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An infrared and SEM study of the margins of some German hyperinflation postage stamps.

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ABSTRACT

Infrared spectroscopy and scanning electron microscopy (SEM) studies were performed on the margins (the area without printed colors) of a set of German postage stamps dated 1923 and corresponding to the hyperinflationary period of economy.SEM results showed the presence of Al, Si, Zn and S in the samples, but in different combinations. Sulfur is considered to be a contaminant. The Al-Si combination is credited to kaolinite, Zn to ZnO and unaccompanied Al possibly to Al_2O_3. Several experimental IR bands of kaolinite agree with the suggestion of its existence in some stamps. In the case of Al and Zn alone we have supposed that, with the passing of time and with the help of moisture, AlOH and ZnOH were formed in a quantity allowing detecting some of the metal-OH bands by IR spectroscopy. On the basis of previous infrared experimental results on Al and Zn hydroxides we assigned some IR bands to the OH groups bonded to these metals. Our results indicate that it is highly probable that the paper used to print these postage stamps was originated in more than one paper mill.

Keywords: Cellulose, postage stamps, infrared spectra, archeology, archeophilately, SEM, kaolinite, white zinc, hyperinflation, Germany, scanning electron microscopy.

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INTRODUCTION

Following Wikipedia, "*stamp collecting is the collecting of postage stamps and related objects*". People practicing this activity are called "*stamp collectors*". The same Encyclopedia says that "*philately is the study of stamps and postal history and other related items. It also refers to the collection, appreciation and research activities on stamps and other philatelic products. Philately involves more than just stamp collecting*". After the first postage stamp was released in Britain in 1840 people began to collect almost immediately [1]. Russell Belk refers to collecting as "*the process of actively, selectively, and passionately acquiring and possessing things removed from ordinary use and perceived as part of a set of non-identical objects or experiences, is an acquisitive, possessive, and materialistic pursuit. It differs from most other types of consumption in the concern for a set of objects, the passion invested in obtaining and maintaining these objects, and the lack of ordinary uses to which these collected objects are put*" [2, 3]. Like most collecting activities, the beginning of the stamp collecting activity was at the commencement a somewhat costly activity because, as Apostolou comments, "*most of the times collectors have to purchase their objects of desire*" [4]. The first postage stamps catalogues were "pure" in the sense that they did not include prices. A good example is Oppen's *Postage Stamp Album and Catalogue of British and Foreign Postage Stamps [*5]. But, like in all collecting activities, there always appear people behaving so as to minimize their work and maximize their profit. This is the very origin of the actual postage stamps market with auctions, stamp buyers and sellers, etc. In 1863 an anonymous writer says that "*twelve months ago* [1862] *not a stamp could be purchased in London at any shop, whereas there are now a dozen or more regular dealers, and numbers of private individuals make a pretty tolerable profit out of the coloured rectangles launched into circulation, with no purpose of that kind, by Sir Rowland Hill*" [6]. And almost immediately, around 1860, the lucrative industry of forged stamps was born [7]. On their side, national postal systems rapidly recognized an opportunity for making extra revenue; and they began issuing postage stamps unequivocally directed to the collector's market. Today there is an undeniable strong marriage between money and stamp collecting [8]. Business is so good that even in Walmart supermarkets postage stamps are sold for collectors! (This is one example among many). Unhappily it seems that today "*pure*" catalogues do not exist: you find only catalogues *with* a price list. And in almost all the catalogues you find inside an endless list of stamps buyers, sellers, auctioneers and dealers. Not longtime ago, when science began to study stamps, some papers were focused in distinguishing between original stamps and forgeries [9-24]. Many scientific studies of postage stamps make an explicit reference to fully mercantilist concepts such as 'investment', 'price', etc. indicating the hidden influence of money. Anyway, and only for the purposes of framing the role of science in philately, we may state the following: a scientific examination of a certain stamp allows stating one and only one of these two statements*: the stamp is clearly a forgery* or *probably this stamp is authentic* (with the word *probably* included). Science cannot proclaim that a stamp is *doubtless authentic* because there is no a test for detecting the property of being authentic as a whole: at most the nature of the paper, ink(s), coating(s) and filler(s), and the printing technique can be declared as belonging to the time of the authentic stamp. What is usually done after is the fallacy of composition and generalizing from the parts to the whole. Even if we were able to employ dating techniques to attribute a date in the past to each element comprising a postage stamp, we cannot prove that they were assembled at that date. Form this point of view, all the stamps used by Imperio et al. for building their spectral database for Italian stamps were only declared as being probably authentic because no proof of genuineness was provided. After these thoughts about philosophy of science, we shall comment about a new aspect of philately.

It is probable that Sumerian clay tablets or Egyptian papyri lasted so long because they were the product of many trial/error procedures. More than 175 years ago the first stamp saw the light. They were not printed to last forever. In some cases, protections against erasure of cancellations and of the effect of humidity were added. The stamp paper(s) suffers from most of the problems of library books, inks disappear from certain stamps, some postmarks dispersed slowly through the paper, etc. But, given the appropriate environmental conditions, they can survive from natural or human-produced (wars, contamination) disasters. Let us provide some examples. The Katyn massacre was a series of mass executions of Polish nationals carried out by the Soviet secret police in April and May 1940 and approved by the Politburo of the Communist Party of the Soviet Union, including its leader, Joseph Stalin (22,000 killings, see Wikipedia). The first mass graves were discovered in the Katyn forest by the Germans in 1943 and in the original filming it is possible to appreciate that paper documentation was found despite the harsh winter conditions at that place (in the Internet Archives the reader may find more visual material). In the Republic of Iraq, the Syrian Arab Republic and the Islamic Republic of Afghanistan, years of war with the associated destruction of cities, towns and villages had produced an enormous quantity of debris possibly containing all kinds of philatelic (postage stamps, setenants, souvenir sheets, sheets of stamps, postal stationery, postal markings, envelopes, boxes, metal dies and plates, etc.) and post office materials. The same holds for the recent ethnic/religious Balkan wars. In an indeterminate future these places will be excavated and the findings will be classified, studied, restored and conserved. It is possible then that in the future a new specialty, called archeo-philately, will be needed for dealing with these materials at the beginning of an excavation (to recognize them as philatelic materials at a first sight for example). The term archeo-philately was employed first by Dasture et al. but it is not clear if with the same meaning used here [25]. Large collections of stamps (and related materials) exist in several parts of the world: National Philatelic Collection held at the British Library, the Smithsonian National Postal Museum, the Singapore Philatelic Museum, the Stamp Museum in Budapest, the National Philatelic Museum at New Delhi, the Philatelic Museum of Indian Stamps at Ahmedabad, etc., that could serve as reference material in some cases. But we think that in the majority of cases only fragments of stamps will be recovered. We do not remember reading a paper focusing on postage stamps from the point of view of '*present or future archeological objects'* or simply '*archeological objects'*. Even if the quality of the materials employed to fabricate them guarantee their continued existence only under very special circumstances, this is not a fact impeding this approach. Therefore, we should accumulate enough scientific data to know the *fingerprints* of each stamp. In this way, if in the future a fragment of paper, colored or not, is found and identified as being possibly a postage stamp, the comparison of the results of its analysis with the many stamps' fingerprints could help to identify it. Unhappily, some studies provoke the destruction of the sample(s). Even if a stamp is damaged to a large extent, for one of us (J.S. G.-J. is a dedicated stamp collector and studious) is really painful to dispose of them in this way.

In this paper we shall focus in the postage stamps produced during the hyperinflation period in the Weimar Republic (Germany). As there is plentiful of literature covering this period [26-29], we only remember the reader that, for example, for a domestic letter of less than 20 grams, a citizen must pay 20 Marks in January 15, 1923 and 50,000,000,000 Marks in December 12, 1923. Considering that the paper for stamp printing was supplied by a large number of paper manufacturers located throughout Germany, we become interested in answering the question if the composition of the paper of stamps with different denominations was exactly the same or not.

Samples and Methods

We selected thirteen stamps of a definitive series of hyperinflation stamps. Figure 1 shows the selected stamps and Table 1 shows their identification accordingly to the Michel Deutschland Spezial Katalog [30].

Figure 1: German postage stamps used in this study. From left to right and top to bottom: S1, S2, S3,---S13.

Table 1: Classification of the postage stamps studied.

Sample	Michel DS 2016
S1	313AP
S2	314AW
S3	315A
S4	316AP
S5	317AW
S6	318AP
S7	320AW
S8	321AW
S9	322AP
S10	323AP
S11	324AP
S12	326AW
S13	327AW

The samples corresponded to mint never hinged stamps. Because experiments were carried out at two different laboratories separated by about 2,000 km, three stamps of each denomination were needed. To guarantee that the paper samples were the same, stamps were taken from blocks of four unseparated stamps. The samples for infrared (IR) analysis were obtained by cutting the external border of the stamps after rising the stamps accordingly to our previous work [31]. The IR spectra were recorded with a Perkin Elmer Systems 2000 spectrophotometer. The standard technique of disk pressing in KBr was employed. Briefly, 0.5-1.0 milligrams of sample and 80-100 mg of IR grade KBr were used. The mixture was submitted to a pressure of 15 t /cm^2 for 30 seconds to form a 13 mm diameter disk. The SEM analysis was performed with a JCM-6000 NeoScope Benchtop equipment. An electron beam with variable energy of 5, 10 or 15 keV is used to scanning the sample under a vacuum of 10^{-2} Pa. The analyzed area can be varied in the range of about 22 mm^2 (magnification 22x) up to 12 µm^2 (magnification 30000x). The reflected backscattered electrons are collected with a detector yielding the morphology-picture (SEM). In the “mapping” mode the distribution of the elements within the sample is presented.

RESULTS AND DISCUSSION

Appendices A and B show, respectively, the IR spectra and the SEM results of the samples. The treatment of the samples to make the KBr pills does not allow us to know if some chemical compounds were used for filling, coating or both purposes. Also we expect that the treatment should expose almost all or all the chemicals that could be located on the cellulose fibers. The analysis of the SEM results (see Appendix B) allows us to separate the thirteen samples in five groups shown in Table 2.

Table 2: Ordering of SEM results by elements content.

