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# Characterization of Solids Deposited on the Modular Caustic-Side Solvent Extraction Unit (MCU) Strip Effluent (SE) Coalescer Media Removed in April 2015

F. F. Fondeur

June 2016

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# REVIEWS AND APPROVALS

AUTHORS:

F. F. Fondeur, Advanced Characterization and Processing Date

TECHNICAL REVIEW:

T. B. Peters, Advanced Characterization and Processing, Reviewed per E7 2.60 Date

APPROVAL:

B. J. Wiedenman, Manager Date
Advanced Characterization and Processing

D. E. Dooley, Director Date
Environmental & Chemical Process Technology Research Programs

J. S. Contardi, Manager Date
Savannah River Remediation Engineering

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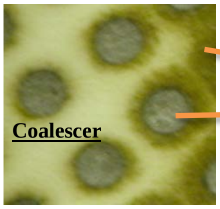
# EXECUTIVE SUMMARY

On June 2015, Savannah River National Laboratory (SRNL) received a Strip Effluent (SE) coalescer (FLT-304) from MCU. That coalescer was first installed at MCU in late October 2014 and removed in April 2015. While processing approximately 48,700 gallons of strip solution, the pressure drop steadily increased linearly from 1 psi to near 16 psi (the administrative limit is 17 psi) with the total filtrate volume (2.1E-4 psi/gal of filtrate). The linear behavior is due to the combined effect of a constant deposition of material that starts from the closed-end to the mid-section of the coalescer reducing the available surface area of the coalescer for fluid passage (linearly with filtrate volume) and the formation of a secondary emulsion (water in NG-CSSX) on the fibers of the coalescer media. Both effects reduced the coalescer porosity by at least 13% (after processing 48,700 gallons). Before the coalescer was removed, it was flushed with a 10 mM boric acid solution to reduce the dose level. To determine the nature of the deposited material, a physical and chemical analysis of the coalescer was conducted.

Characterization of this coalescer revealed the adsorption of organic containing amines (secondary amides and primary amines), TiDG, degraded modifier (with no hydroxyl group), MaxCalix, and oxidized hydrocarbon (possibly from Isopar™L or from lubricant used at MCU) onto the coalescer media. The amide and amines are possibly from the decomposition of the suppressor (TiDG) as follows:

$$\text{(R-NH-)}_2\text{C=NR} + \text{H}_2\text{O (caustic)} \Rightarrow \text{R-NH-CONH-R} + \text{R-NH}_2 \quad \text{(R=aliphatic hydrocarbon)}$$

The modifier and MaxCalix were the largest components of the deposited organic material, as determined from leaching the coalescer with dichloromethane. Both the Fourier-Transformed Infrared (FTIR) and Fourier-Transformed Hydrogen Nuclear Magnetic Resonance (FT-HNMR) results indicated that some of the modifier was degraded (missing their OH groups). The modifier was observed everywhere in the examined coalescer pieces (FTIR) while the TiDG and its decomposition products were observed at the entrance discs of the coalescer. A summary of the deposits found in the coalescer is shown below.



Composition of the deposits found on the coalescer

Gray color: Mercury, steel debris, titanium, aluminum, silicates, modifier, and MaxCalix

Entrance Discs:
Primary amines, poly-urea, TiDG, modifier, oxidized lubricant and silicates

A solvent trim (a cocktail of solvent components with a high concentration of modifier) was added to the solvent on 2/22/2015. It is believed that the trim did not mix completely with the solvent and that it was subsequently spread around the MCU components including the coalescers, where it may have deposited.

Chronologically, the modifier, the TiDG's decomposition products and silicates deposited on the entrance discs first and after the pressure drop increased significantly, parts of the coalescer media detached itself from the central porous steel mandrel and a significant amount of steel debris, mercury, titanium, and additional aluminum and silicates deposited on the coalescer. This is responsible for staining the coalescer media with a gray color.

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| | |
|---|---|
| ARP | Actinide Removal Process |
| CSSX | Caustic-Side Solvent Extraction |
| DI-DD | De-ionized Doubled-distilled |
| DSS | Dissolved salt solution |
| EDS | Energy dispersive spectroscopy |
| FTIR | Fourier transformed infrared Spectroscopy |
| FT-HNMR | Fourier Transformed Hydrogen Nuclear Magnetic Resonance |
| IC | Ion chromatography |
| ICP-AES | Inductively coupled plasma atomic emission spectroscopy |
| MCU | Modular caustic side solvent extraction unit |
| MST | Monosodium titanate |
| NAS | Sodium aluminosilicate |
| NGS | Next Generation Solvent |
| PPS | Polyphenylene sulfide |
| SB | Salt batch |
| SE | Strip effluent |
| SEM | Scanning electron microscopy |
| SRNL | Savannah River National Laboratory |
| XRD | X-ray diffraction |

## 1.0 Introduction

During operations of the Modular Caustic-Side Solvent Extraction [CSSX] Unit (MCU), pressure drops across the MCU coalescers have been found to increase over time as the volume of solution processed through the coalescer increases. Such pressure drop increases have been attributed to pluggage and /or fouling of the coalescers associated with accumulation of inorganic /organic compounds and /or changes in the characteristics of the coalescer fibers. The net result is reduction in coalescer porosity.

When pressure drops reach a level impeding normal operations, the coalescers are replaced so that effective processing operations can resume. Alternatively, the coalescers are also replaced opportunistically as a routine maintenance practice when other facility outages occur. This applies to the Decontaminated Salt Solution (DSS) coalescers, the SE coalescers, and the DSS coalescer prefilters.

In this document, characterization of spent SE coalescer samples is reported. The SE coalescer (FLT-304) examined in this report was removed in April 2015 (after it has processed 48,700 gal). As a back drop, a recent history of previous coalescer characterization analyses is shown in Table 1. Another objective is to determine any spatial variation in the solid deposition within a coalescer.

**Table 1. A recent history of the coalescer sent to SRNL for characterization**

| Coalescer Type<br>DSS (20 µ, 40” long)<br>SE (10 µ, 24” long) (gallons processed K= 1000 gallons) | Solution that contacted the coalescer | Entrained Solids Found and Approximate Concentration on the Coalescer |
|---|---|---|
| DSS [1] | Simulated salt simulant (1.9 M [OH]) | 40 – 170 g of NAS |
| DSS[2] | Water with MST | 15 -20 g of MST |
| DSS coalescer A[3] | Simulated salt simulant (10K) | Sodium Carbonate and Boehmite |
| DSS coalescer B (40K)[3] | Salt simulant | Sludge, Silica, Bayerite (1.4 g), and Titania |
| DSS coalescer C (80K)[3] | Salt simulant | Bayerite (7.8 g) |
| SE coalescer Alpha (9.5K)[4] | 1 mM Nitric Acid | Bayerite $Al(OH)_3$ and NAS |
| SE coalescer Beta (40K)[4] | 1 mM Nitric Acid | Bayerite and Boehmite $Al(OH)_3$ |
| DSS[5] | Saltbatch 2 & 3 | $Al(OH)_3$, NAS, and Titania |
| DSS[5] | Saltbatch 3 | $Al(OH)_3$, Silica, and Titania |
| DSS[6] | Salt Batch 3 | No solids found |
| DSS (800K)[7] | Salt Batch 5 & 6 | Sludge, Titania, NAS, Silica, Oxalate, $Al(OH)_{3,}$ and modifier |
| SE (65K)[7] | 3 mM boric acid (SB 5/SB 6) | Sludge, Titania, NAS, Silica, iron oxide, and modifier |
| DSS (700K) and SE (40K) [8] | Salt Batch 6 and 7 | $Al(OH)_3$, Titania (48 mg /mL of coalescer) |
| SE[9] | 3 mM boric acid (SB 6 / SB 7) | $Al(OH)_{3,}$ Silica (1.5 mg/ mL of coalescer) |
| SE (30K) [10] | 3 mM boric acid (SB 7) | Stainless steel debris. Low solids concentration |

## 2.0 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. [11] This report was developed in accordance with the protocols identified in Task Technical and Quality Assurance Plan SRNL-RP-2013-00536.[12]

## 3.0 Background Information

The coalescer samples addressed in this report are those associated with MCU operations during part of Salt Batch 7 (SB 7). Salt batches are blended in Tank 21, sampled, and analyzed for qualification. This designates the Salt Batch number. Once qualified the batch (or partial batch) is moved from Tank 21 to

Tank 49. If partial batches are sent from Tank 21 to Tank 49, then an alphabetical designation is attached to the number. Salt batches may be adjusted with 50 wt% NaOH solution to reach a free hydroxide concentration of 2.0 ± 0.2 M.[9,13] A batch number is assigned to the content of Tank 49H when its compositional chemistry changes slightly due to the content transfer from Tank 21H. In the amount transferred along with cold chemical additions, the concentration of major chemical components of concern such as hydroxyl, sodium, carbonates, aluminates, phosphate, nitrate, nitrite, and sulfate are typically similar from batch to batch. Batches with slight change in chemical composition are identified with an alphabet letter (for example A, B, C, and D). A history of the coalescer media from which the samples in this study were collected are summarized below.

