ATR-FTIR Spectroscopic Studies of Polymer-Based Identification Cards

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ATR-FTIR spectroscopic studies of polymer-based identification cards

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Abstract
Counterfeit production of polymer identity cards poses a significant economic cost to society and a threat to national security. Identifying these counterfeits is a challenge for 'frontline' personnel who lack training in specialised document examination. This study investigates the use of attenuated total reflectance Fourier Transform infrared (ATR-FTIR) spectroscopy with chemometrics as a potential approach to assessing polymer card authenticity. *In situ* analysis of several cards found that differentiation could be achieved based on the core polymer composition. A chemometric model was thus built for three driver's licence series produced in Western Australia and tested using a separate set of seven licences. The majority of test samples were correctly matched to the series of issue, with atypical samples recognisable based on their discriminant values. Synchrotron FTIR imaging revealed that differentiation between each series was possibly related to the adhesive used between the core layers. The approach presented in this work has the potential to be developed as a rapid screening method to identify suspect polymer cards warranting further examination.

1. Introduction

The possession and use of polymer-based (plastic) cards has become routine in modern society. Individuals often own multiple polymer cards for various purposes including loyalty, credit or debit, insurance, health care, security access or driver's licence cards. Many of these cards can be used as a means of proving identity, gaining access to financial accounts, or obtaining high-value security documents such as passports and visas. These cards are thus a frequent target for counterfeit production, posing significant economic and security threats on an international scale [1, 2].

Plastic cards generally consist of a polymer core with printed information on one or both sides, covered with a laminate film. The core can be a single or multi-layer system, typically consisting of polyvinyl chloride (PVC), polyethylene terephthalate (PET), polycarbonate, or a polymer blend [3-5]. European cards frequently use polycarbonate as a core material due to its high durability, whilst PVC is thought to be the main polymer used in Australia due to its lower cost and ease of mass production [6]. The laminate can similarly be applied as a single or multi-layer system, and may consist of polyester, polycarbonate, cellulose ester, or polysulfone [7-9]. The various layers are fused through the application of heat, or using adhesives such as polyester amide, polyester urethane, or polyolefin [7]. Depending on the intended purpose of the card, additional features such as holographic images or radio frequency identification tags may also be present [4, 8].

Forensic examination of polymer cards largely relies on visual inspection of the printing and security features under various light sources. These examinations often make use of a stereomicroscope or specialised viewing systems, such as a video spectral comparator (VSC). Although visual examination is a widely practiced and valued method of document examination, it is heavily reliant on the training and expertise of the examiner [10]. This is challenging for 'frontline' personnel such as immigration officials or licensing agents, who do not necessarily have extensive training in questioned document examination. This may result in counterfeit cards remaining undetected at critical checkpoints. A simple, non-destructive and reliable screening tool would thus be of high value to combating this form of crime.

Vibrational spectroscopic techniques are well suited to this purpose as they provide high chemical specificity, require minimal sample preparation and are non-destructive when applied *in situ*. Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy is particularly appropriate for this application as it is simple, robust and readily portable. Multiple studies have demonstrated the use of ATR-FTIR spectroscopy to characterise polymer-based products such as automotive paint [11, 12], food packaging films [13-15], and textile fibres [16, 17]. However, to the best of our knowledge, the use of ATR-FTIR spectroscopy for polymer card examination has not previously been reported in the open literature.

This work used ATR-FTIR spectroscopy to identify compositional differences amongst a variety of polymer cards, and to determine whether these could act as an indicator of authenticity for driver's licence cards issued in Western Australia. The potential structural differences between cards were further investigated using synchrotron infrared microscopy.

2. Materials and Methods

2.1. Sample collection and preparation

All polymer cards for this study were donated by individuals. Nine polymer cards issued by different organisations were used for a preliminary investigation into compositional diversity (Table 1). Subsequent investigation focussed specifically on Western Australian driver's licence cards. Three series of cards were used; those that began distribution in 2001, 2011, and 2014 (Figure S1). 15 cards were used as a calibration set for the construction of chemometric models (Table 2). An additional set of six Western Australian driver's licence cards issued from 1999 – 2017, as well as a New South Wales driver's licence expiring in 2017, were used as an external validation set (Table 2). Each card was thoroughly wiped with ethanol and a lint-free tissue in order to remove any contaminants prior to spectral collection.

Table 1: Polymer cards used for preliminary investigations into compositional diversity.

Card	Issuer
ANZ Access Visa Debit	ANZ Bank, Australia
RAC Rewards	Royal Automotive Club, Australia
HBF Healthcare	HBF Health Insurance, Australia
Working with Children	Department of Communities, Western Australia
Western Australian Driver's Licence	Department of Transport, Western Australia
Club Sobeys	Sobeys Food Retailer, Canada
Saskatchewan Health Card	Government of Saskatchewan, Canada
Curtin Student	Curtin University, Western Australia
Medicare	Department of Human Services, Australia

Table 2: Driver's licence cards used for series investigations. (*) denotes cards used as an external validation set. N/A = Not applicable.

Card Identifier	Series	Card Identifier	Series
AH17	2014	WVB20	2014
JM10	2001	JOM10	2001
PC20	2014	TT12	2001
RB20	2014	MB14	2011
TS12	2001	AR01*	N/A (Pre-2001)
CM19	2011	AR06*	2001
JM19	2011	AR11*	2001
KVDP18	2011	AR12*	2001
SL16	2001	AR17*	2011
SL21	2014	AR22*	2014
TL16	2001	AY17*	N/A (NSW issued)

An expired 2001-series Western Australian driver's licence was used for synchrotron-source spectroscopic imaging. Microtomed cross-sections approximately 7 μm thick were taken for cross-sectional imaging.