Elements	Samples	Suggested chemical compounds
Al, Si, S	S1, S2, S3, S4	Kaolinite
Al, Si	S5, S10	Kaolinite
Al	S6, S7, S11, S12, S13	Al_2O_3 (?)
Al, S	S8	Al_2O_3 (?)
Zn, Al, Si	S9	ZnO, kaolinite

We can see that sulfur appears in five samples (S1-S4, S8). We think that the presence of this chemical is the result of anthropogenic activity. We do not know the history of each one of these stamps from the place of printing in Germany to our hands in Chile after about ninety five years. A possible source of sulfur contamination is the pulp mills but also the atmosphere because of the chemical properties of paper. Many atmospheric constituents (hydrogen sulfide, oxides of nitrogen and sulfur, etc.) are absorbed by the water previously absorbed by the paper and, when the paper dries out again, some of these chemicals remain [32]. The presence of zinc in sample S9 could correspond to “zinc white” (ZnO that was beginning to be produced around 1840).This molecule has strong IR bands at 450, 475 and 501 cm^{-1} [33].Figure 2 shows the IR spectrum of S9 in the range 400-600 cm^{-1} (see also Fig. A9 of Appendix A).

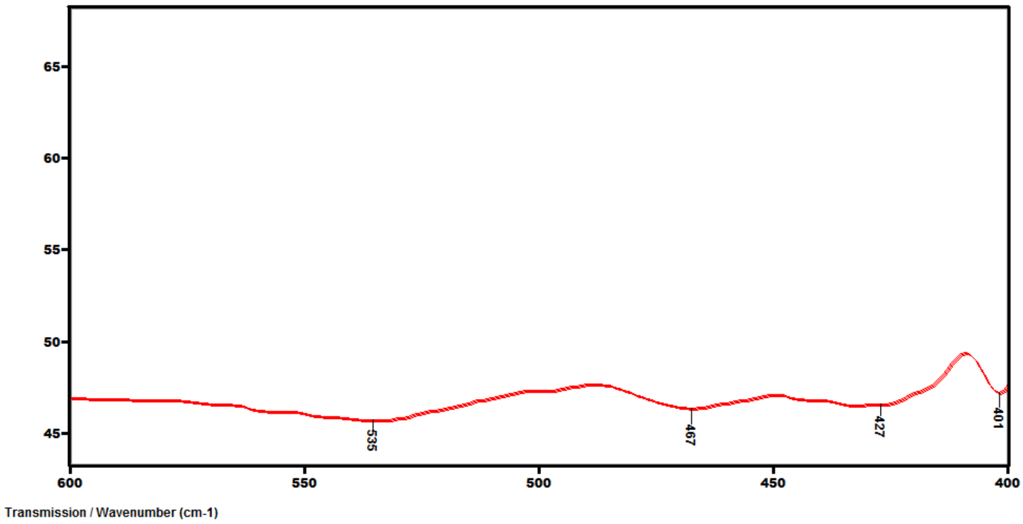

Figure 2: IR spectrum of S9 in the400-600 cm^{-1} range.

It seems that ZnO is not present in the sample. Brittain observed that ZnO in ink can undergo a chemical reaction with the soap used as surfactant in the oil component of the ink formulation [34]. This ZnO-soap-microcrystalline cellulose complex (ZSM) should produce bands at about 1396, 1456 and 1537 cm^{-1}[18, 34]. These bands are not present in the IR spectrum of S9 (see Fig. 3).

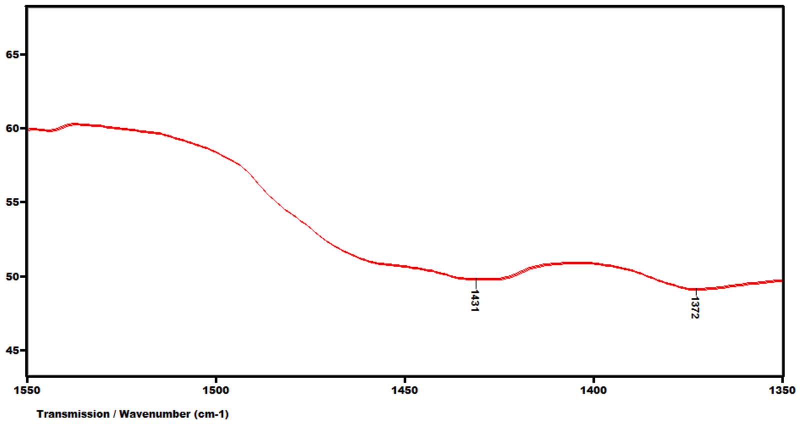

Figure 3: IR spectrum of S9 in the 1350-1550 cm^{-1} range.

In addition, the ZSM complex exhibit two strong bands at 2849 and 2916 cm^{-1}. S9 shows two bands at about 2852 and 2916 cm^{-1} as shown in Fig. 4.

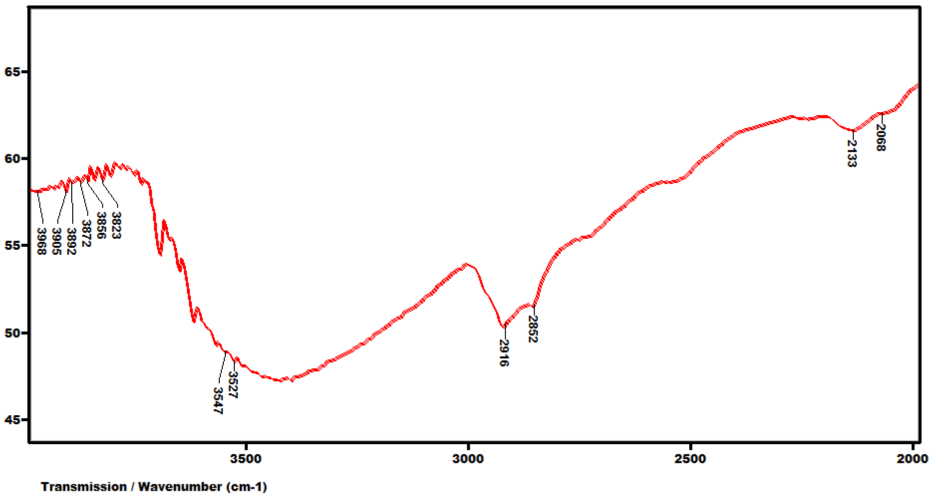

Figure 4: IR spectrum of S9 in the2000-4000 cm^{-1} range.

These bands are assigned to hydrocarbon adsorption bands; therefore their presence is not an indication of ZnO or ZSM. New research concerns the role of pigments containing zinc on the discoloration of paper [35, 36].A very interesting hypothesis is the possibility that the interaction of ZnO with paper humidity (water) produces $Zn(OH)_2$ or Zn(OH)-X [37].A infrared study of zinc oxide surfaces suggested that the 3670, 3620, 3555 and 3440 cm^{-1} bands could be ascribed to isolated and hydrogen-bonded surface hydroxyls [37]. Figure 4 shows the IR spectrum of S9 in the 2500-4000 cm^{-1}.

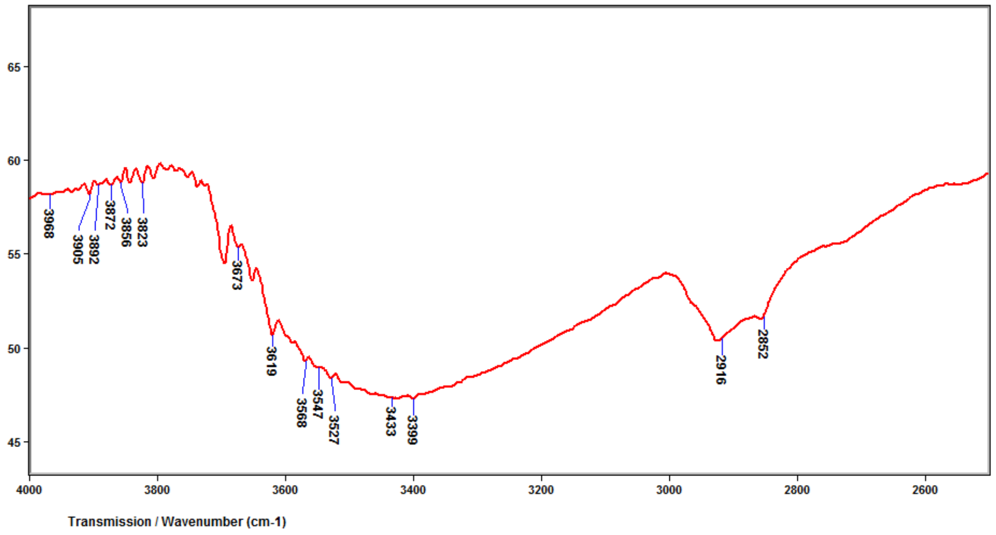

Figure 5: IR spectrum of S9 in the 2500-4000 cm^{-1} range.

It can be easily seen the existence of bands at 3673 and 3619 cm^{-1}. Therefore, the results give some support to the idea that we are in presence of a Zn-OH system.

There are five samples containing only aluminum (S6, S7, S11, S12 and S13) and one containing aluminum and sulfur (S8, see Appendix B).Probably it is aluminum oxide that was employed as filler but it is not seen as such in the IR spectrum. But we can approach this problem from another point of view. Tombácz mentions that aluminum oxides and hydroxides absorb at least a monolayer of water when exposed to moisture and that the top layer of oxides ions is converted to hydroxyl ions [38]. In the case of Al_2O_3 Kiselev and Uvarov state that in the IR spectrum of aluminum oxide three absorption bands corresponding to surface hydroxyl groups are observed in the region of 3500 to 3705 cm^{-1} [39]. Peri and Hannan report that absorption bands at 3698, 3737 and 3795 cm^{-1} correspond to “isolated” hydroxyl groups. Wang and Andrews listed three AlO-H stretching frequencies at 3787.0, 3752.4 and 3738.7 cm^{-1} [40]. Also they found bands for Al-O stretching

(921.5, 886.7 and 810.4 cm^{-1}) and weak O-H bending absorptions at 655.0 and 576.7 cm^{-1}. In Table 3 we have listed the bands of S6-S8 and S11-S13 that could be associated with Al(OH)$_x$ (we employed x because we do not know how aluminum is really present in the sample).

Table 3: Bands possibly associated with Al(OH)$_x$.

Sample	Bands (cm^{-1})
S6	3801, 3693
S7	3797, 3696
S8	3786, 3752, 3694
S11	3798, 3732, *c.* 3700
S12	3797, 3731, *c.* 3697, 659
S13	3796, 373, *c.* 3698, 809

We may conclude tentatively that aluminum can be detected using IR only if enough Al-OH is formed through the time. Then, if you cannot get SEM results, a good approach consists in looking for these bands after discarding the presence of kaolinite (see below).

There are seven samples containing aluminum and silicon. We have hypothesized that the chemical compound containing both elements is kaolinite ($Al_2Si_2O_5(OH)_4$). In the case of kaolinite, it has been suggested that '*the presence of just one peak*[of the four OH stretching bands] *in the spectrum can be related to the position of the kaolin filler into the fibers*' [9]. In our case, if kaolinite is present in one or more samples some of its IR bands should be visible because of the mechanical treatment of the paper to fabricate the KBr pill.