The SE coalescer was installed in October 2014 and remained in operations until April 2015. A total of 48.7K gallons of strip solution (mostly SB 7) were processed through the SE coalescer and the corresponding maximum pressure drop across the coalescer was approximately 16 psi (see Fig. 1) which is close to the administrative pressure drop limit.[14] Before it was removed, the SE coalescer (FLT-304) was backed flushed with 10 mM boric acid to reduce its radioactive load with the risk of removing solids deposited on the entrance discs of the coalescer media. A quick glance at Fig. 1 reveals the pressure drop data consists of two distinct lines (the second straight line is noticeable after 33,500 gallons of filtrate). The appearance of the second line coincides with the solvent trim addition to the SHT on Feb. 22, 2015. The two lines in Fig.1 may indicate a sudden increase in particle concentration of the solution that is filtered. The data does not appear to support other plugging mechanisms such as surface pore blocking or deep bed filtering.[15] The data can be fit with the mathematics of “cake filtration” or that of Kozeny-Carman equation for a porous media.[16] Assuming Kozeny-Carman (see Table 2), the porosity of the coalescer dropped drastically from the first line to the second line in Fig. 1. Please recall that prior to sampling (but following completion of the normal SE operations), the SE coalescer was rinsed with approximately 500 gallons of a solution containing 10 mM boric acid to reduce the gamma emitting radionuclide concentration in the media. That rinsing can possibly remove any cake film on the inner surface of the SE coalescer.

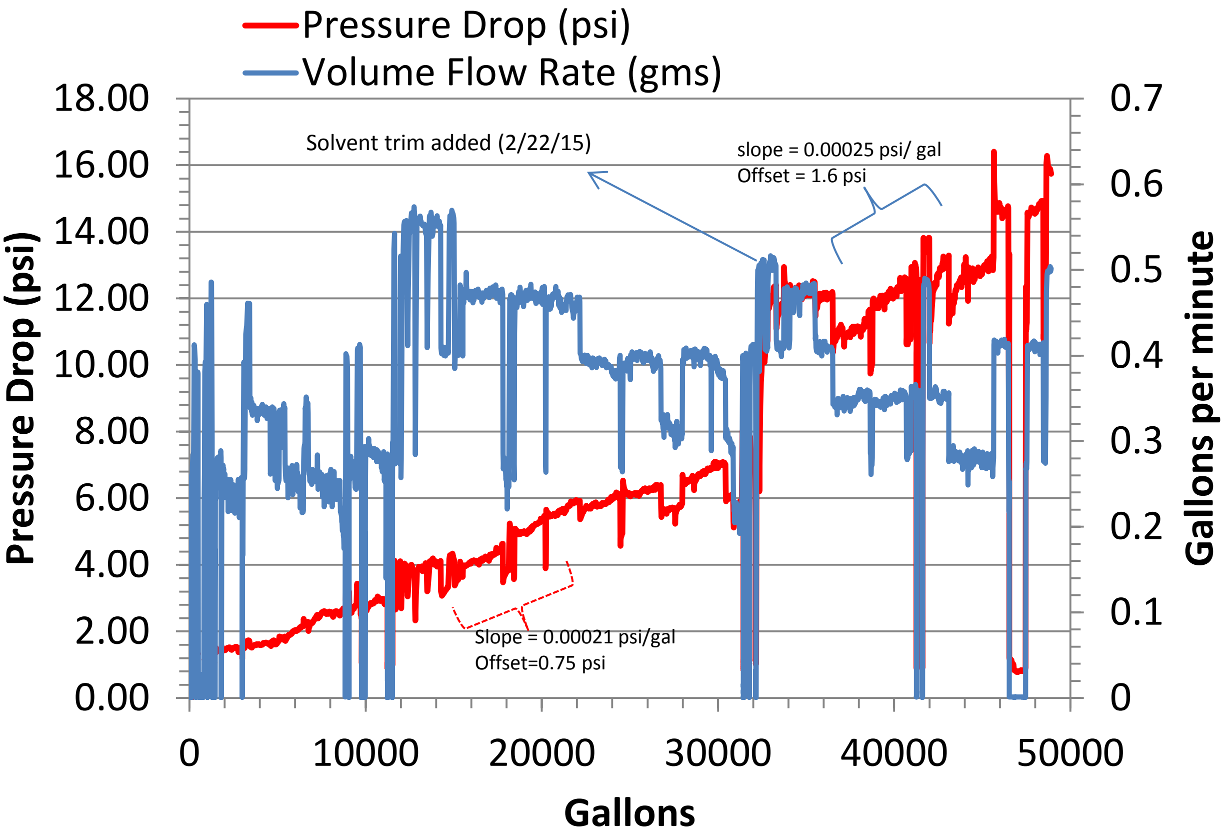


**Figure 1 Pressure Drop Across the April 2015 SE Coalescer**

**Table 2. Calculated porosity using the Kozeny-Carman equation of Figure 1**

| Volume (gallons) | Pressure Drop (psi) | Porosity |
|---|---|---|
| 6412 | 2.0967 | 0.90 |
| 28927 | 6.828 | 0.83 |
| 37818 | 11.05 | 0.79 |
| 45658 | 13.01 | 0.78 |

$$\Delta P = \left(\frac{Volume\ Flow\ Rate}{Area}\right) \times viscosity \times media\ thickness \times resistivity,$$

$$resistivity = \left(\frac{80}{D^2}\right)\frac{(1 - porosity)^2}{porosity^2}, D = fiber\ diameter$$

The SE coalescer arrived at SRNL in August 2015. The coalescer characterization activities at SRNL began in early September 2015. Given this time frame, it is clear that the lag times between the sample collection dates and sample analysis dates were on the order of 6 months for the SE coalescer sample. The lag times provide an indication of the extent that aging could possibly have caused potential crystallization and /or decomposition of compounds.

## 4.0 Experimental Procedure

### 4.1 Sample Preparation and Laboratory Analyses

Upon arrival at SRNL (August 2015), the coalescer was placed in the Shielded Cells, where it was visually inspected for “gross” flaws and /or defects. No such abnormalities were found (See Fig. 2). The coalescer was then prepared for leaching and chemical analysis. Six one-inch wide “ring” segments of the coalescer were removed (cut). Three “rings” were cut from the closed-end of the coalescer and the other three from the middle. A picture of a cut ring from the SE April 2015 coalescer is shown in Figure 2. A closer look at one of the coalescer pieces is shown in Fig 2. The rings inner surfaces appeared clean. A set of two “ring” segments (one from the closed-end and one from the middle) were submitted to the laboratory “as is,” for solids characterization by XRD, FTIR spectroscopy, SEM, and EDS. A second set of two “rings” were placed in its own “leaching bottle” and immersed in 400 mL of 3 M nitric acid at ambient temperature and under quiescent conditions. The remaining set of two “ring” segments was placed in 400 mL of deionized and double-distilled (DI-DD) water. The weights of each coalescer piece leached in water and nitric acid are listed in Table 2. Leachate aliquots (about 6 mL each) were removed from each bottle (both the nitric acid and DI-DD water) after periods of 1, 7, 14, and 28 days. The respective leachate aliquots from the nitric acid and DI water were submitted to the laboratory for elemental analysis by ICP-AES and for anions by IC.