2.2. Attenuated total reflectance infrared spectroscopy

Spectra were obtained using a Nicolet iS50 FTIR spectrometer equipped with an iS50-ATR attachment (single-bounce diamond crystal). Measurements were taken over the 4000 – 400 cm^{-1} range in absorbance mode, with a resolution of 4 cm^{-1} and the co-addition of 32 scans. Data collection was performed using OMNIC software (version 9.1.24), with an air background spectrum obtained prior to each sample spectrum. As many polymer cards contain multiple layers, 'bulk' spectra were obtained from the card edge to provide representative cross-section sampling of the laminar structure (Figure S2).

2.3. Synchrotron Fourier transform infrared imaging

Cross-sectional mapping was completed at the infrared microspectroscopy (IRM) beamline at the Australian Synchrotron, Melbourne. The beamline consists of a Bruker Vertex 80v FTIR spectrometer and Hyperion 2000 IR microscope (Bruker Optik GmbH, Ettlingen, Germany) with a 20× objective and liquid nitrogen cooled mercury cadmium telluride (MCT) detector. High-resolution maps were collected using a germanium crystal (n_{Ge} – 4.0) ATR attachment, using a 0.5 mm pinhole and step size of 3 μm. An air background spectrum was first collected using 12 co-added scans, with sample spectra subsequently recorded with 32 co-added scans. All data collection was performed using OPUS software version 7.50, under 19 % relative humidity.

2.4 Data analysis

Chemometric data pre-processing and analysis was performed using the Unscrambler® X 10.5 software (Camo Software AS, Oslo, Norway). The 1880 – 2340 cm^{-1} region was omitted due to high absorbance from the diamond ATR crystal in this region. A baseline correction and range normalisation were then applied to account for spectral variations related to card thickness. Spectra were mean-centred and subjected to principal component analysis (PCA) using the non-linear iterative partial least squares algorithm. Samples were plotted using up to the first four principal components (PCs) to visualise the distribution of the samples and identify any outliers.

An LDA model was constructed for the discrimination of Western Australian driver's licences from the calibration set (12 cards), treating each series as a distinct class. This model was constructed using the linear distance measure and scores from the first four PCs (accounting for 93.0 % of the variance in the dataset). The model was then used to predict spectra from the validation set (seven cards), comparing the actual and predicted classifications to evaluate the efficacy of the model.

FTIR imaging spectra were baseline corrected and analysed using Bruker OPUS v7.0 and CytoSpec 2.00.03 software (Cytospec Inc., Boston, MA, USA). Images were further processed with ImageJ 1.50i software.

3. Results and discussion

3.1 Preliminary considerations

Spectra were initially collected from the front, back and across the edges of each card. It was decided that collection across the edge of the card was the most suitable for subsequent investigation, as it allowed a spectral cross-section of the full layer system to be obtained. However, this required the card to be manually held in contact with the ATR crystal, as no appropriate sample mount was available. Consequently, the reproducibility of the spectra must be assessed.

This was investigated by collecting spectra from a 2001 series Western Australian driver's licence card over multiple days (Figure S3). Differences in relative intensity were observed in the 3335 cm^{-1}, 1610 cm^{-1} and 400 cm^{-1} regions, which could be attributed to differences in the contact pressure and angle of contact. The evanescent wave only penetrates $1 - 2$ µm beyond the crystal surface, making handheld contact reproducibility difficult. An additional challenge is the malleable nature of the card, allowing it to bend or slip during spectral acquisition such that parts of the laminate or other layers were no longer in contact. This may result in higher intra-group spectral variation, resulting in less distinct separation between cards using chemometric analysis.

3.2 Population diversity

This investigation began with the collection of spectra from a variety of polymer cards (Figure 1). Peak assignments for these spectra are presented in Table 1. All cards exhibited a shoulder at ca. 690 cm^{-1} corresponding to a C-Cl stretch, indicative of polyvinyl chloride (PVC). This is consistent with the proposition that PVC is the core polymer used in the manufacture of Australian identity cards. The Western Australian driver's licence, Working with Children, Medicare, Saskatchewan Health Services, Curtin Student and Club Sobeys cards also show characteristic peaks at 1450 cm^{-1} and 1250 cm^{-1}. These were attributed to in-phase CH_2 and CH bending, again associated with PVC. In the RAC, HBF Health Care and ANZ Access cards, the CH_2 bending peak is shifted to ca. 1430 cm^{-1}, denoting the presence of polyethylene terephthalate (PET). These cards also give rise to a C-C-O stretching band at 1240 cm^{-1} associated with PET, and a grouping of peaks in the $1100 - 960$ cm^{-1} region associated with long-chain stretches in both PVC and PET. Based on this visual inspection, two groupings are evident based on the use of different polymers for their core. The RAC, HBF and ANZ cards appeared to consist of a PVC and PET blend, while the remaining cards were thought to be solely PVC-based.

Table 1: ATR-FTIR peak assignments for nine different polymer cards [18-20].