The IR spectrum of white powdery aggregate kaolinite from Margaritas mine (Mexico) presents the following band positions: 3693s, 3655sh, 3620, 1115s, 1023s, 1006s, 939, 914s, 792, 753, 696, 642, 600sh, 536s, 429 cm^{-1}. Saikia and Parthasasarathy analyzed the IR spectrum of six kaolinite samples from Assam and Meghalaya (India)and found that 'a typical spectrum of kaolin show four bands, at 3697, 3669, 3645 and 3620 cm^{-1}' [41]. Therefore we shall proceed as follows. Using the above data together with those of other references searched for the following bands (approximate values): 3620 (inner OH stretching), 3695 (in-phase stretching of inner surface OH groups), 3690 (Si-OH stretching mode), 3669 (anti-phase stretching of inner surface OH groups), 3652 (anti-phase stretching of inner surface OH groups), 1134 (Si) 1112 (symmetric stretch of equatorial Si-O bonds), 1103 (?), 1032 (anti-symmetric stretch of equatorial Si-O bonds), 1008/1016 (anti-symmetric stretch of equatorial Si-O bonds), 1000 (Si), 941 (Al-OH), 917 (Al-OH), 900-950 (H atoms), 566 (Al) and 542 (Si-O-Al) [9, 21, 41-43]. We analyzed the corresponding spectra searching for bands suggesting the presence of kaolinite. Table 4 shows the results.

Table 4: Bands possibly associated with kaolinite.

Sample	IR bands (cm^{-1})
S6	3693, 3669, 3649, 1027, 1000, 917, 593.
S7	3399, 3697, 3645, 1115, 1031.
S8	3694, 3651, 3619, 1113, 1031, 941, 912, 536.
S11	3699 (?), 3645, 1114, 1031.
S12	3667, 3644, 1116, 1037 (?).
S13	3667, 3644, 3625 (?), 1117, 1036 (?).

We can see that each sample has its own set of frequencies possibly associated with kaolinite. Besides the possibility of a different location of this compound on the cellulose fibers we have no other explanation for this fact. The information to solve this problem seems to be a detailed infrared study of different mixtures and fabrication modes of pure cellulose and kaolinite. The influence of crystallization water should be included. For the IR bands of cellulose we have examined a book and some papers [9, 10, 18, 44]. Valli et al. mention cellulose signals at 1158, 1106, 1055, 1030 (all C-O stretching), 1647 and 1546 cm^{-1} (primary and secondary amide bands from protein glue) [9]. Brittain cited bands at 1025 and 3330 cm^{-1} [18]. Ferrer and Vila found bands at 1158, 1111, 1061, 1036 and 3346 cm^{-1} [10]. We searched only for these bands. Table 5 shows our findings.

Table 5: Suggested cellulose bands.

Sample	Cellulose bands (cm^{-1})
S1	1641, 1162, 1113, 1057, 1039
S2	1645, 1161, 1112, 1061, 1031
S3	1644, 1555, 1113, 1061, 1031
S4	1647, 1114, 1064, 1030
S5	1645, 1113, 1055, 1031
S6	1649, 1159
S7	1644, 1115, 1062, 1031
S8	1639, 1545, 1113, 1060, 1031
S9	1647, 1113, 1053, 1030
S10	1649, 1544, 1112, 1060, 1031
S11	1646, 1555, 1114, 1060, 1031
S12	3333, 1643, 1116, 1065, 1037
S13	1646, 1554, 1117, 1064, 1036

We can see that only the band at about 1647 cm^{-1} is common to all samples. We shall not attempt to provide assignations to the cellulose bands or to analyze more bands because we are preparing a larger set of diverse papers for a study centered specifically on cellulose. But what Table 5 tells us is that not all the papers seem to be exactly the same.

The main conclusion of this study is that the paper used to print these stamps originated in more than one paper factory. The reason (or reasons) for this lack of uniformity in paper fabrication was probably one of the results of the state of disorder in which Germany was immersed at that time.

APPENDIX A: Infrared spectra of samples S1-S13.

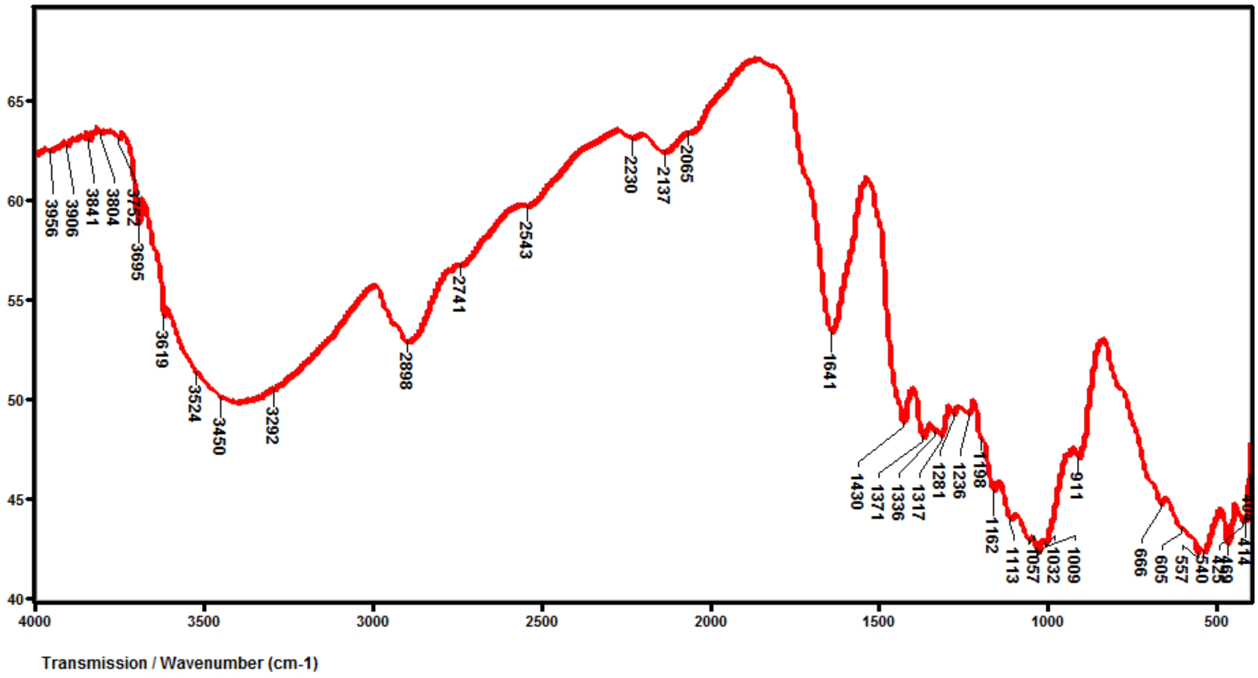

Figure A1: IR spectrum of sample S1.

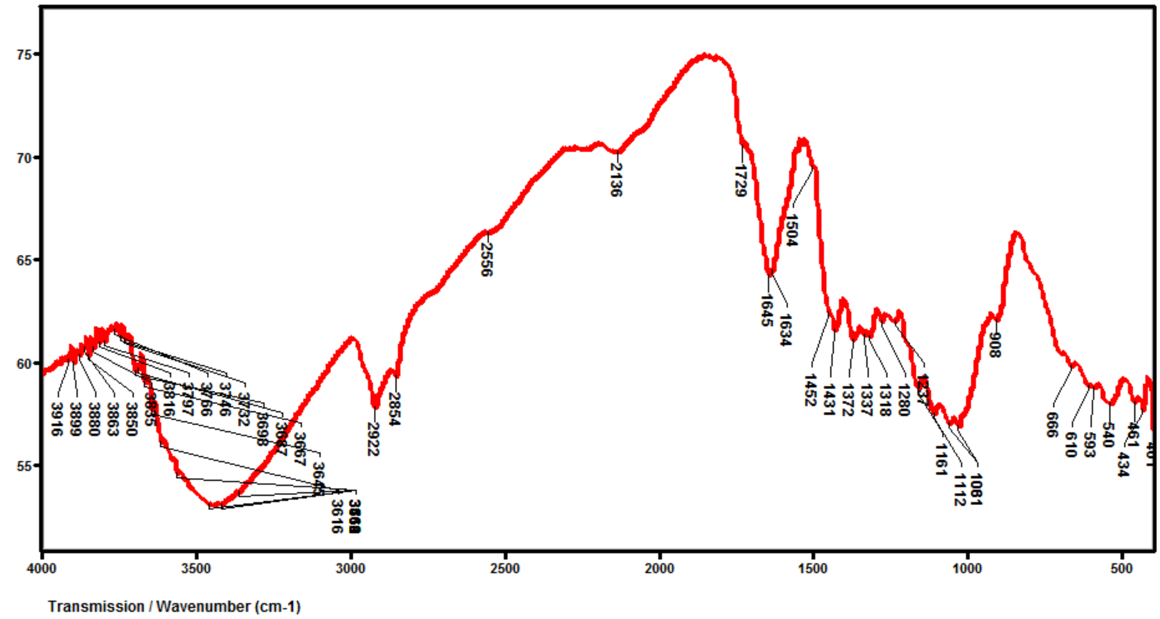

Figure A2: IR spectrum of sample S2.

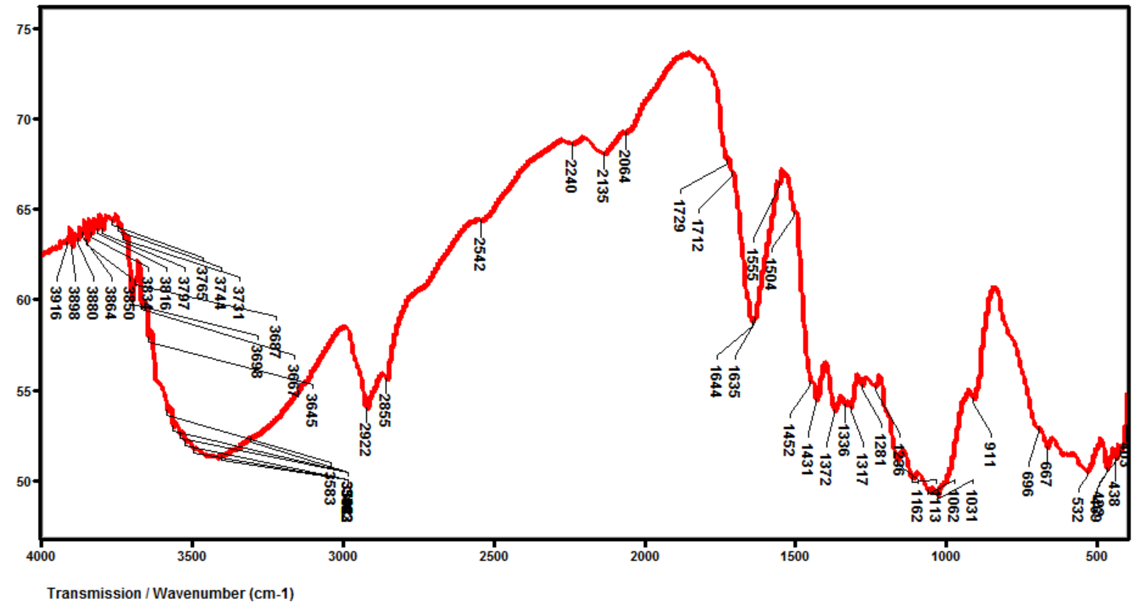

Figure A3: IR spectrum of sample S3.