**Table 3.** Gravimetric weights of the cut samples used in the nitric acid and water leaching tests

| Coalescer and cut piece location | Donut weight (g) Nitric acid leaching | Donut weight (g) Water leaching |
|---|---|---|
| SE middle piece | 12.901 | 12.118 |
| SE closed-end piece | 10.560 | 9.280 |

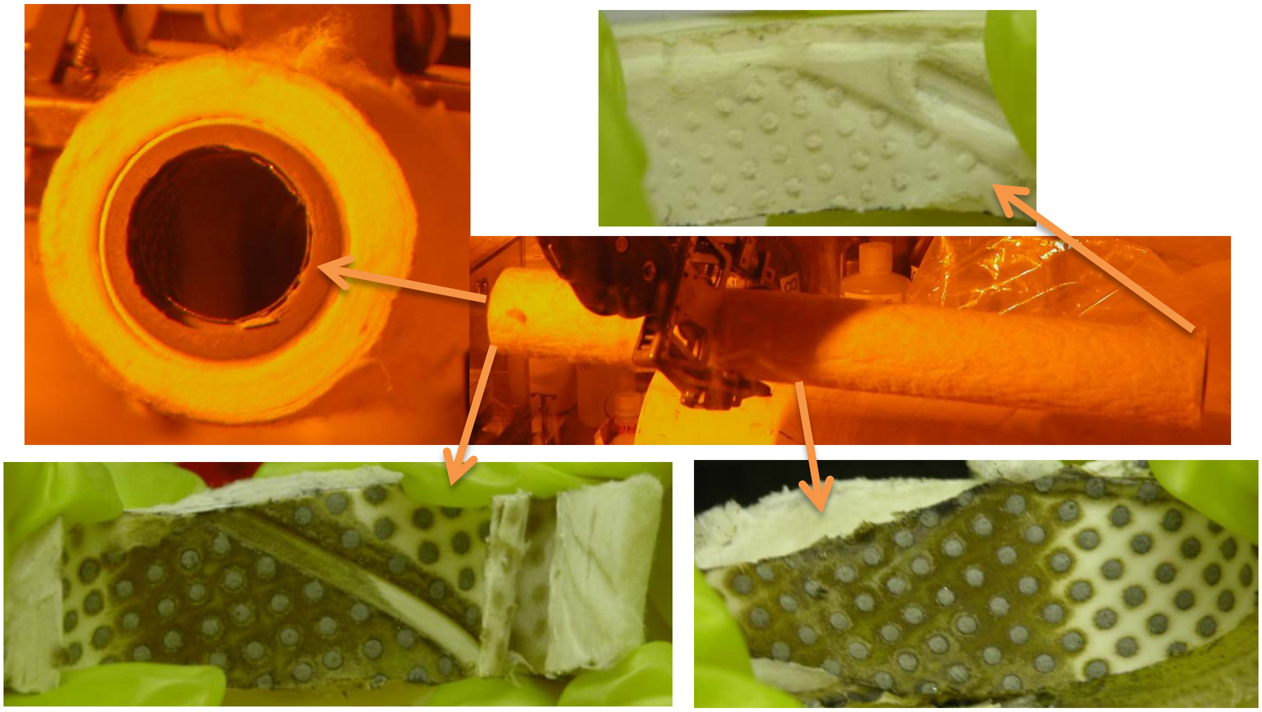


**Figure 2.** A picture of the April 2015 SE coalescer sent to SRNL.

## 5.0 Results and Discussion

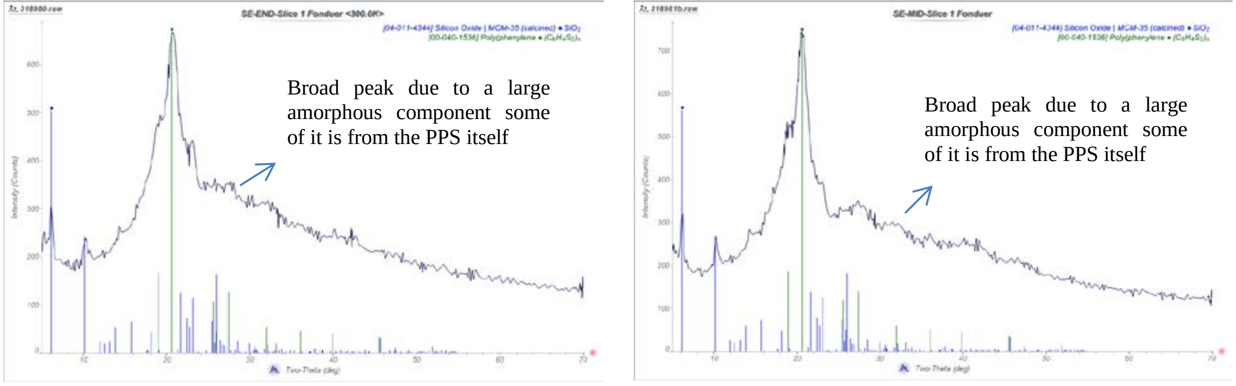
### 5.1 Characterization: XRD

A low concentration of crystalline $SiO_2$ (glass) was detected by XRD in this coalescer (see Fig. 4). It is likely that a silicon-saturated solution carry-over salt solution (from centrifuge 501 that is heated to 33 °C) precipitated past the SE coalescer filter (ACC-309) at MCU. In a low dielectric media, such as the MCU solvent, silica (as well as other elements capable of forming oxygen polyhedral) readily precipitates as a crystalline oxide. Silica has been previously observed in the DSS pre-filter element (coalescer), used for filtering the saltbatches of salt solution from Tank 49H, and in the extraction contactors (SEP-401).[16] The few peaks in the XRD spectrum do not assure the absence of solids – it merely indicates that solid-phase constituents were not present in high concentrations and /or that existing solids were non-crystalline. In addition, the flushing of the SE coalescer with 10 mM boric acid to reduce its radioactivity may have physically dislodged and removed solids from the coalescer. There is also the effect of the deposit orientation in the coalescer that can minimize its interaction with the X-rays in the XRD analysis. The pore network in the coalescer is radially directional and the deposited solids in this network may appear unidirectional to the X-ray beam from the XRD. Thereby, XRD peaks may not be proportional to the solid concentration (bias low relative to solid concentration) on the coalescer. The broad peak observed in Fig. 3 is due to the amorphous Polyphenylene sulfide (PPS) fibers.

Non-detection of crystalline solid-phase constituents does not necessarily mean that these constituents were absent from the samples, but it indicates that such constituents did not dominate the solids and/or were not present in crystalline form. Also, given the likely spatial variations associated with solids precipitating in the coalescer media, there is the understanding that concentrations of deposited solids in one segment of a given sample can be markedly different from those in another segment.

Typically aluminosilicates or pure aluminum hydroxide compounds have been observed in previous SE coalescers.[4,8] In a low dielectric media, such as the MCU solvent, silica (as well as other elements capable of forming oxygen polyhedral) readily precipitates as a crystalline oxide. As discussed above, the few peaks in the XRD spectrum do not assure the absence of solids – it merely indicates that solid-phase constituents were not present in high concentrations and /or that existing solids were non-crystalline. Moreover, the pore network in the coalescer is radially directional and the deposited solids in this network may appear unidirectional to the X-ray beam from the XRD. Thereby, XRD peaks may not be proportional to the solid concentration (bias low relative to solid concentration) on the coalescer.

Given the relatively long storage period of the SE coalescer (~ 6 months), it is surprising that no bicarbonate was observed by XRD given the continuous absorption of carbon dioxide from the air into the (neutral pH) residual solution of the coalescer during storage.


April 2015 Closed-end SE Coalescer

April 2015 Mid-point SE Coalescer

Broad peak due to a large amorphous component some of it is from the PPS itself

Broad peak due to a large amorphous component some of it is from the PPS itself

**Figure 3. XRD spectra of the mid and end-section of the SE (April 2015) coalescer**

### 5.2 Characterization: FTIR Spectroscopy

Given the low crystallinity extent of the solids deposited on the coalescer, further analyses that are insensitive to the amorphous state of the deposits were conducted. These analyses included spectroscopy (FTIR and FT-HNMR) and chromatography (water/acid leaching coupled to ICP-ES and IC-Anions). Starting with the FTIR analysis of the closed-end of the SE coalescer, a small concentration of silicate particles, some with modifier and amines-containing material (amino and secondary amide) on it, was observed (see Fig. 4). The silicate is consistent with previous observations of Sodium Alumino Silicate (NAS) and compounds of aluminum in past SE coalescers characterizations.[4,8] The silicate may have formed from the precipitation of a silica saturated solution during or after contacting the Next Generation Solvent (NGS) . A second mechanism of depositing solids in the coalescer is the passing of small silica particles (or frit) through the filter (ACC-309) and this is considered less likely to happen. The primary amine and secondary amide observed in Fig. 4 from the closed-end of the coalescer are possibly from the decomposition of the TiDG molecules.

$$(\text{R-NH-})_2\text{C=NR} + \text{H}_2\text{O} \Rightarrow \text{R-NH-CO-NH-R} + \text{R-NH}_2 \ (\text{R=aliphatic hydrocarbon})$$

The secondary amide (urea) can also be from proteins (bacteria) but the spectral features (peak intensity at the N-H stretch peak and the peak separation of the amide I and II peak) in Fig. 4 do not fit that of proteins. None of the peaks in Fig. 4 match the peaks from urea (1683, 1631, and 1603 $\text{cm}^{-1}$) or di-urea (1689 and 1604 $\text{cm}^{-1}$) but they are similar to that of polyurea (1647 and 1570 $\text{cm}^{-1}$).[17] Polyurea and di-urea are the main ingredients of HT-51 and GJN lubricants used at MCU. Lubricant leaching or replacement is expected to be minimal. Another chemical detected was oxidized hydrocarbon (R-O-CR-

O). This oil may have come from the oxidation of Isopar™L or it may be the degradation product of the lubricant added to the centrifuge shaft and motors at MCU (HT51or GJN).