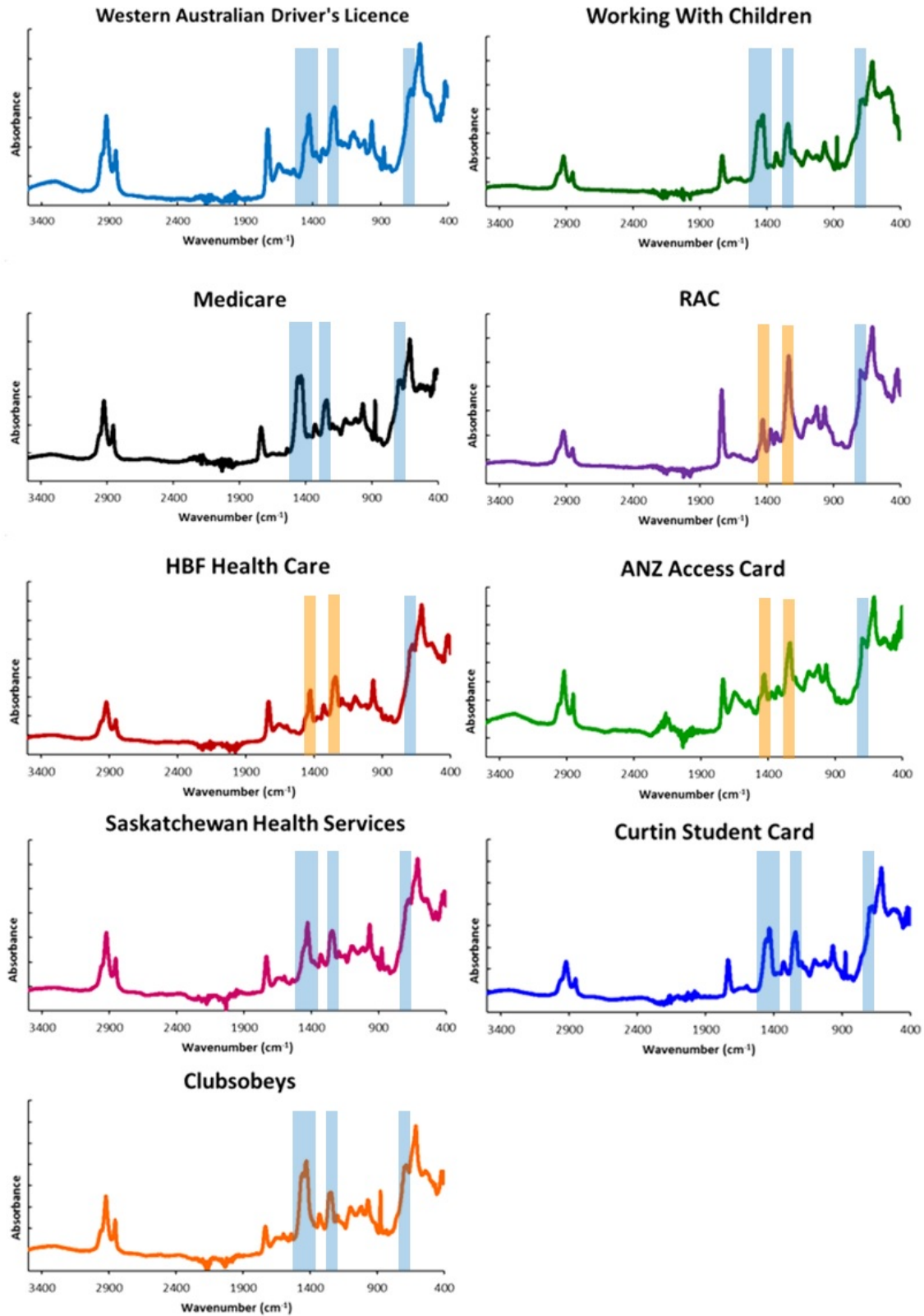| Wavenumber (cm^{-1})	Vibration	Polymer
2930-2920 cm^{-1}	Symmetric C-H stretch	PVC and PET
2900-2850 cm^{-1}	Asymmetric C-H stretch	PVC and PET
1450	In-phase CH_2 bend	PVC
1430	CH_2 bend	PET
1250	In-phase CH bend	PVC
1240	C-C-O	PET
1100-960	Chain stretch	PVC and PET
690	C-Cl	PVC

Figure 1: ATR-FTIR spectra of nine polymer card types. Blue highlighted regions are indicative of polyvinyl chloride and yellow regions are characteristic of polyethylene terephthalate.

The observed spectral differences indicated the potential to separate these groups using chemometric methods. The spectra from each card were therefore subjected to principal component analysis (PCA) to reduce the data dimensionality. This is achieved by transforming the initial set of correlated variables (wavenumbers) into orthogonal variables known as principal components (PCs) [21, 22]. The samples can then be plotted using the first few PCs in order to visualise the sample distribution and identify latent groupings. The scores plot generated from this data is presented in Figure 2, which shows that separation of the groups occurs along PCs 1 and 2. Interestingly, the Western Australian driver's licence clustered with the ANZ, HBC and RAC cards along PC1, but with the remaining cards along PC2, reflecting compositional similarities to both groups. Although the driver's licence is thought to have a PVC core, it is possible that the laminate layers share a similar composition to the cards thought to consist of a PVC-PET blend.

The factor loadings for the first two PCs (Figure 3) were examined to identify spectral regions contributing to sample separation. Both components produced strong correlations against peaks in the alkyl C-H region. PC1 was positively correlated with 2930 cm^{-1} and 2900 cm^{-1} bands, while PC2 was negatively associated with 2920 cm^{-1} and 2850 cm^{-1} bands. This likely reflects a downshift in C-H stretching frequencies in PET compared to PVC. As the lower stretching frequencies associated with PET are negatively correlated against PC2, it is expected that cards containing PET would obtain more negative scores against this component, which is reflected in the scores plot distribution. PC2 also has a strong negative correlation at ca. 1020 cm^{-1}, reflecting possible variations in polymer chain length. The higher C-H stretching frequencies likely associated with PVC are positively correlated with PC1, causing samples with a greater proportion of PVC to attain more positive scores along this component. This also aligns with a positive correlation between PC1 and the 1250 cm^{-1} C-H bend

associated with PVC. However, it should be noted that as the specific composition of the cards is unknown, these assignments cannot be made with absolute certainty.
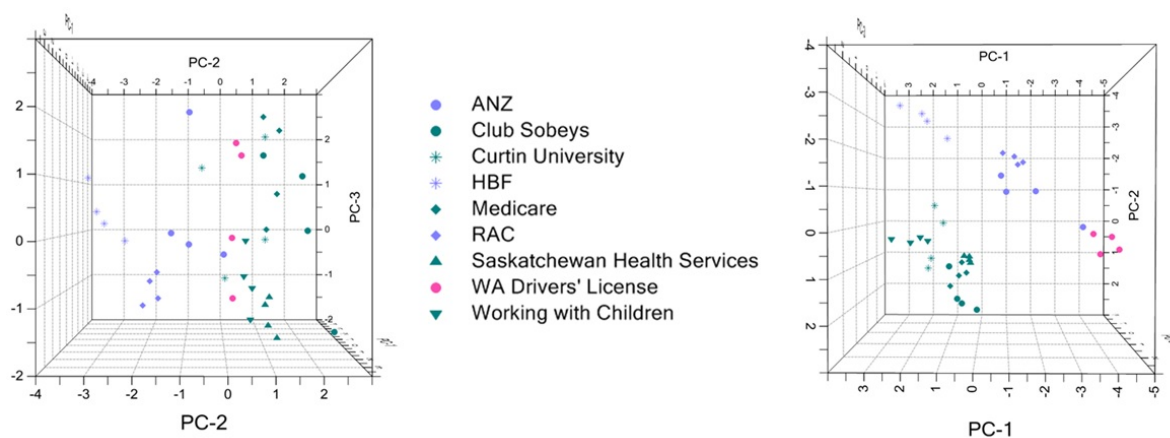