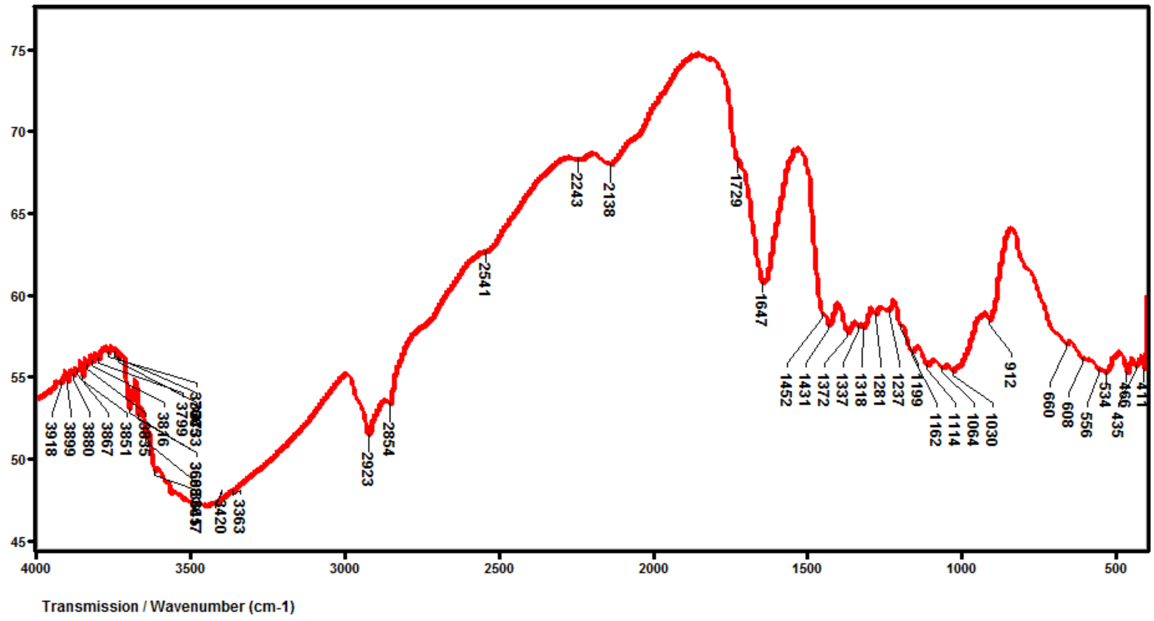

Figure A4: IR spectrum of sample S4.

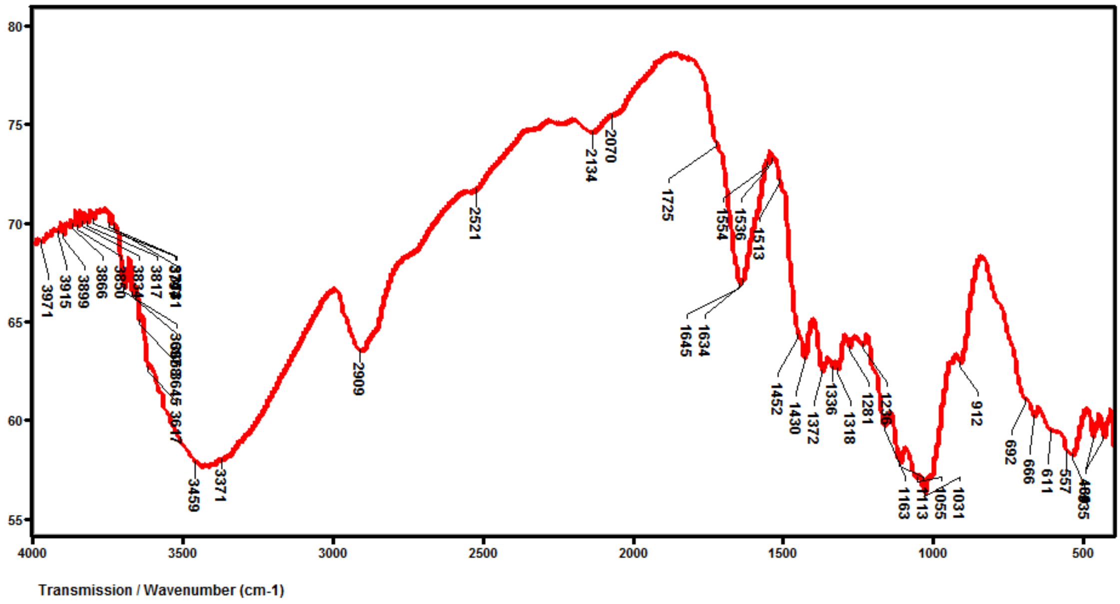

Figure A5: IR spectrum of sample S5.

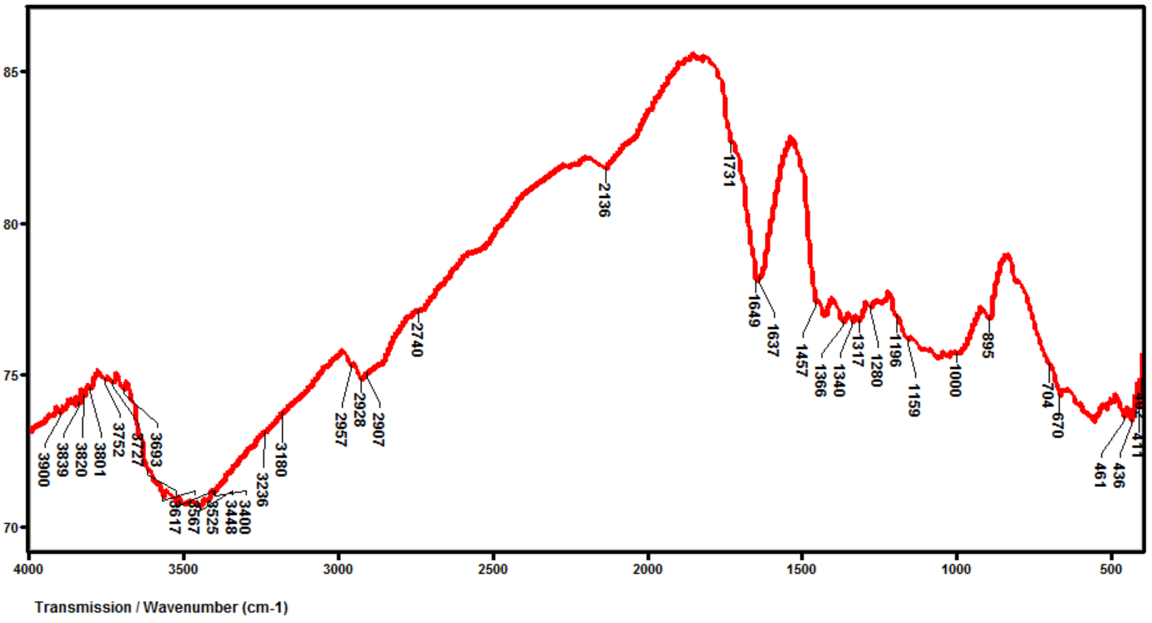

Figure A6: IR spectrum of sample S6.

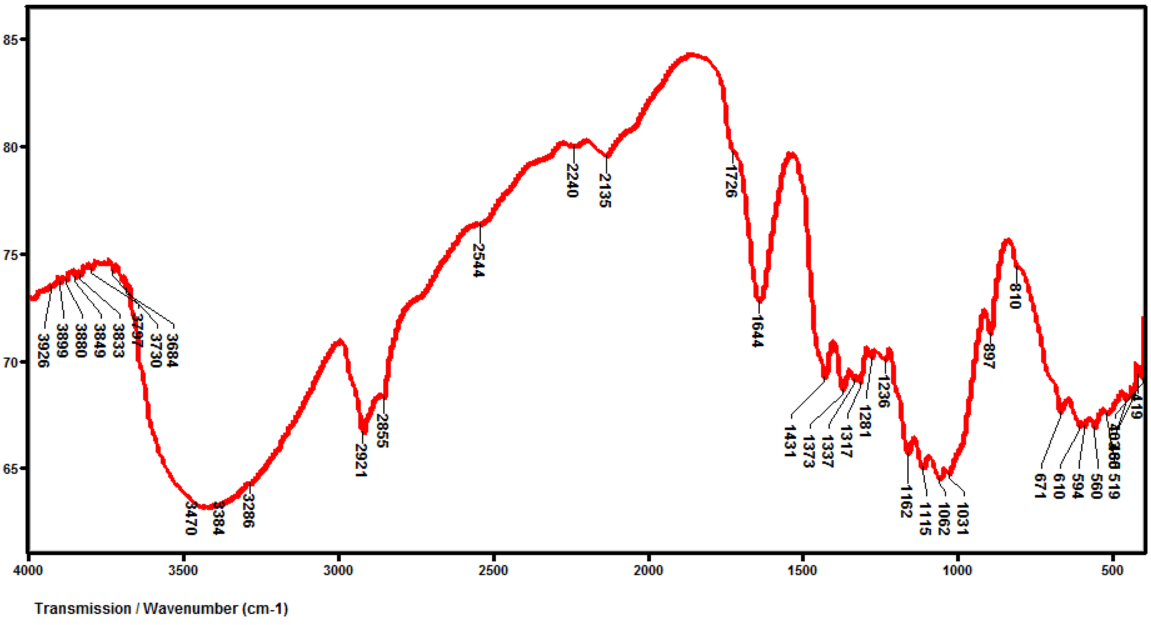

Figure A7: IR spectrum of sample S7.