The FTIR also detected a high concentration of the MCU modifier. The presence of MCU modifier in the coalescer is reasonably consistent with past observations of the modifier sorbing on different surfaces (as the Isopar™L evaporates during storage), as seen in previous cold simulant testing. At least one modifier addition was made to MCU while this SE coalescer was in service. Soon after the addition, it was discovered that un-mixed or partially-mixed modifier-rich cocktail was circulated through the MCU pipe network and possibly sorbed on surfaces (see A. G. Garrison, SRR-LWE-2015-00022, March 2015). The discontinuity at 32,000 gallons of strip solution in Fig. 1 coincided with the trim addition to the solvent.

The inner most surface of the media that touches the perforated tube had minimal solid loading. The lateral porosity (the available empty space between the fibers when viewing the inner surface in a normal direction or the surface porosity of the coalescer) at the hole entries was found to be closer to 51% (lateral porosity for an "As Received" or un-used coalescer is approximately 68%). A 25% decrease in surface porosity significantly reduces the permeance of the coalescer- thus requiring higher pressure to maintain the same flow rate. The fiber thickness in these media ranged from 18 to 23 microns.

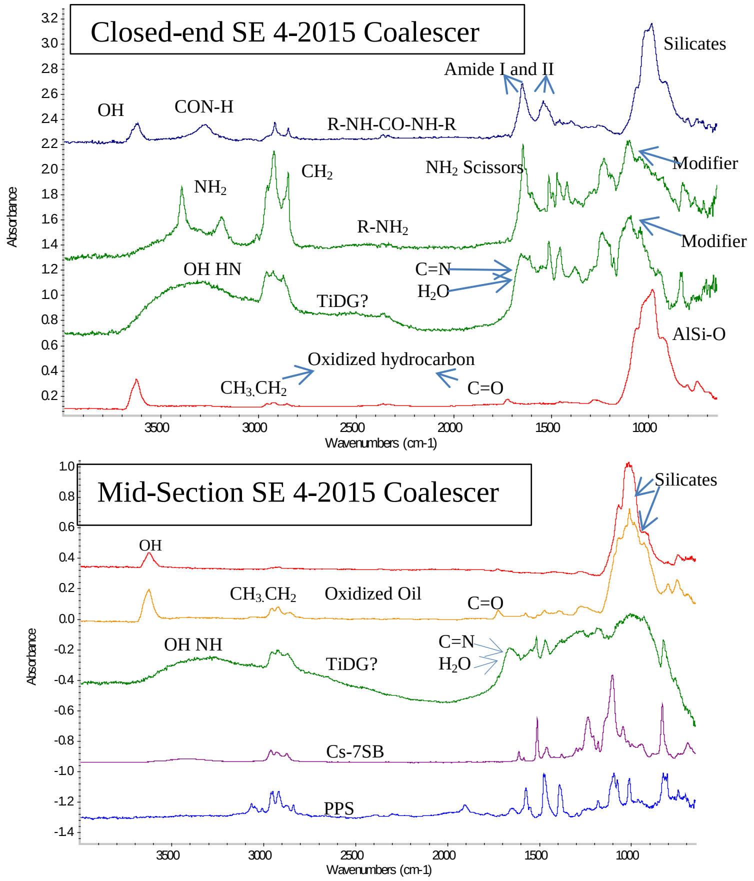
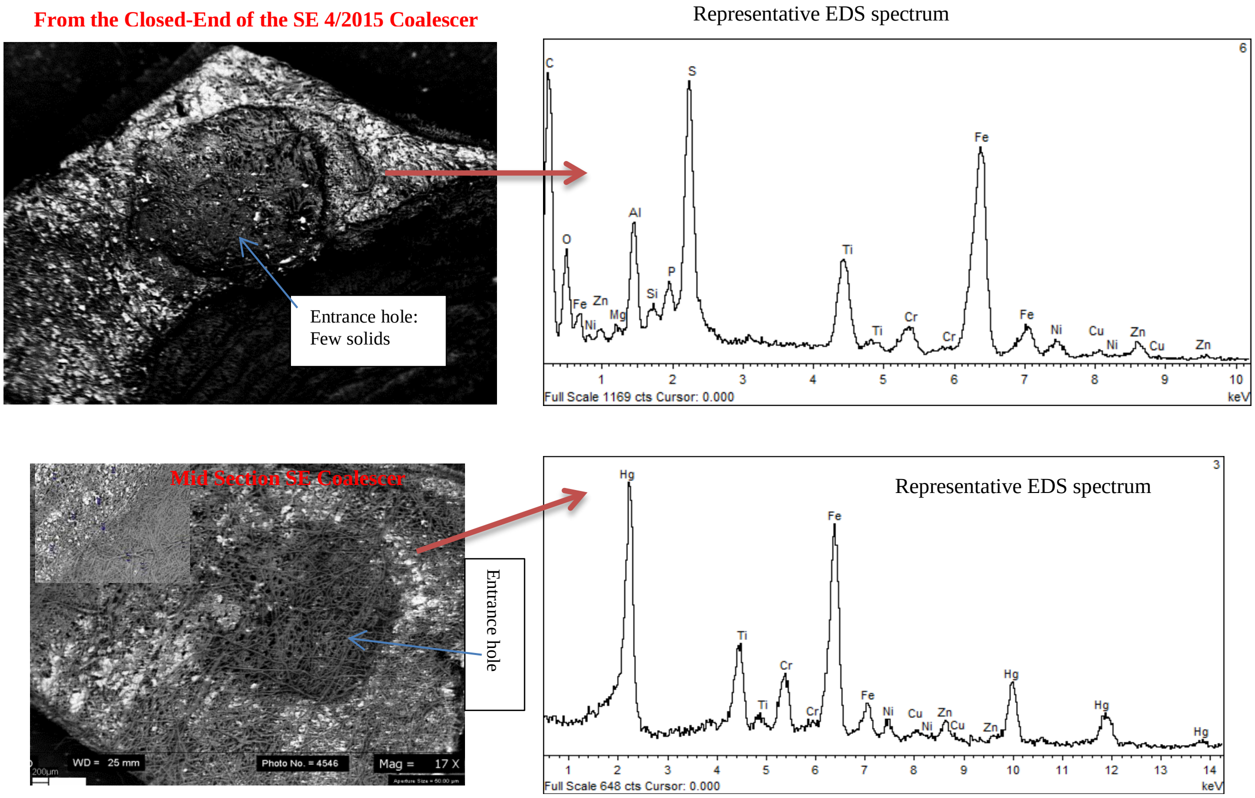


Figure 4. FTIR Results of the middle portion of the SE coalescer April 2015

## 5.3 SEM and EDS

The EDS analyses of both the closed-end and mid-section of the SE coalescer removed in April 2015 (shown in Fig. 5) clearly showed that the large concentration of particles outside of the entranced holes (shown as "dark" circles in Fig. 5) are a mixture of stainless steel debris, aluminum hydroxide, silica, aluminosilicate, and titania particles. The largest fraction of the particles examined was stainless steel (debris from moving parts at MCU). Given that a few of these particles were observed “in” or “on” the entrance discs, they were probably deposited around them when at some point during processing, the coalescer detached itself from its steel mandrel and solution then flowed into those areas depositing those solids that includes mercury. These second deposition of solids may explain the second line (higher slope) observed after processing 36,000 gal of boric acid (see Fig. 1). Phosphorus (phosphate) was also detected and it may be associated with the phosphate content of the salt batch solution or degraded lubricant or from the extractant used in the PUREX process.

