques, stability tests on the paper support and solubility tests on the artistic media. The tests were realized to verify the interactions between the paper supports, the artistic media and the chosen removal methods.
The intervention of removal of the PSTs was articulated in two phases: 1) removal of the backing and of the adhesive; 2) removal of the adhesive penetrated within the paper support. During the first phase the backing and a part of the adhesive layer were removed with the organogels in DEC, the only non-toxic solvent revealed to be stable towards almost all of the artistic media analysed (Fig. 13).
For the tests on the artworks the most retentive organogels were chosen, as these had given the best results in terms of removal, stability towards the artistic media and the paper supports: PMMA E3 with ESC 75% and ED50 with ESC 82%.
These systems were used both on PSTs applied in correspondence of the artistic media and in areas where no artistic media were present. The gels were applied for

a maximum of ten minutes, as established during experimental tests on the samples. The mechanical removal of the adhesive residues was done by dabbing with the solvent DEC on a cotton swab and with the aid of a Teflon spatula and tweezers.
In the second phase we removed the adhesive penetrated within the paper matrix with a low pressure table and using the pure solvents DEC and EA (Ethyl Acetate). This operation was necessary for the PSTs with a synthetic rubber SBS based adhesive, differently from acrylic adhesives which have proved to be more viscous and with less tendency to penetrate into the support.

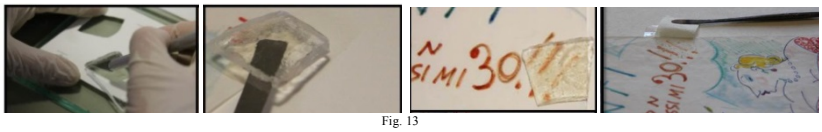

Fig. 13

3.3 Results and discussion

Federico Fellini*, Self portrait with the Oscar. "To Rinaldo, friend forever"*, 1993, felt-tip pens on paper, Giuliano Geleng Collection (2nd)

The first phase of PST removal was realized by using an organogel in DEC 100% v/v which allowed to remove the backing and a partial removal of the adhesive, slightly lightening the stain.
The second phase showed a complete asportation of the adhesive penetrated within the paper support, with a significant reduction of the stain (Fig. 14).

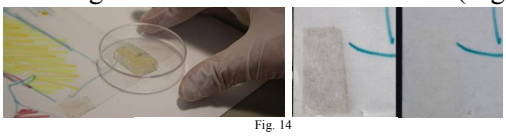

Fig. 14

Federico Fellini, *Titta and Federico in front of the remains of the frescoes of the Church of Sant'Agostino,* Rimini, black ballpoint pen on paper, 1989

This intervention was particularly delicate due to the presence of PST above the artistic media; in the first step the organogel PMMA E3 in DEC 100% v/v was used (Fig. 15), allowing to remove the backing and the surface layer of the adhesive with a slight reduction of the stain, attenuated more in a second step using a suction table and the pure solvents DEC and EA (Fig. 16).

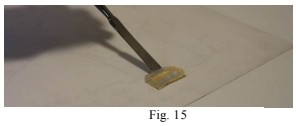

Fig. 15

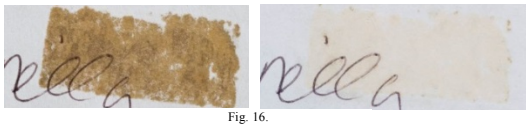

Fig. 16.

Federico Fellini, *Anniversary: "14 May 1957 – 14 May 1987"*, felt-tip pens and ballpoint pen on paper, 1987, Anna Giovannini Collection

In this case study the removal of PSTs was even more delicate due to two PSTs on the front of the drawing above the felt-tip pen medium; in this case also we used the most retentive organogel, allowing the removal of the backing and the adhesive (Fig. 17).

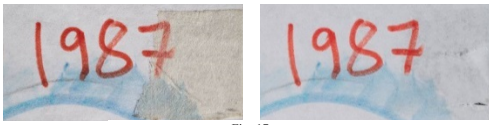

Fig. 17

Conclusions

This study presents a new approach for PST removal on contemporary artworks, in this case felt-tip pen and ballpoint pen on paper, with technical methods less invasive than those used so far, as well as effective.

The combined use of organogels and the non toxic solvent DEC proved to be a valid alterative to the traditional PST removal methods.

The results obtained on Federico Fellini's drawings have demonstrated this method's efficacy: the application of the organogels allowed the complete removal of the backing and of part of the PSTs' adhesive layer. In the case of adhesive penetrated within the paper support, the organogels in DEC contributed to the partial removal of the adhesive and the attenuation of the stains, further reduced with the use of the suction table and the solvents DEC and EA. This method proved effective also on stains above the artistic media, which resulted stable.

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