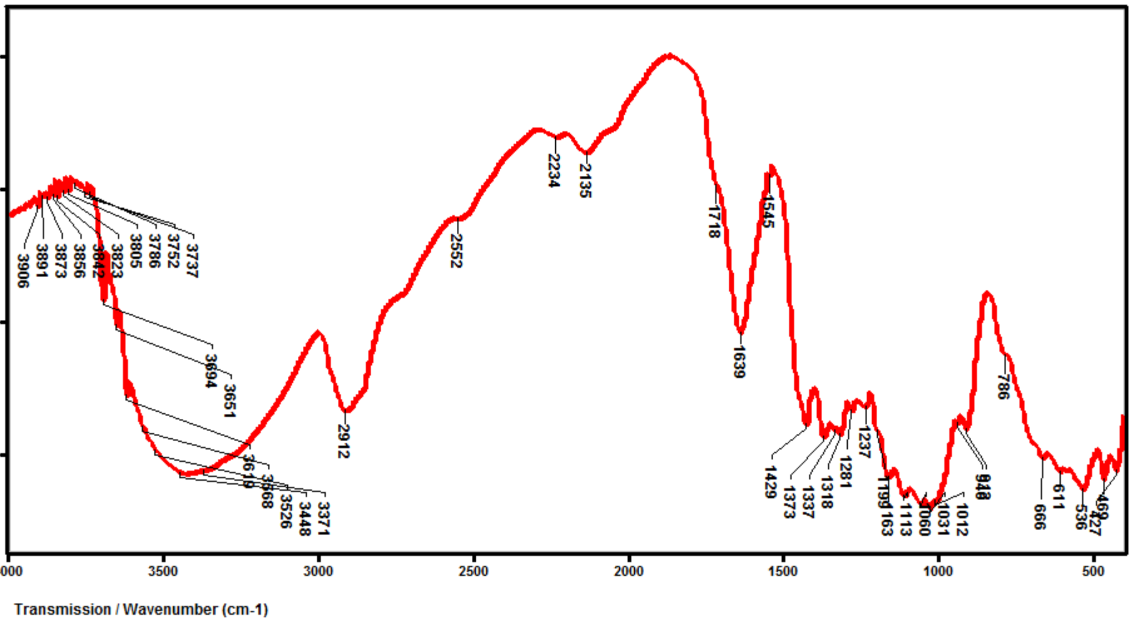

Figure A8: IR spectrum of sample S8.

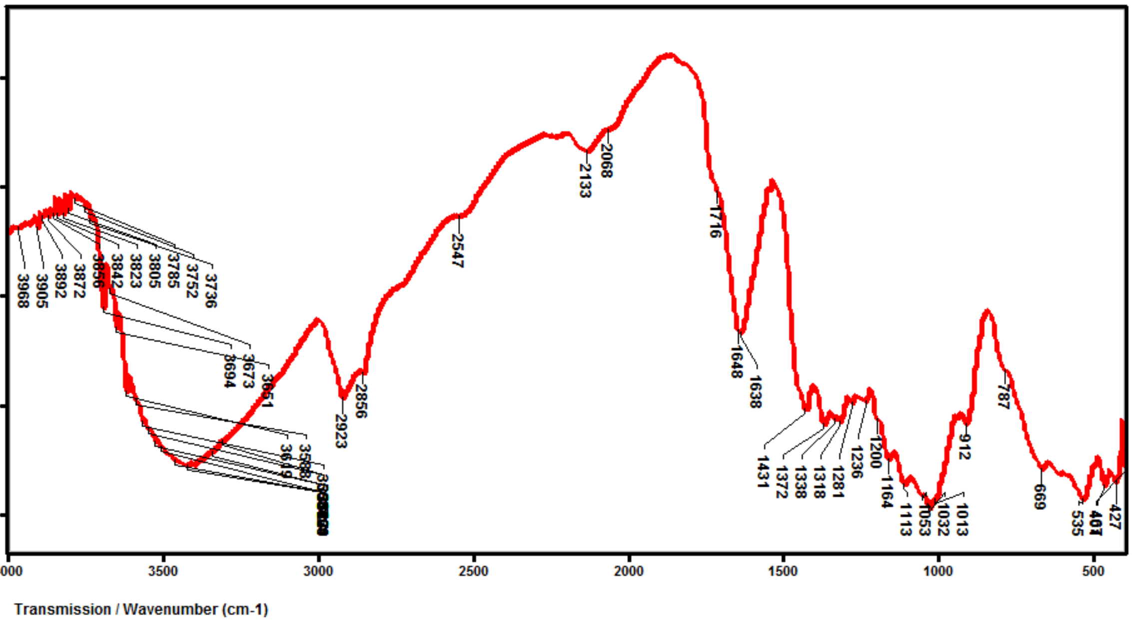

Figure A9: IR spectrum of sample S9.

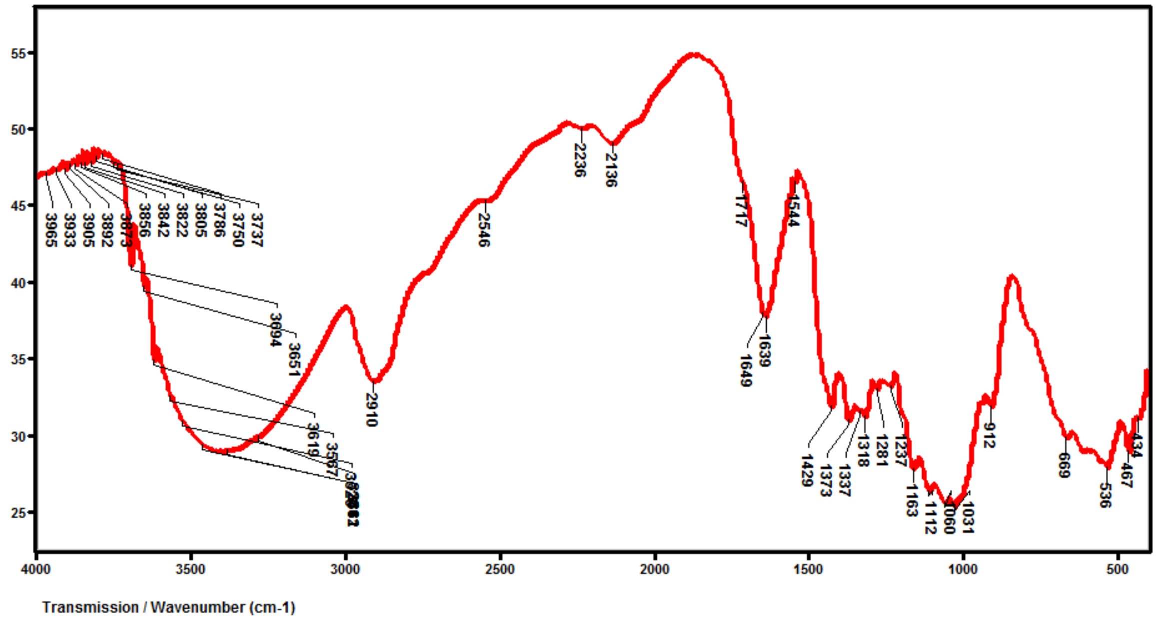

Figure A10: IR spectrum of sample S10.

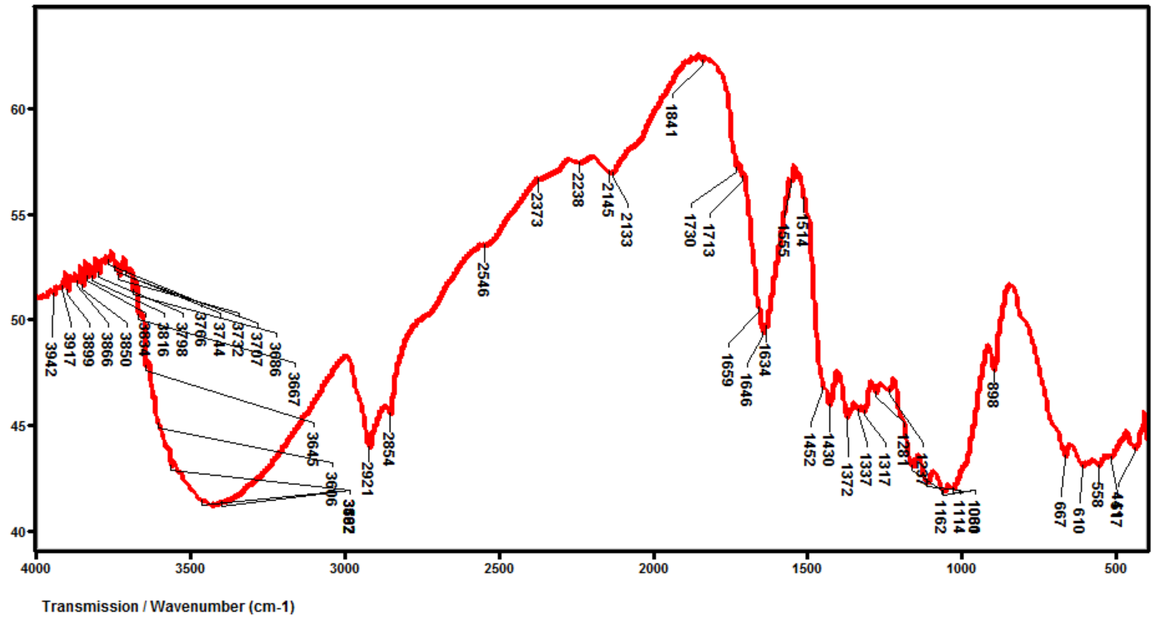

Figure A11: IR spectrum of sample S11.