In some particles, elements like Potassium and Magnesium were detected. Potassium, a potential competitor to cesium for MaxCalix in NGS, is possibly an impurity from the cold chemical additions to the tanks for corrosion control or from radioactive decay processes in the Tank Farm. Titanium (from the degradation of monosodium titinate [MST]) was also detected as it was detected in the past [Ref. 18]. Mercury, present in the MCU feed, was also observed in Fig. 5. The large concentration of debris on the inner surface of the coalescer may be viewed as a layer of particles blocking the pores of the coalescer. If the suspension concentration is constant and the retention of the particles in the coalescer is constant (for example a constant retention rate of particles), the pressure drop is expected to increase linearly with time which is observed experimentally. If the particles in the suspension simply block the pores at the surface of the coalescer, then the pressure drop should increase non-linearly with time or filtrate volume (this is not observed).


From the Closed-End of the SE 4/2015 Coalescer

Representative EDS spectrum

Mid Section SE Coalescer

Representative EDS spectrum

**Figure 5. SEM and EDS spectra of the closed-end portion from the SE coalescer (April 2015)**

5.4 <u>ICP-AES of Coalescer Leachates</u>

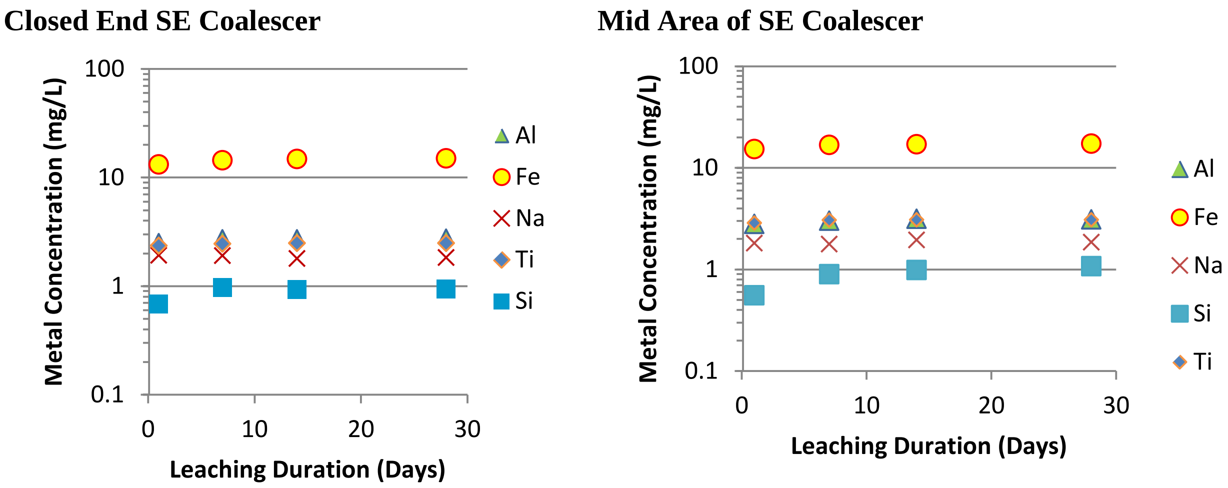
A summary of the metals found in the coalescer leachate solutions from the closed-end and mid-section of the coalescer is shown in Table 3. Twenty eight days of leaching were sufficient for the metals in the leachates to reach a steady state level (see Fig. 6). As clearly seen in Table 3, the elements with significant concentrations in the leachates were Aluminum (fuel cladding), Calcium (trapped chemical and nuclear decay processes), Chromium (steel debris), Iron (steel debris and/or sludge), Molybdenum (steel debris), Nickel (steel debris), Mercury (catalyst for digestion), Sodium, Thorium (from $^{233}$U production at the Savannah River Site in the 1960's), and Vanadium (steel debris). Elements of lesser concentration were Phosphorous and Boron. Phosphate may also originate from the phosphate-based extractant used at the solvent extraction process at the F & H-Canyons. Boron is believed to come from the strip solution (boric acid) used at MCU. No confident conclusions can be made on the Titanium and Lead concentrations due to their high detection limits. The total elements found and their corresponding total molecular mass were consistent between middle and closed end.

A further examination of Table 3 shows that the concentration levels of the elements are higher than those reported in Ref. 7 where the SE coalescer had no visible deposits.

**Table 4. Elemental Constituent Concentrations in SE Coalescer**

| **Element** | **SE April 2015 Leachate Concentration, mg/L (1 sigma 10% from the analytical measurement)** | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | **Closed End** | | | | **Middle Portion** | | | |
| | **t = 1 day** | **t = 7 days** | **t = 14 days** | **t = 28 days** | **t = 1 day** | **t = 7 days** | **t = 14 days** | **t = 28 days** |
| Ag | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 |
| Al | 2.63 | 2.85 | 2.85 | 2.89 | 2.8 | 3.01 | 3.16 | 3.11 |
| B | 0.333 | 0.369 | 0.379 | 0.377 | 0.432 | 0.46 | 0.496 | 0.499 |
| Ba | 0.0945 | 0.105 | 0.107 | 0.107 | 0.107 | 0.125 | 0.127 | 0.128 |
| Be | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 |
| Ca | 1.08 | 1.11 | 1.13 | 1.44 | 1.11 | 1.16 | 1.21 | 1.18 |
| Cd | < 0.08 | < 0.08 | < 0.08 | < 0.08 | < 0.08 | < 0.08 | < 0.08 | < 0.08 |
| Ce | < 0.655 | < 0.655 | < 0.655 | < 0.655 | < 0.655 | < 0.655 | < 0.655 | < 0.655 |
| Co | < 0.063 | < 0.063 | < 0.063 | < 0.063 | < 0.063 | < 0.063 | < 0.063 | < 0.063 |
| Cr | 1 | 1.06 | 1.11 | 1.13 | 1.21 | 1.32 | 1.34 | 1.37 |
| Cu | 0.527 | 0.547 | 0.55 | 0.53 | 0.637 | 0.637 | 0.633 | 0.633 |
| Fe | 13.2 | 14.4 | 14.8 | 15 | 15.3 | 16.8 | 17.1 | 17.3 |
| Gd | < 0.072 | < 0.072 | < 0.072 | < 0.072 | < 0.072 | < 0.072 | < 0.072 | < 0.072 |
| Hg[$] | NM | NM | NM | **33.4** | NM | NM | NM | **41.9** |
| K | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 |
| La | < 0.443 | < 0.443 | < 0.443 | < 0.443 | < 0.443 | < 0.443 | < 0.443 | < 0.443 |
| Li | 0.71 | 0.743 | 0.75 | 0.796 | 0.834 | 0.87 | 0.877 | 0.874 |
| Mg | 0.201 | 0.216 | 0.22 | 0.22 | 0.228 | 0.245 | 0.256 | 0.253 |
| Mn | < 0.177 | < 0.177 | < 0.177 | < 0.177 | < 0.177 | < 0.177 | < 0.177 | < 0.177 |
| Mo | 1.92 | 1.91 | 1.8 | 1.83 | 1.81 | 1.78 | 1.95 | 1.86 |
| Na | 1.04 | 1.15 | 1.12 | 1.11 | 1.31 | 1.27 | 1.35 | 1.37 |
| Ni | 1.11 | 1.17 | 1.51 | 1.66 | 1.24 | 1.78 | 1.92 | 1.93 |
| P | 0.965 | 1.07 | 1.04 | 0.954 | 1.32 | 1.16 | 1.31 | 1.28 |
| Pb | < 83.2 | < 83.2 | < 83.2 | < 83.2 | < 83.2 | < 83.2 | < 83.2 | < 83.2 |
| S | < 1.74 | < 1.74 | < 1.74 | < 1.74 | < 1.74 | < 1.74 | < 1.74 | < 1.74 |
| Sb | 0.684 | 0.971 | 0.929 | 0.942 | 0.558 | 0.897 | 0.985 | 1.08 |
| Si | < 1.1 | < 1.1 | < 1.1 | < 1.1 | < 1.1 | < 1.1 | < 1.1 | < 1.1 |
| Sn | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 | < 0.005 |
| Sr | < 0.313 | < 0.313 | < 0.313 | < 0.313 | < 0.313 | < 0.313 | < 0.313 | < 0.313 |
| Th | 2.36 | 2.46 | 2.48 | 2.49 | 2.87 | 3.06 | 3.08 | 3.09 |
| Ti | < 2.57 | < 2.57 | < 2.57 | < 2.57 | < 2.57 | < 2.57 | < 2.57 | < 2.57 |
| U | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 | < 0.036 |
| V | 1.62 | 1.63 | 1.63 | 1.63 | 2.03 | 2.06 | 2.05 | 2.05 |
| Zn | < 0.021 | < 0.021 | < 0.021 | < 0.021 | < 0.021 | < 0.021 | < 0.021 | < 0.021 |
| Zr | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 | < 0.11 |

[$] Mercury was measured by Cold Vapor Atomic Adsorption (CVAA). NM means "Not Measured".