Figure 2: Three-dimensional PCA scores plots of nine polymer card types based on their ATR-FTIR spectra. Left and right images show different perspectives of the same plot.
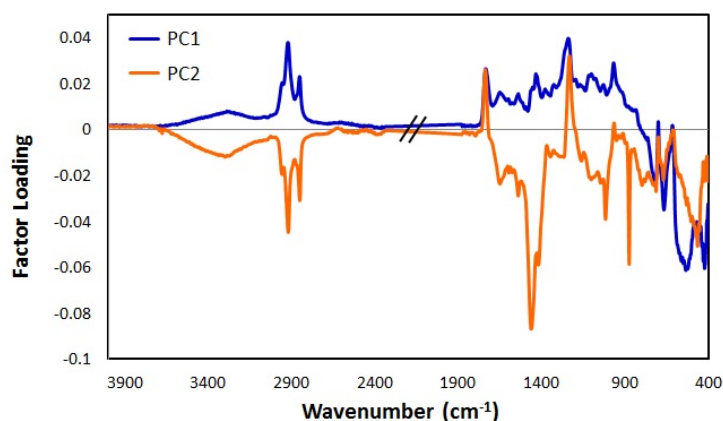

Figure 3: Factor loadings plot of PCs 1-2 for nine polymer card types based on their ATR-FTIR spectra.

3.3 Discrimination and classification of Australian driver's licences

Further work focussed on Western Australian driver's licence cards to determine whether separation between different versions of this card type could be achieved. Identity cards such as driver's licences are the most commonly detected form of fraudulent document; often used to facilitate organised crimes such as migrant smuggling or human trafficking [1, 2]. Licence cards can also be used as "breeder" documents to obtain legitimate forms of identification, such as passports [6].

There are currently two series of driver's licence valid within Western Australia, which began distribution in 2011 and 2014. An expired series distributed from 2001-2011 was also investigated. PCA was carried out on fifteen licence cards, with 89.4 % of total variance described by the first three PCs. The resulting scores plot (Figure 4a) showed a clear separation of the 2011 series along PC1, whilst the 2001 and 2014 series were overlapping. The 2014 series produced a high level of dispersion along PC3, suggesting that the composition of this series is potentially not as consistent

between individual licence cards. Separate PCA of the 2001 and 2014 series cards resulted in some level of separation along PC1 and PC3 (Figure 4b). However, as the *intra*-group and *inter*-group variation were of similar magnitudes, these series were not considered to be reliably distinguished.
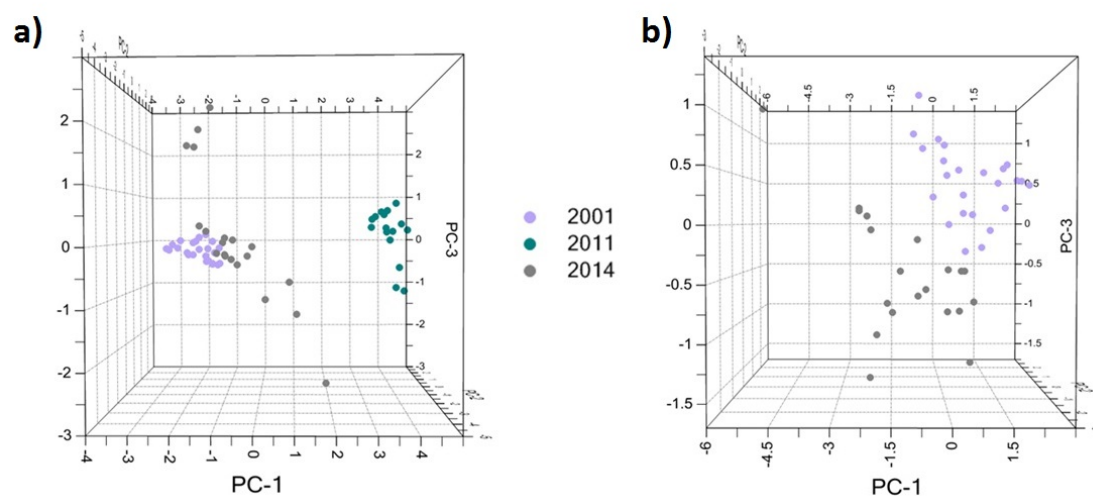

Figure 4: Three-dimensional PCA scores plots of (a) 2001, 2011 and 2014 series; and (b) 2001 and 2014 series Western Australian driver's licences based on their ATR-FTIR spectra.