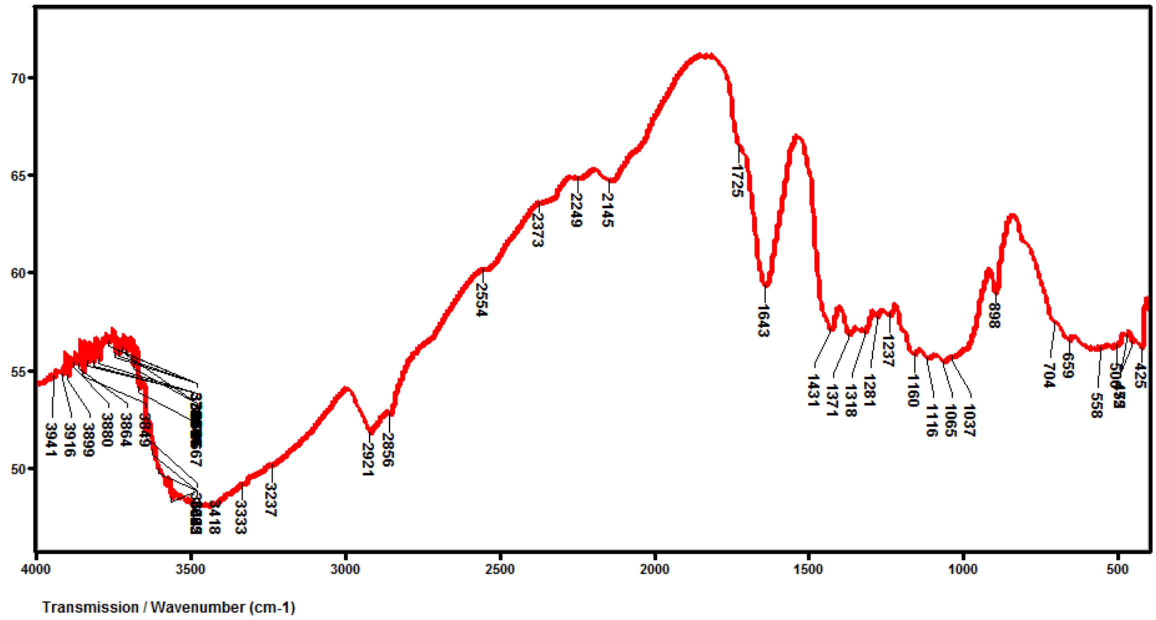

Figure A12: IR spectrum of sample S12.

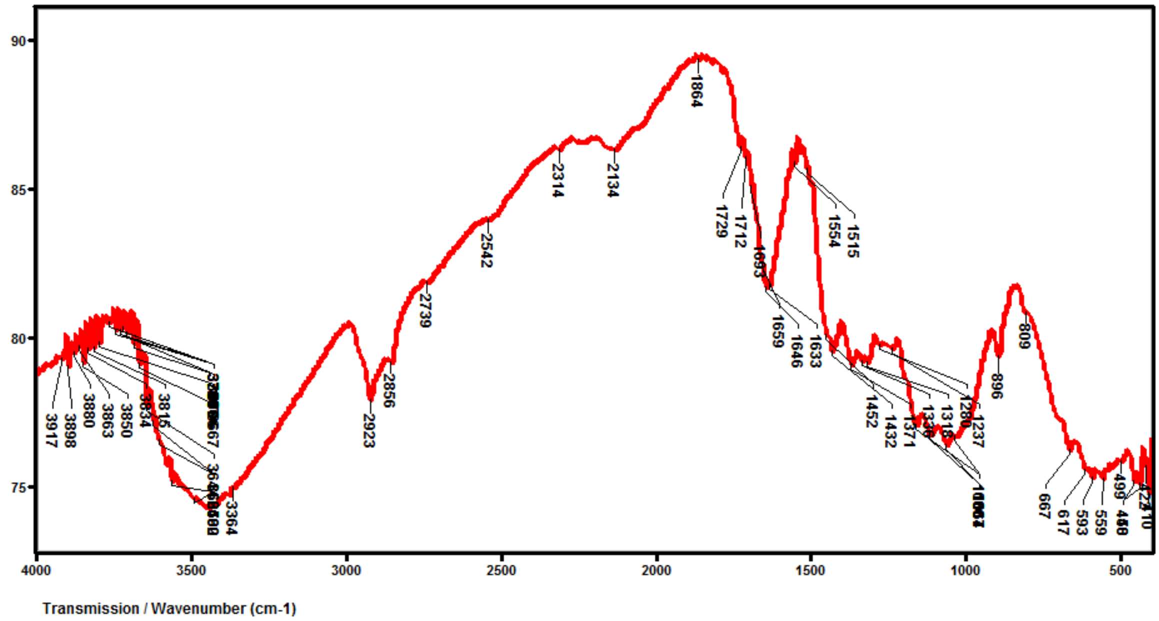

Figure A13: IR spectrum of sample S13.

Appendix B. SEM results for samples S1-S13.

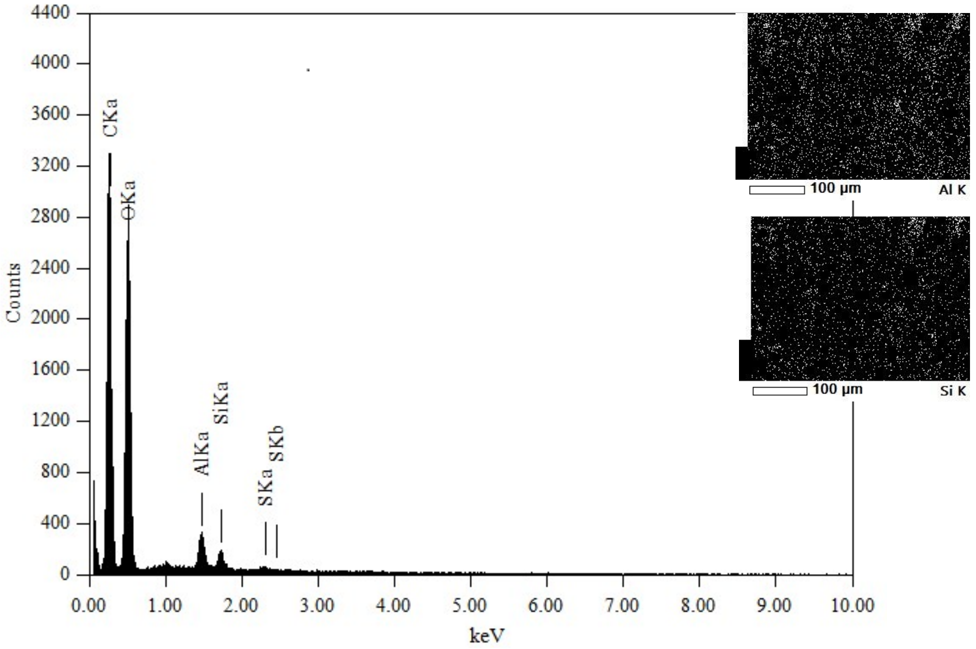

Figure B1: SEM results for S1. At the right side we show the distribution of important elements within the sample.

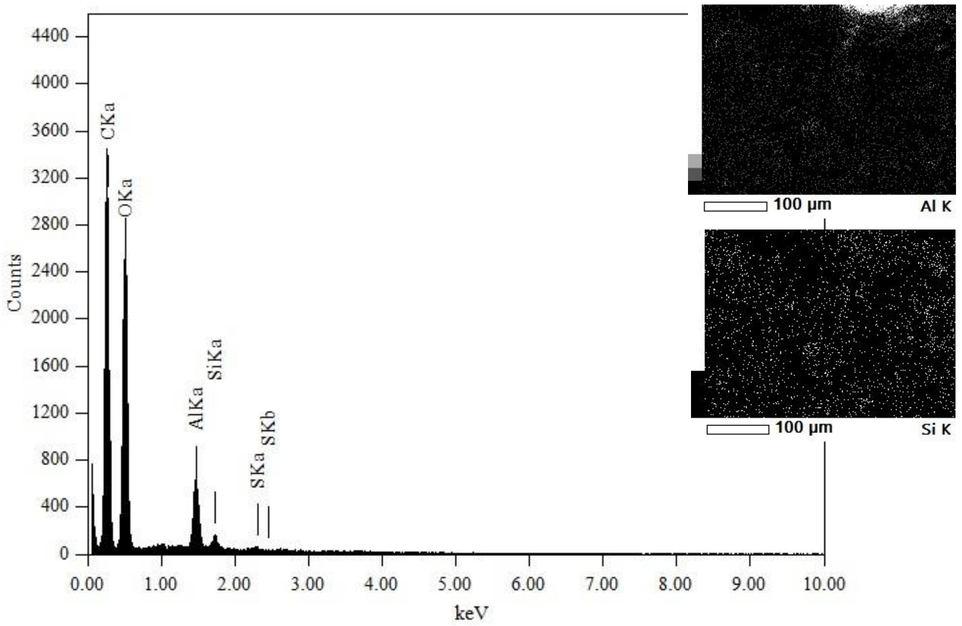

Figure B2: SEM results for S2. At the right side we show the distribution of important elements within the sample.

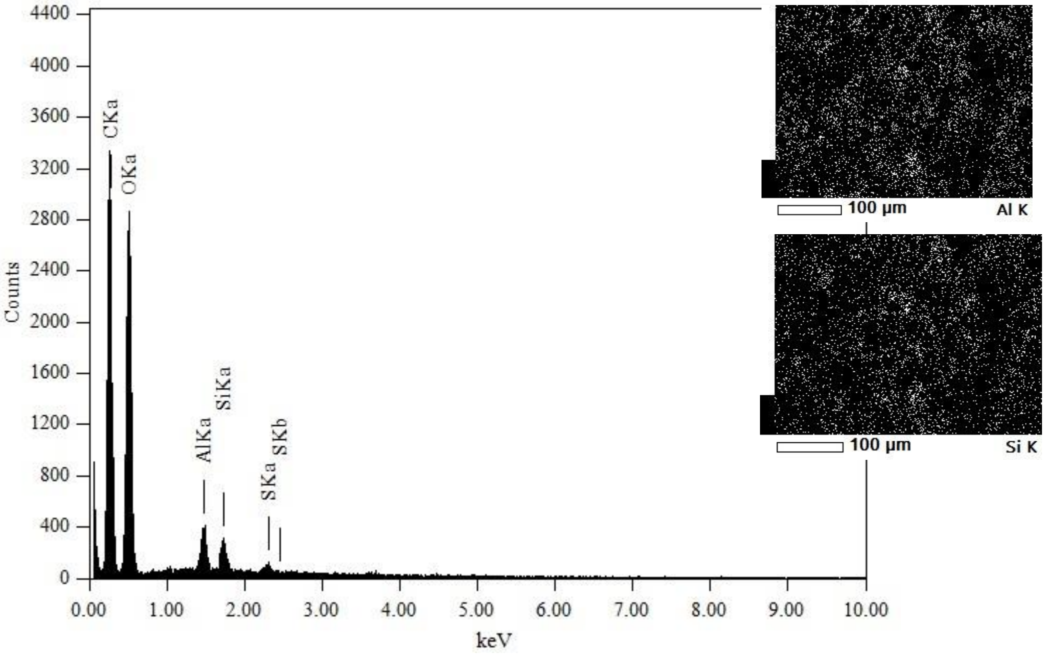

Figure B3: SEM results for S3. At the right side we show the distribution of important elements within the sample.