**Closed End SE Coalescer**

**Mid Area of SE Coalescer**

**Figure 6. Dominant metal concentration from the closed-end and middle portion of the SE coalescer leachates**

Using the data in Table 3, an estimate of the amount of solids deposited on the coalescer was found to be approximately 200 micrograms of solids per mL of coalescer bed media. This calculation assumed, excluding P, Ba, Ca, and Na, all detectable metals listed in Tables 3 are assumed to be non-hydrated oxides. Examples include AlOOH for aluminum, $Fe_2O_3$ for iron (instead of ferric hydride), $SiO_2$ for silicon, $TiO_2$ for titanium, and $ZnO_2$ for zinc. This calculation under estimate the actual solid loading that include nitric acid-resistant austenitic stainless steel (316L) debris observed by the SEM method. A high concentration of mercury was also detected on the coalescer (by the CV-AA method). The mercury levels are much higher than the level of solids needed to plug deep bed filters in other systems (approximately 5 mg/mL of bed). It is believed that both the stainless steel debris and possibly particles (40 to 80 microns in diameter) containing Mercury are associated with the increased in the slope of the pressure drop after 36,000 gallons of filtrated had been processed.

The high concentration of steel debris and Mercury could have plugged the closed-end of the coalescer first and reducing the available coalescer area for permeation which results in higher pressure drop.[14] Since the coalescers are approximately one meter long, the pressure drop along the perforated tube (mandrel) is insufficient to slow the axial flow down the coalescer and the highest discharge flow out of the coalescer (or out of the perforated tube) then occurs at the closed-end of the tube. Thus, if a suspension of solids enters the coalescer, it will flow down the coalescer and exit at the closed-end first. Once the discharge friction at the closed-end of the coalescer increases, the discharge flowing out of the coalescer shifts toward the inlet of the coalescer. In the case of a supersaturated solution, a high liquid flow over the PPS may cause heterogeneous precipitation over these fibers. Longer coalescers or increased axial flow friction (possibly from solid deposition along the wall of the coalescer) may cause significant discharge flow at the middle and inlet region of a coalescer.

In a previous report (Ref. 7), it was recommended to look for oxalates in the leachate of the coalescers. Along this line, IC analysis of the water leachate from the coalescer showed no anion (those that can be observed by IC) was detected (see Table 4).

The steel debris and Mercury on the coalescer are practically insoluble in boric acid and a once-pass through rinsing of the coalescer may not remove sufficient amount of solids to restore the permeation through a bed of nonwoven fibers. Although NAS precipitation is more prevalent on DSS coalescer, it is far easier to trim the salt solution to prevent NAS precipitation and replace the coalescers with new elements rather than clean a plugged coalescer, it is recommended that this practice should be continued.

**Table 5. IC-Anion analysis of the water leachates from the closed-end and mid-point of the SE coalescer**

| IC-Anions | Coalescer Closed End, µg/mL (1σ =10%) | | | | SE Coalescer, µg/mL (1σ =10%) | | | |
|---|---|---|---|---|---|---|---|---|
| Component | 1 day | 7 days | 14 days | 28 days | 1 day | 7 days | 14 days | 28 days |
| Fluoride | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Formate | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Chloride | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Nitrite | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Bromide | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Nitrate | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Phosphate | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Sulfate | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |
| Oxalate | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 |

### 5.5 CP-AES of Coalescer Leachates

Given that a few inorganic solids were found at the entrance discs of the coalescer (see Fig. 5), additional leaching tests were conducted on the coalescer to determine any organic loading. To this end, a portion of the coalescer was leached with dichloromethane (5 to 1 liquid to solid volume ratio) for 4 hours. The leachate was analyzed by the SVOA, H-NMR and FTIR methods.

The SVOA analysis of the leachate is shown in Table 6. Several alkylated ring compounds were detected. Some of these compounds may be degradation products of TBP but they are not likely.[19] A more likely source of these compounds is that they are likely added to the polymer to suppress oxidation and deterioration under UVA-UVB rays. It is believe that these chemicals are not released when the coalescer contacts caustic and/or boric acid solution. These chemicals are released only when they are in contact with a favorable organic solvent. However, it is possible that these chemicals are leached when the coalescer contact the NGS-CSSX solvent. No modifier or extractant was detected by the SVOA method.

The SVOA method is limited to low molecular weight and/or volatile materials. Less volatile material in the leachate was investigated by the HPLC, FTIR and H-NMR methods. No organics above 10 mg/L level was detected by the HPLC method (300319002 and 300319003). The H-NMR spectra of the leachate from the closed-end and mid-section of the coalescer are shown in Figure 7. A large concentration of MaxCalix and modifier was observed in the spectrum of the leachate from the mid-section. A closer look at the modifier's NMR peaks suggests the absence of hydroxyl groups in the modifier molecules which could make the modifier insoluble in aqueous solutions. The FTIR spectrum (Fig. 8) also shows a large aliphatic content in the modifier molecules proving the absence of hydroxyl groups. Both the H-NMR and FTIR indicate a high concentration of modifier (mostly without an OH group) in the entrance discs of the coalescer and this is consistent with the recent addition of modifier that did not properly mix with the solvent and probably flowed around the MCU loop, possibly attaching itself to surfaces.

**Table 6. SVOA analysis of the dichloromethane leachate of the April 2015 SE coalescer. All units are given in mg/L with 20% uncertainty. Limit of detection (method) was 1 mg/L.**

| **Component**<br>**Result** | Mid-section<br>300319003 | Closed-End<br>300319002 |
|---|---|---|
| 2,6 di-t-butylphenol | 480 | 440 |
| 2,4 di-t-butyl-6-nitrophenol | 78 | 74 |
| 4-tert-Butyl phenol | 15 | 14 |
| Triphenyl phosphate | 24 | 21 |
| 2,5-Di-tert-butylphenol | <1 | 3.8 |
| 4,4-Dichlorodiphenylsulfide | 5.9 | 3.5 |
| 4-tert-butyl-2,6-diisopropylphenol | 2.6 | <1 |
| Diisooctyl adipate | 6.5 | <1 |
| All Other SVOA Organics | <5 | <5 |

*"As received" coalescer (unused).