Inspection of the PC1 factor loadings for analysis of all three series (Figure 5a) revealed strong positive correlations at ca. 2925 cm^{-1} and 1245 cm^{-1}, along with negative correlations at 540 cm^{-1} and 425 cm^{-1}. Comparing the normalised spectra for the centroid of each series (Figure 5b), the 2011 series has a greater relative absorbance at the 2925 cm^{-1} and 1245 cm^{-1} peaks, and much lower absorbance below 570 cm^{-1}, resulting in more positive scores along PC1. This may reflect a greater relative proportion of PVC compared to other constituents in the non-core layers. PC3 was positively associated with the chain stretch region at 1020 cm^{-1}, indicating that the dispersion amongst 2014 series cards along this component may be due to chain length variations. However, due to the layered nature and unspecified composition of these cards, it is again difficult to determine exact differences leading to separation. It should be emphasised that these spectral differences may not be readily identifiable to field personnel such as customs or licensing agents. The development of a chemometric model to flag suspect cards would thus be highly beneficial for frontline use.
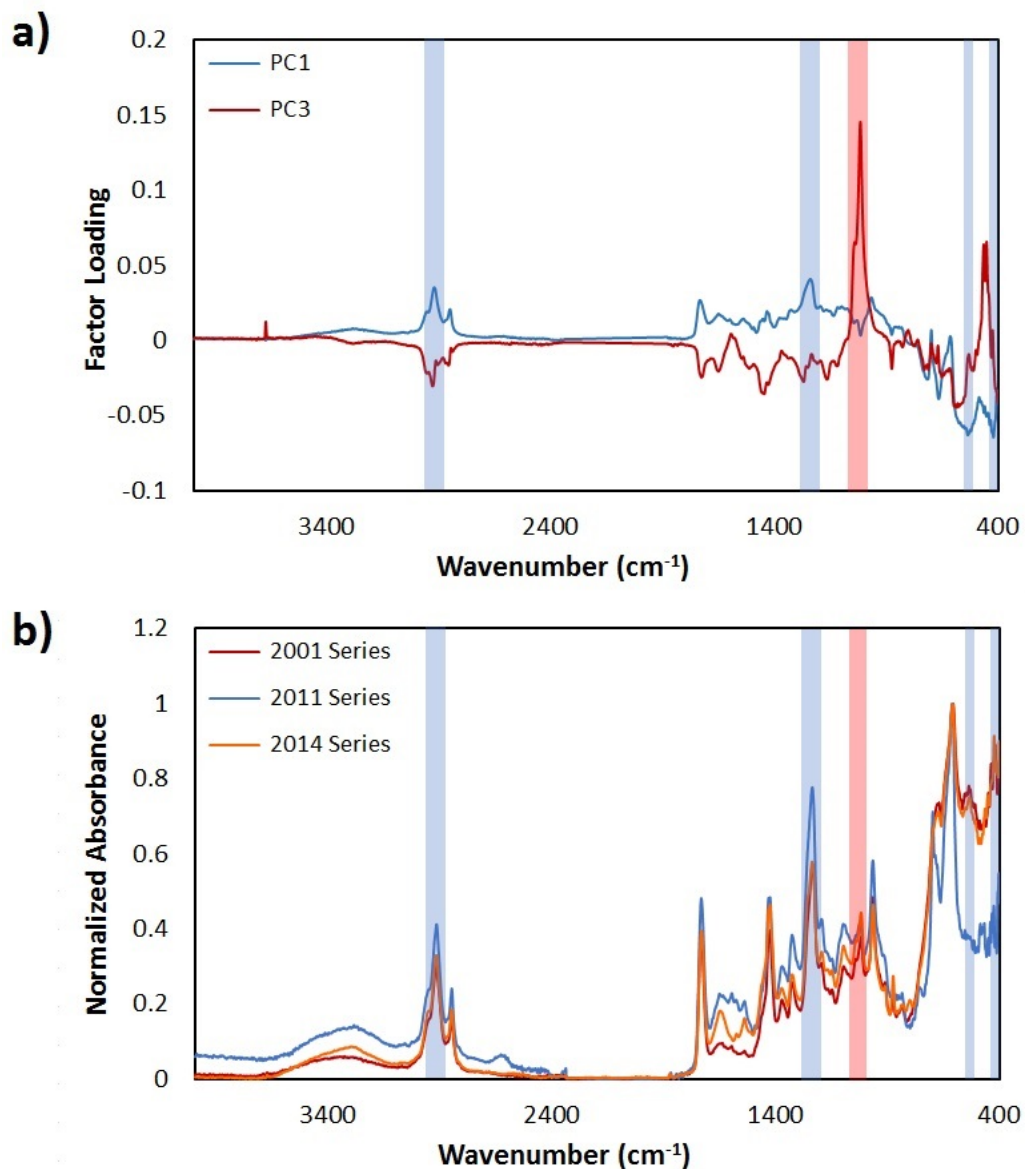
Figure 5: a) Factor loadings of PC1 and PC3 for three series of Western Australian driver's licences based on their ATR-FTIR spectra; and b) Normalized spectra from the centroid of each series.

A discriminant model was therefore constructed from the calibration set using the first four PCs, treating each series as a discrete class. The resulting model gave 98.3 % correct classifications for the calibration set (Table 2). As expected, the only incorrect classification was between the 2001 and 2014 series. Development of a separate discriminant model incorporating only these two series did not alter this misclassification. Nonetheless, the incorrect classification was limited to a single replicate, and thus the overall assignment of this card remained correct.

Table 2: Classifications of samples in the calibration set using a four-PC LDA model. The overall classification accuracy was 98.3 %.

Series	Correct	Incorrect
2001	24	0
2011	16	0
2014	19	1 (2001)

Using the same data to both construct and test the model can often result in an over-prediction of its efficacy. The model was hence further evaluated using a separate validation set of seven driver's licences not incorporated in the model. These included two 'atypical' cards that did not belong to any of the three series investigated; a pre-2001 series Western Australian licence and an interstate licence issued in New South Wales.

Of the five licences belonging to one of the series included in the model, four were correctly classified, with the single misclassification again occurring between the 2001 and 2014 series. The pre-2001 licence was predicted as belonging to the 2001 series, indicating that the composition did not significantly change when the 2001 series began distribution. The New South Wales licence was predicted as corresponding to a 2014 series Western Australian licence. As it is not known whether this licence in fact consists of the same polymer as the Western Australian 2014 series, it cannot be ascertained whether this is a 'correct' assignment.

Table 3: Classifications of samples in the validation set using a four-PC LDA model. Labels in brackets indicate assigned groups. N/A = Not Applicable.