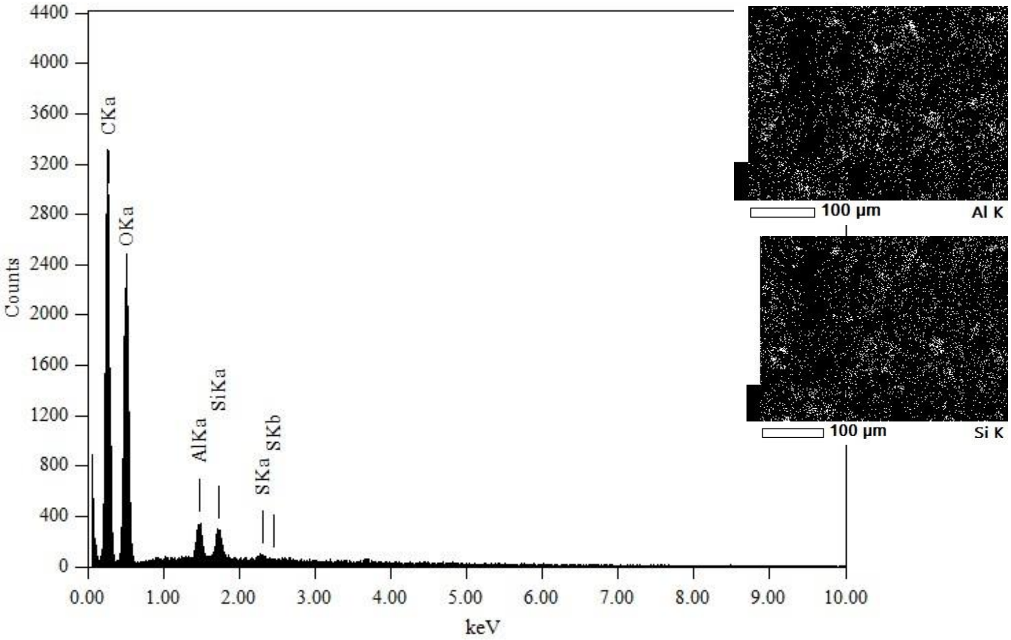

Figure B4: SEM results for S4. At the right side we show the distribution of important elements within the sample.

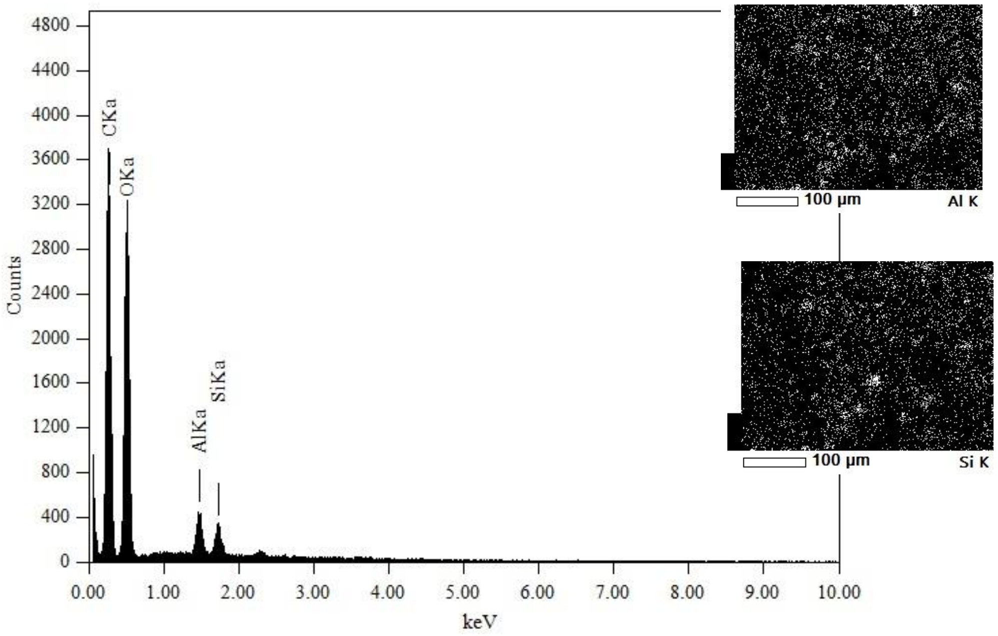

Figure B5: SEM results for S5.At the right side we show the distribution of important elements within the sample.

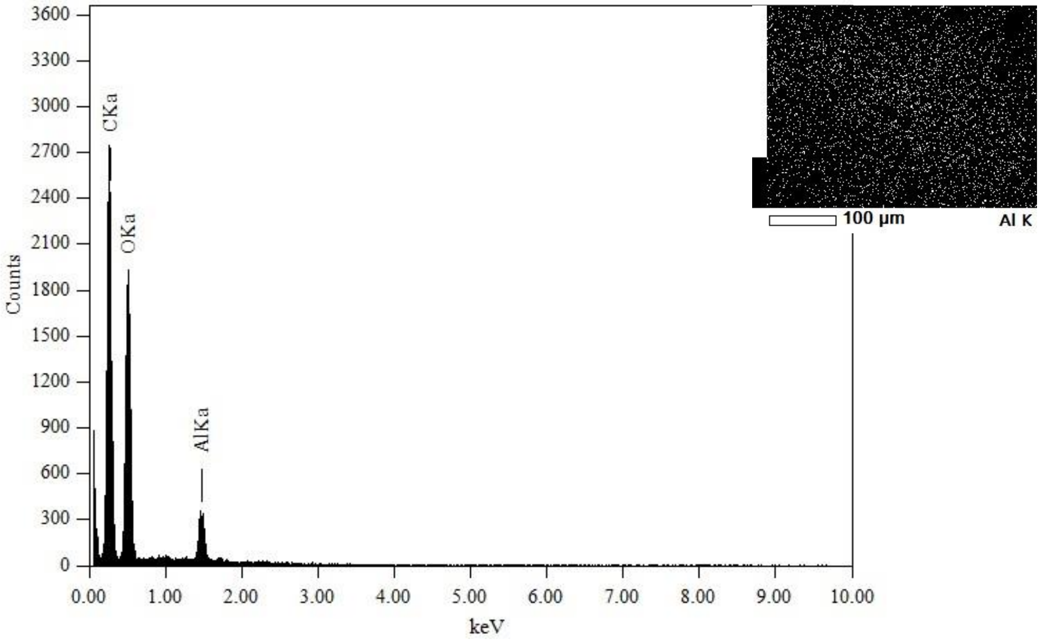

Figure B6: SEM results for S6. At the right side we show the distribution of important elements within the sample.

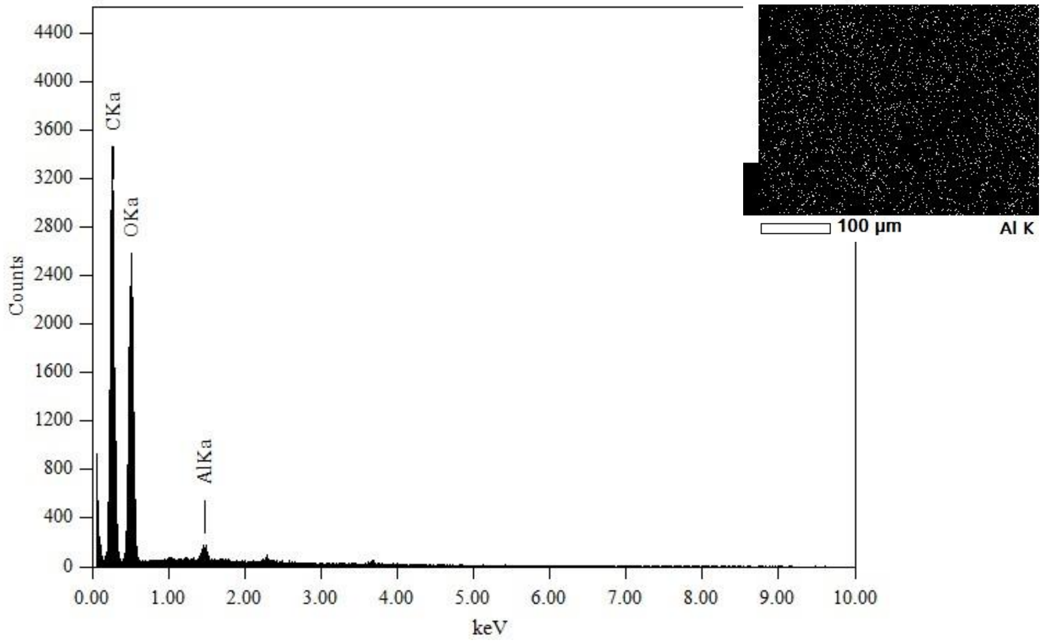

Figure B7: SEM results for S7. At the right side we show the distribution of important elements within the sample.

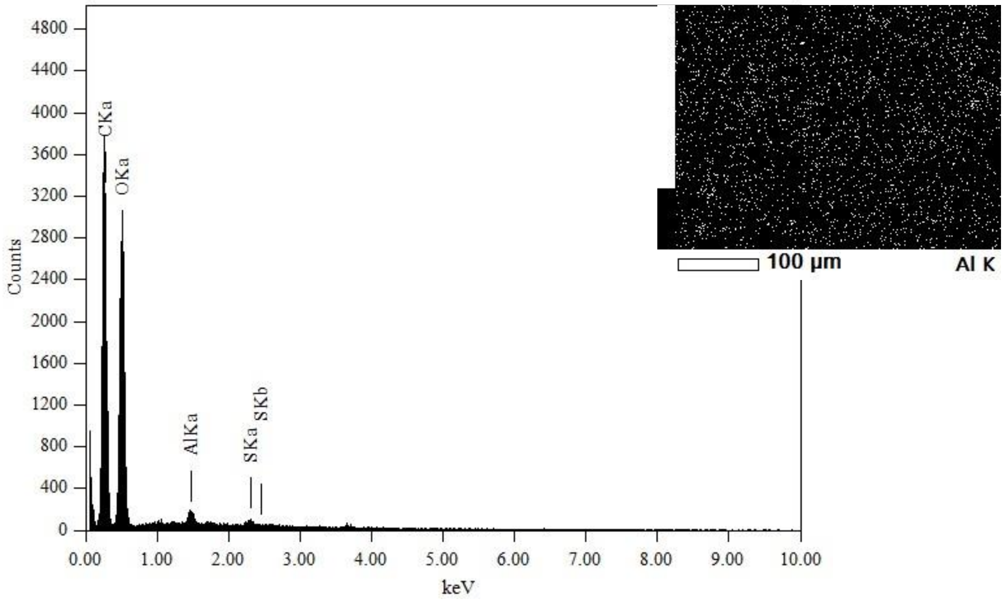

Figure B8: SEM results for S8.At the right side we show the distribution of important elements within the sample.