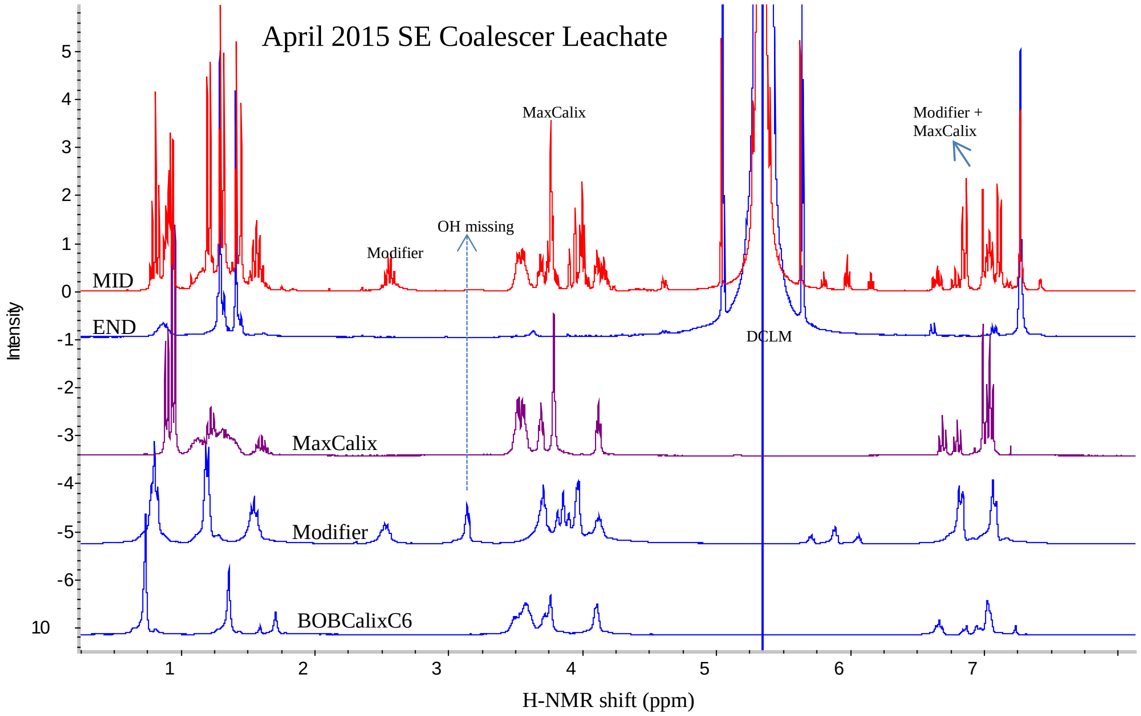


**Figure 7. H-NMR spectra of the dichloromenthane leachate of the closed-end and mid-section of the SE Coalescer.**

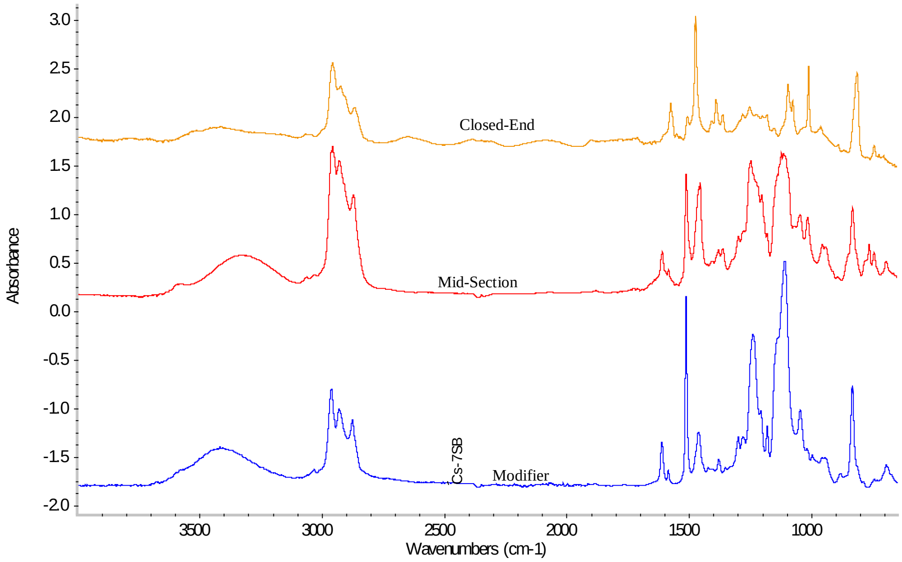


**Figure 8. FTIR analysis of the DCLM leachate from the closed-end and mid-section of the SE coalescer removed in April 2015**

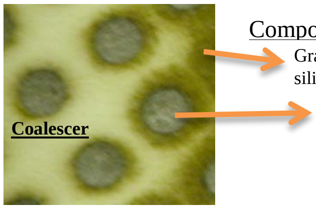
## 6.0 Conclusions

In June 2015, SRNL received a SE coalescer (FLT-304) from MCU. That coalescer was first installed at MCU in late October 2014 and removed in April 2015. While processing approximately 48,700 gallons of strip solution, the pressure drop steadily increased linearly from 1 psi to near 16 psi (the administrative limit is 17 psi) with the total filtrate volume (2.1E-4 psi/gal of filtrate). The linear behavior is due to the combined effect of a constant deposition of material that starts from the closed-end to the mid-section of the coalescer reducing the available surface area of the coalescer for fluid passage (linearly with filtrate volume) and the formation of a secondary emulsion (water in NG-CSSX) on the fibers of the coalescer media. Both effects reduced the coalescer porosity by at least 13% (after processing 48,700 gallons). Before the coalescer was removed, it was flushed with a 10 mM boric acid solution to reduce the dose level. To determine the nature of the deposited material, a physical and chemical analysis of the coalescer was conducted.

Characterization of this coalescer revealed the adsorption of organic containing amines (secondary amides and primary amines), TiDG, degraded modifier (with no hydroxyl group), MaxCalix, and oxidized hydrocarbon (possibly from Isopar™L or from lubricant used at MCU) onto the coalescer media. The amide and amines are possibly from the decomposition of the suppressor (TiDG) as follows:.

$$(R\text{-}NH\text{-})_2C{=}NR + H_2O\ (\text{caustic}) \Rightarrow R\text{-}NH\text{-}CONH\text{-}R + R\text{-}NH_2 \quad (R{=}\text{aliphatic hydrocarbon})$$

The modifier and MaxCalix were the largest components of the deposited organic material, as determined from leaching the coalescer with dichloromethane. Both the FTIR and FT-HNMR results indicated that some of the modifier was degraded (missing their OH groups). The modifier was observed everywhere in the examined coalescer pieces (FTIR) while the TiDG and its decomposition products were observed at the entrance discs of the coalescer. A summary of the deposits found in the coalescer is shown below.



Composition of the deposits found on the coalescer

Gray color: Mercury, steel debris, titanium, aluminum, silicates, modifier, and MaxCalix

Entrance Discs:
Secondary amines, poly-urea, TiDG, modifier, oxidized lubricant and silicates

A solvent trim (a cocktail of solvent components with a high concentration of modifier) was added to the solvent on 2/22/2015. It is believed that the trim did not mix completely with the solvent and that it was subsequently spread around the MCU components including the coalescers, where it may have deposited.

Chronologically, the modifier, the TiDG's decomposition products and silicates deposited on the entrance discs first and after the pressure drop increased significantly, parts of the coalescer media detached itself from the central porous steel mandrel and a significant amount of steel debris, mercury, titanium, and additional aluminum and silicates deposited on the coalescer. This is responsible for staining the coalescer media with a gray color.

## 7.0 Recommendation

The recommendations listed in Ref. 7 will benefit and improve lengthening the services of the SE and DSS coalescers.

## 8.0 References

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## Appendix A. SEM images of the Closed-End from the SE Coalescer removed on April 2015

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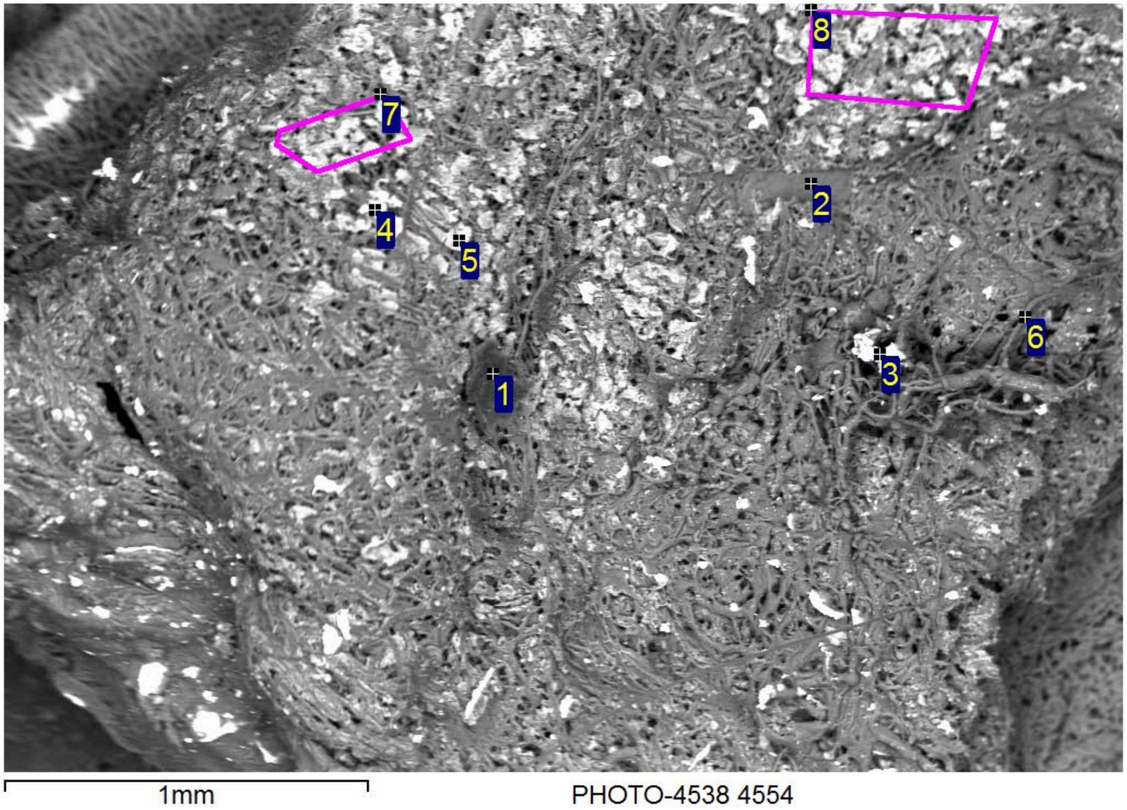