Card Type	Actual Series	Predicted Series
WA Driver's License (1999)	N/A	2001
WA Driver's License (2001)	2001	2001
WA Driver's License (2006)	2001	2001
WA Driver's License (2007)	2001	2001
WA Driver's License (2012)	2011	2011
WA Driver's License (2017)	2014	2001
NSW Driver's License	N/A	2014

It should be noted that when using LDA, all samples will be assigned to a pre-existing class. This assignment is made according to the sample's discriminant values (DVs), which act as distance measures between the sample and each known group. An unknown sample is classified as belonging to the group yielding the smallest magnitude discriminant value, indicative of the 'closest fit'. DVs for a correctly classified Western Australian driver's licence, incorrectly classified licence and the New South Wales licence are displayed in Table 4.

Table 4: Discriminant values for a correctly classified driver's licence, incorrectly classified driver's licence and an atypical licence using a four-PC LDA model. Values are averaged across four replicate spectra.

Licence	2001 Series	2011 Series	2014 Series
WA Driver's License (2017)	-2.1	-66.3	-8.1
WA Driver's License (2012)	-83.4	-3.0	-63.9
NSW Driver's License	-25.7	-33.1	-10.9

The incorrectly classified Western Australian license obtained similar, low-magnitude values against both the 2001 series (-2.1) and 2014 series (-8.1). The similar values indicate that the licence was actually placed in the region between the 2001 and 2014 groupings and is not well classified. The

fact that both values are of low magnitude also indicates poor class separation, with a distance measure of only 10.2 between the centroids of each series. The New South Wales licence, although assigned to the 2014 series, similarly obtained similar magnitude values against all three series. Projection of these spectra onto the initial PCA model confirmed that this licence was placed between the three series and thus could not be confidently assigned (Figure S4).

Comparatively, the correctly classified licence obtained a small magnitude value against the correct 2011 series (-3.0) and much larger values against the remaining series, indicating that this licence is both well classified and distinctly separated from the remaining groups. In this way, atypical samples can be readily identified from those that are confidently assigned. These results highlight the importance of examining the discriminant values to evaluate the quality of predictions made using chemometric models.

3.4 Synchrotron imaging

To gain a better understanding of what might be causing the separation between licence series, an investigation into the structural differences was undertaken. This work was undertaken at the Infrared Microscopy (IRM) Beamline at the Australian Synchrotron, Melbourne. Thin cross-sections approximately 7 μm thick were taken from a 2001 series Western Australian driver's licence for FTIR imaging (Figure S5). False-colour maps of the cross-sections were generated from selected integrated peaks, and then superimposed over an optical image of the sample to create a set of composite images (Figure 6).

Inspection of these images revealed a small feature in both cards which appears to be positioned between the primary layers. Spectra extracted from this feature exhibit peaks centred at ca. 2850 cm^{-1}, 1740 cm^{-1} and 1650 cm^{-1}. These peaks were assigned as C-H stretching, C=O stretching and N-H bending vibrations respectively. Based on the spectra, this feature is suspected to be a thin layer of adhesive such as polyester amide [23]. This indicates that discrimination between licences may be influenced by the use of a different adhesive between each licence series.
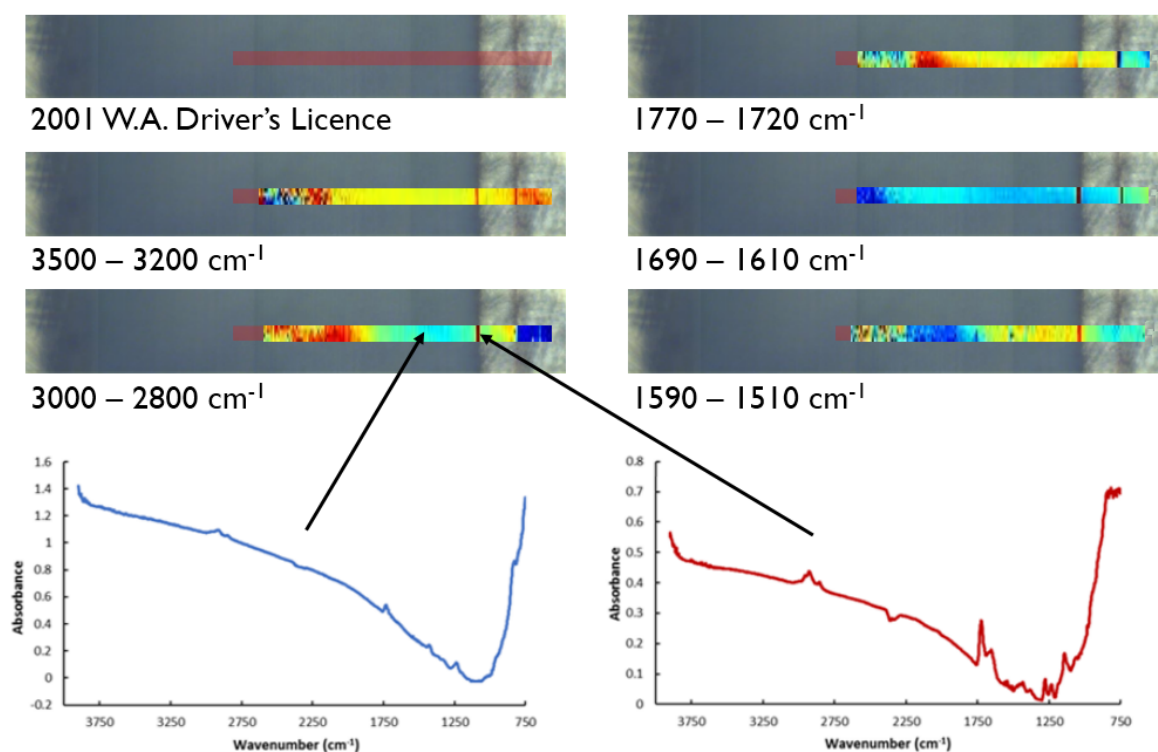
Figure 6: Composite FTIR false colour images from cross-sections of a 2001 series Western Australian driver's licence, based on integration of selected infrared peaks. Representative spectra are shown for the core polymer and suspected adhesive layer. Decreasing intensity from red to blue.