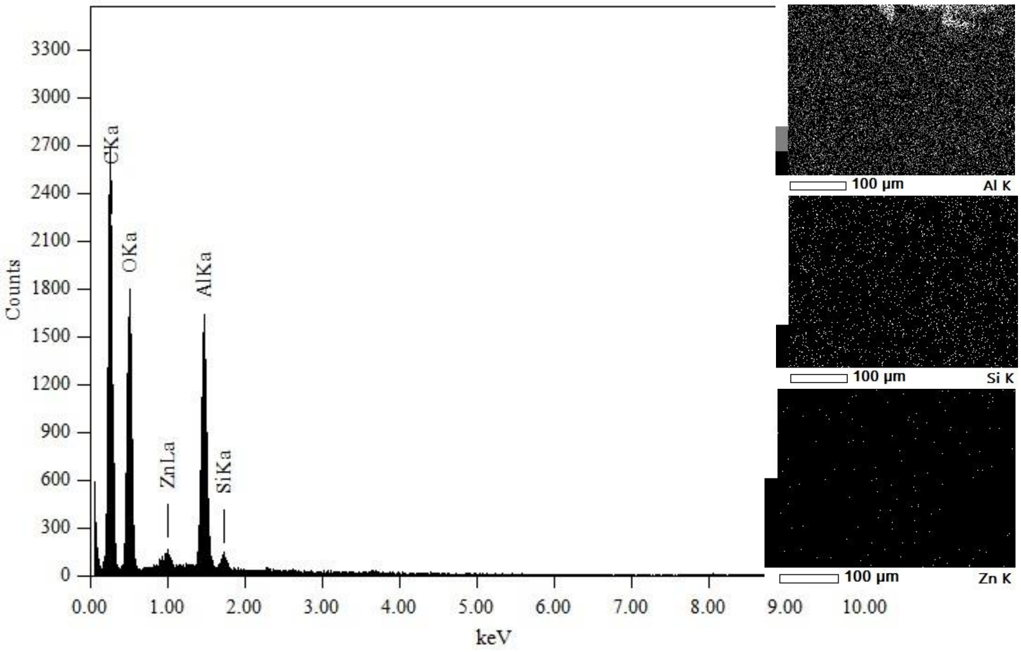

Figure B9: SEM results for S9. At the right side we show the distribution of important elements within the sample.

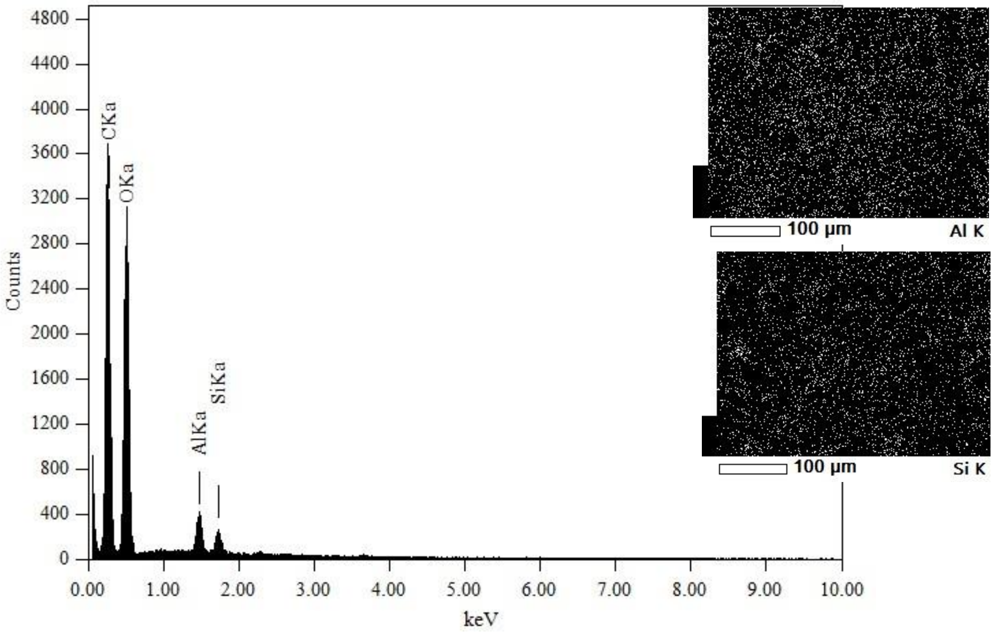

Figure B10: SEM results for S10. At the right side we show the distribution of important elements within the sample.

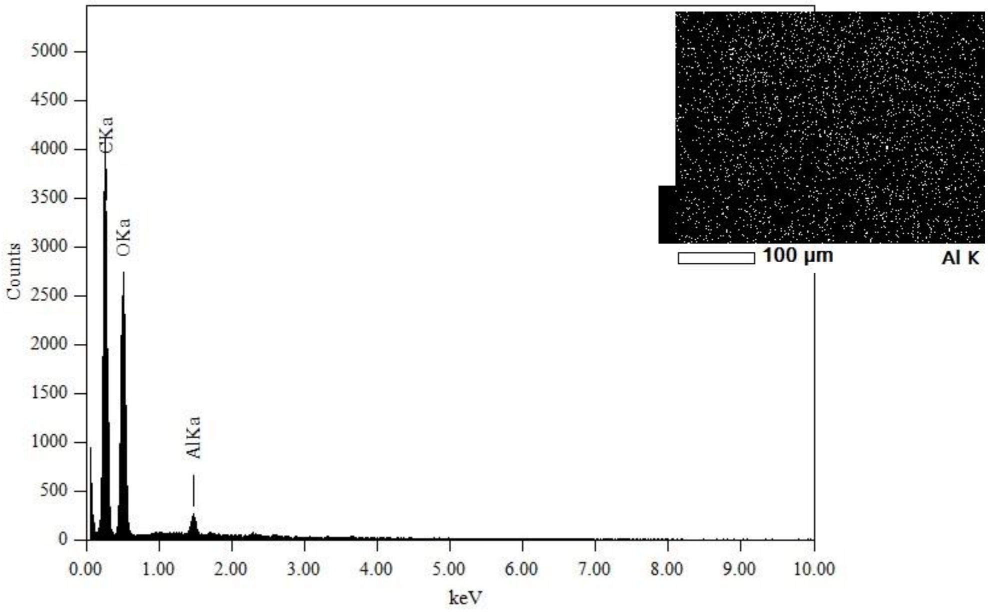

Figure B11: SEM results for S11. At the right side we show the distribution of important elements within the sample.

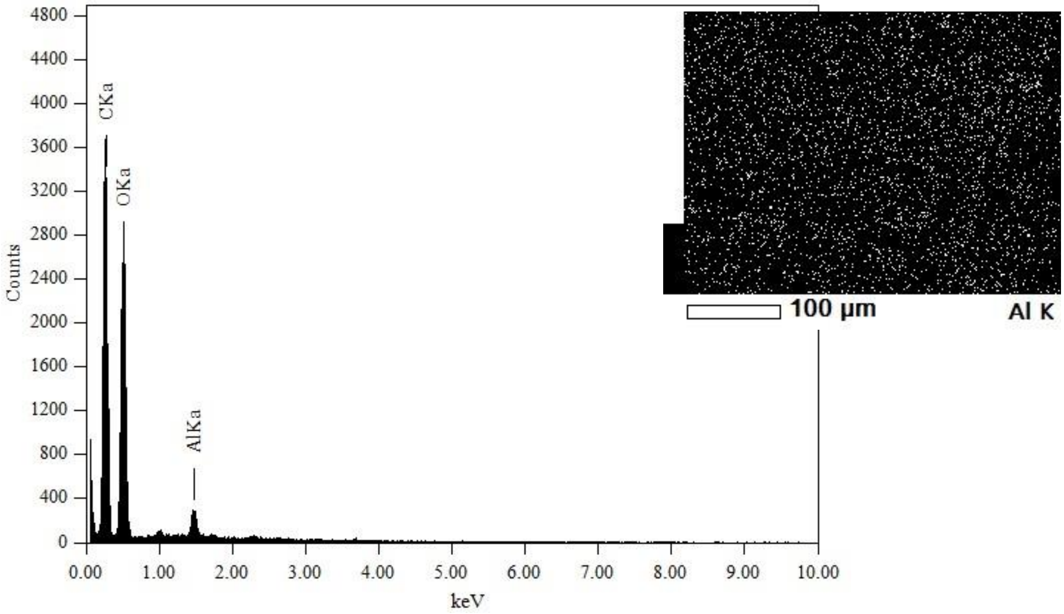

Figure B12: SEM results for S12. At the right side we show the distribution of important elements within the sample.

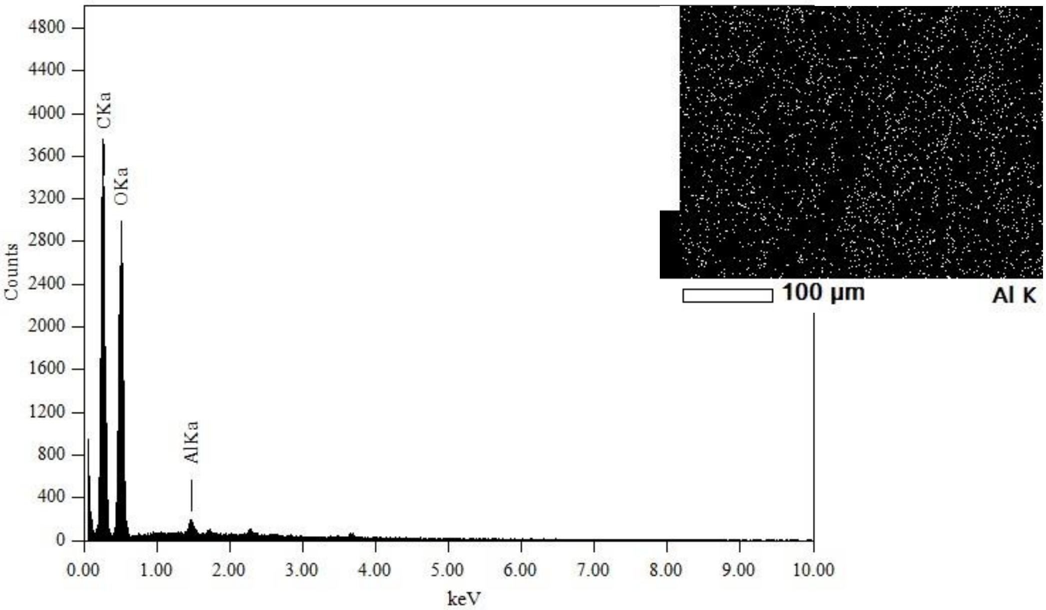

Figure B13: SEM results for S13. At the right side we show the distribution of important elements within the sample.

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