4. Conclusions

ATR-FTIR spectroscopy with chemometrics was successfully used to distinguish broad groupings of polymer cards based on their core composition. Similarly, a focussed investigation of Western Australian driver's licence cards resulted in clear discrimination between the two currently valid series (2011 and 2014). A subsequent predictive model was able to correctly classify four of five test licences, and atypical or poorly classified licences were readily identifiable from their discriminant values. The developed model may be used as a simple and high-throughput screening method for 'frontline' personnel to identify cards that have a composition or structure that is inconsistent with their purported type and origin. This would enable more rapid and accurate referral of potential counterfeits to a document examiner for further examination. This approach could also provide document examiners with a statistically validated basis on which to support their conclusions from visual examinations.

Synchrotron FTIR imaging identified a potential adhesive layer, suspected to be a polyester amide, which could potentially be contributing to separation between different cards. However, this requires confirmation through analysis of additional samples. It would also be of interest to conduct Raman imaging as a complementary technique to FTIR, allowing a more complete chemical characterisation to be obtained.

Transitioning this approach to operational use will first require further validation using a wider range of cards, including known counterfeits. Development of an appropriate sample mount for the collection of edge spectra would also improve the reproducibility of spectral measurements,

potentially allowing for more reliable separation between groups. Future work may also examine the effect of card storage conditions on their characterisation using spectroscopic techniques.

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Supplementary Information

All data associated with this work is available open-access via Research Data Australia;

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Figure S1: Sample images of Western Australia driver's licence series used in this study. Image adapted from Western Australia Department of Transport [1].
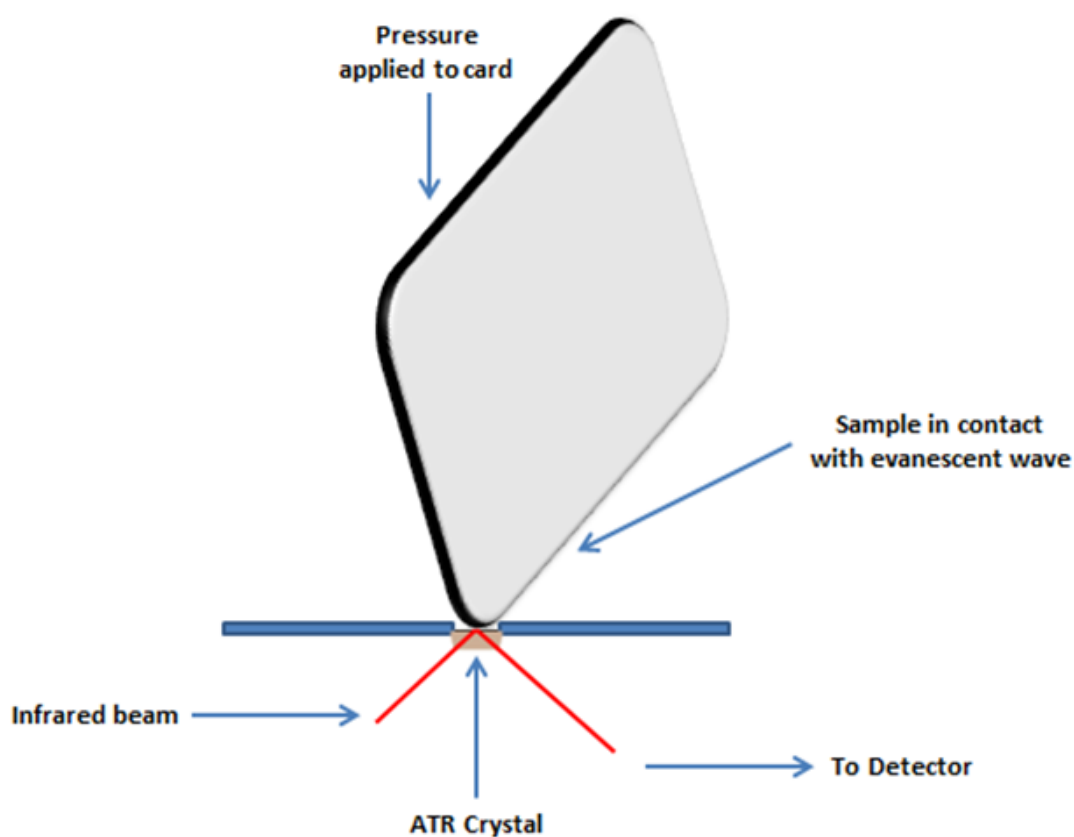
Figure S2: Measurement set-up for cross-sectional ATR-FTIR analysis of polymer cards.
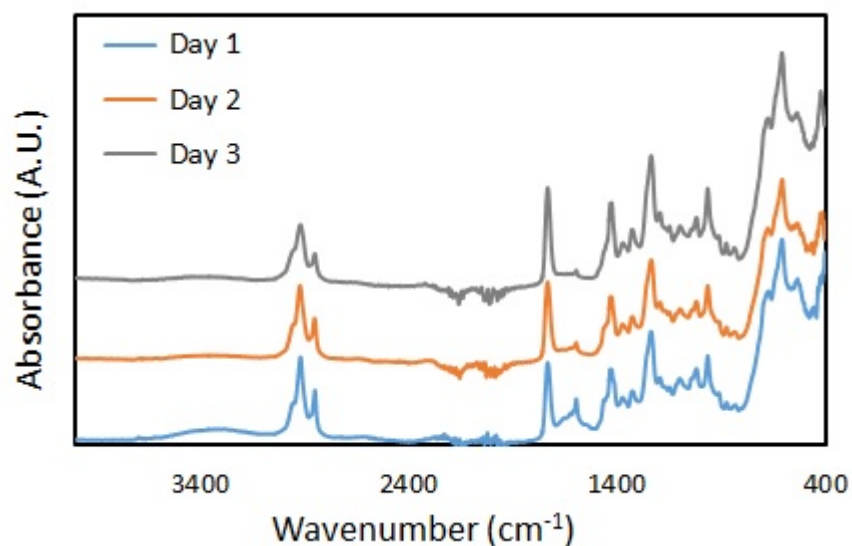

Figure S3: Replicate ATR-FTIR measurements acquired from a 2001 series Western Australian driver's licence across three different days. Spectra have been offset for clarity.
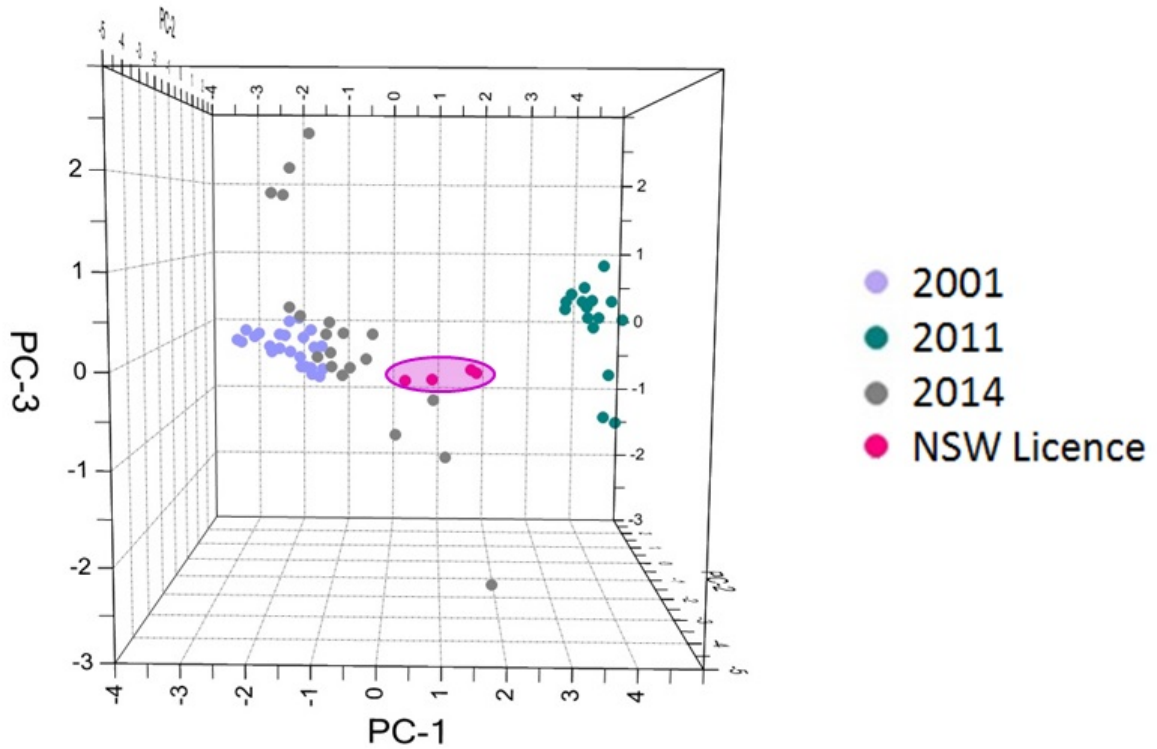
Figure S4: Three-dimensional PCA scores plot showing projection of spectra collected from a New South Wales driver's licence (circled) onto the PCA model constructed using Western Australian driver's licences.
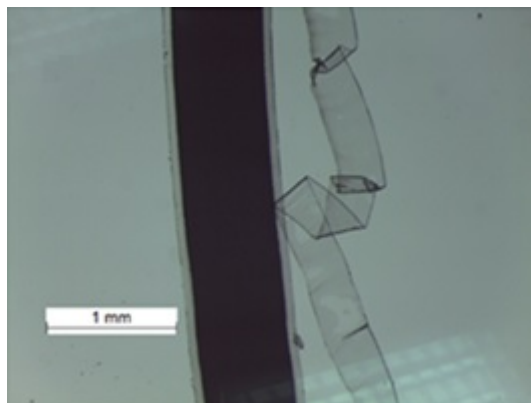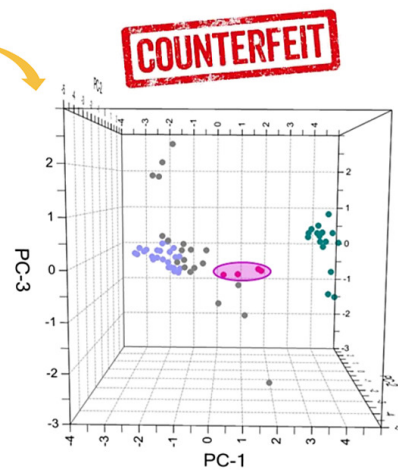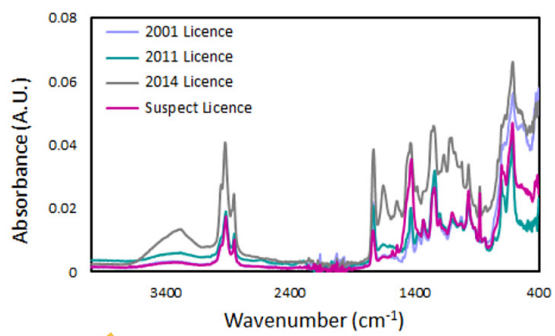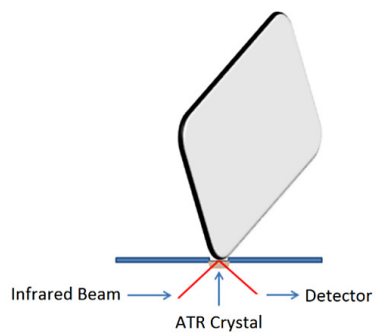

Figure S5: Microtomed cross-section of a 2001 series Western Australian driver's licence.

References

1.   Department of Transport. *Increased security of licence cards*. 2017; Available from: https://www.transport.wa.gov.au/licensing/drivers-licence-cards.asp.

IDCards_ATR_GraphicalAbstract.png (540.26 KiB)