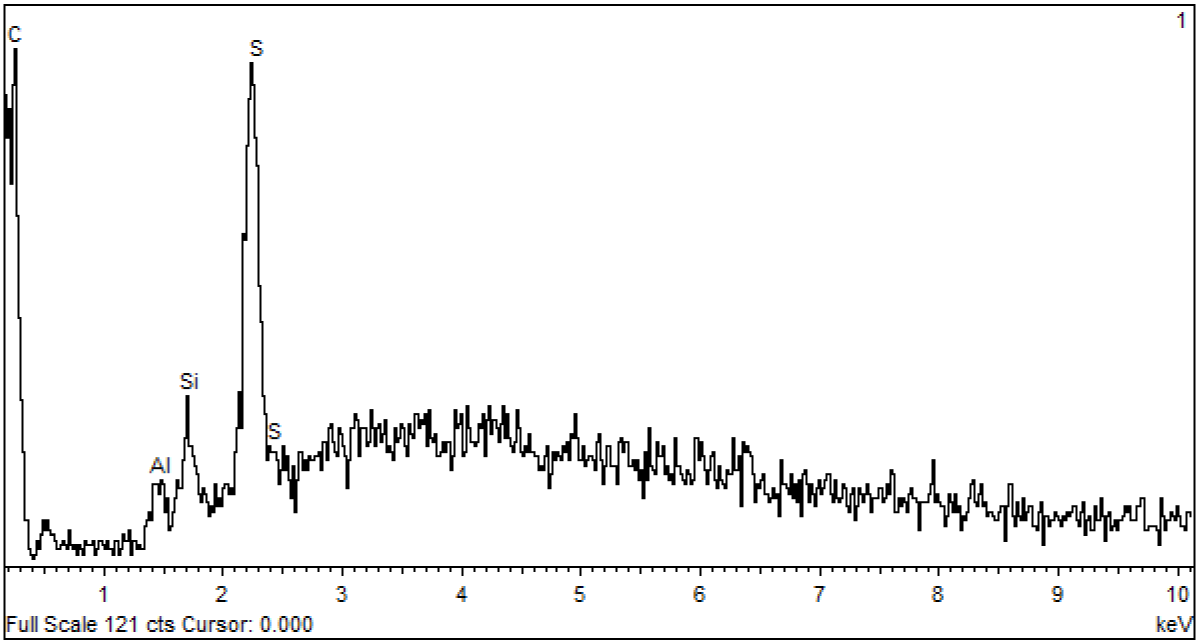

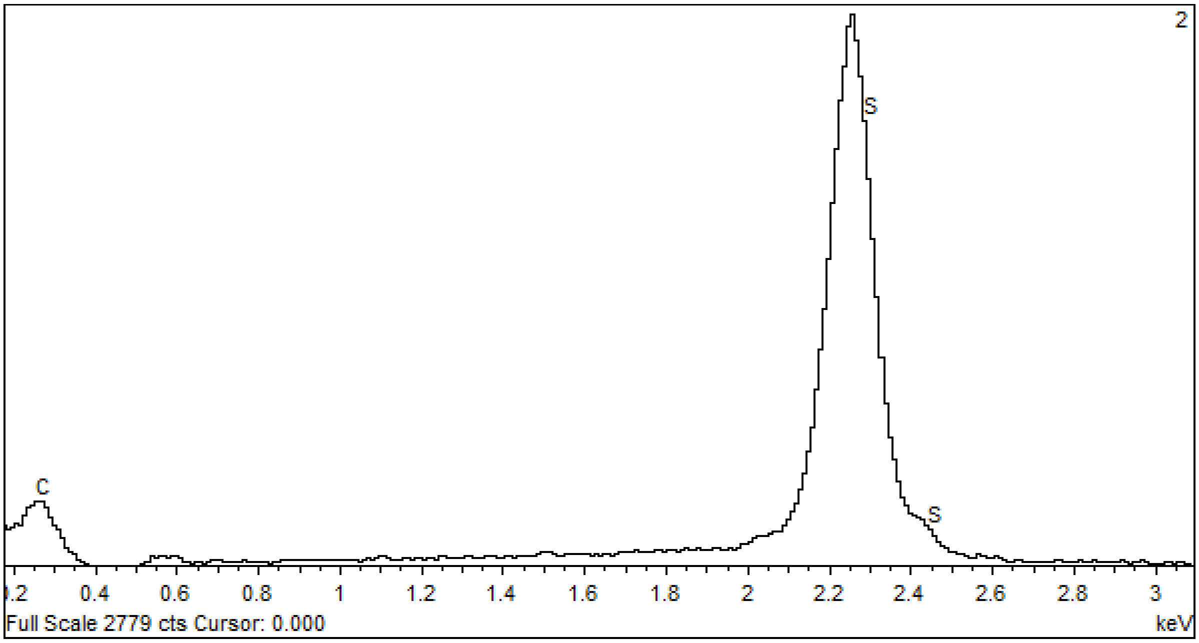
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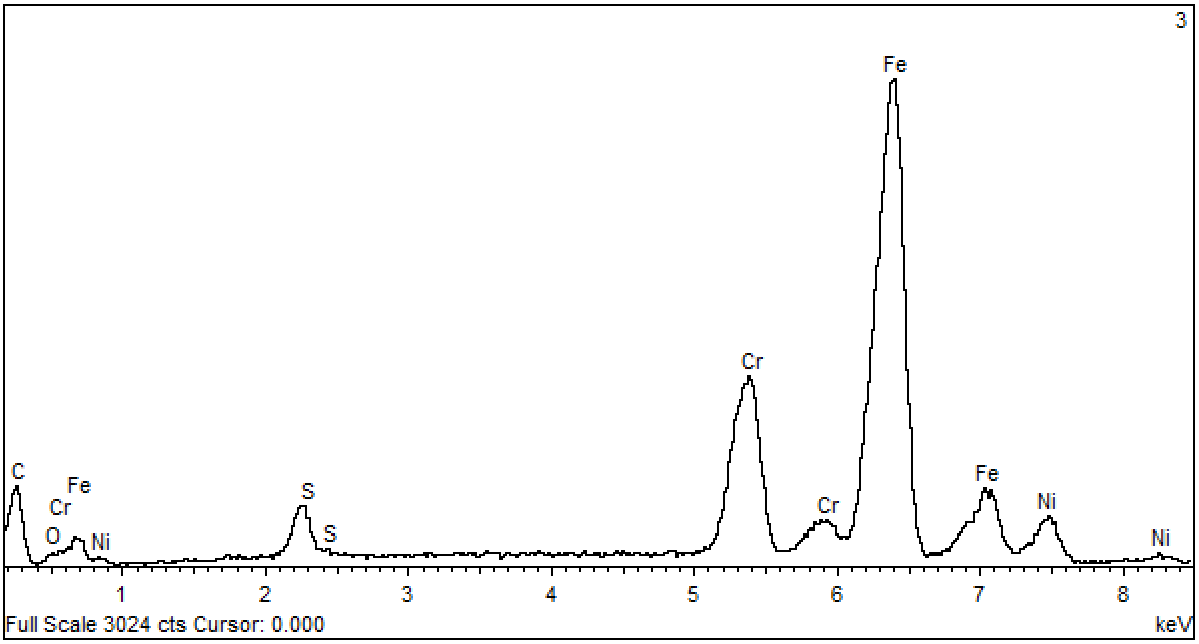
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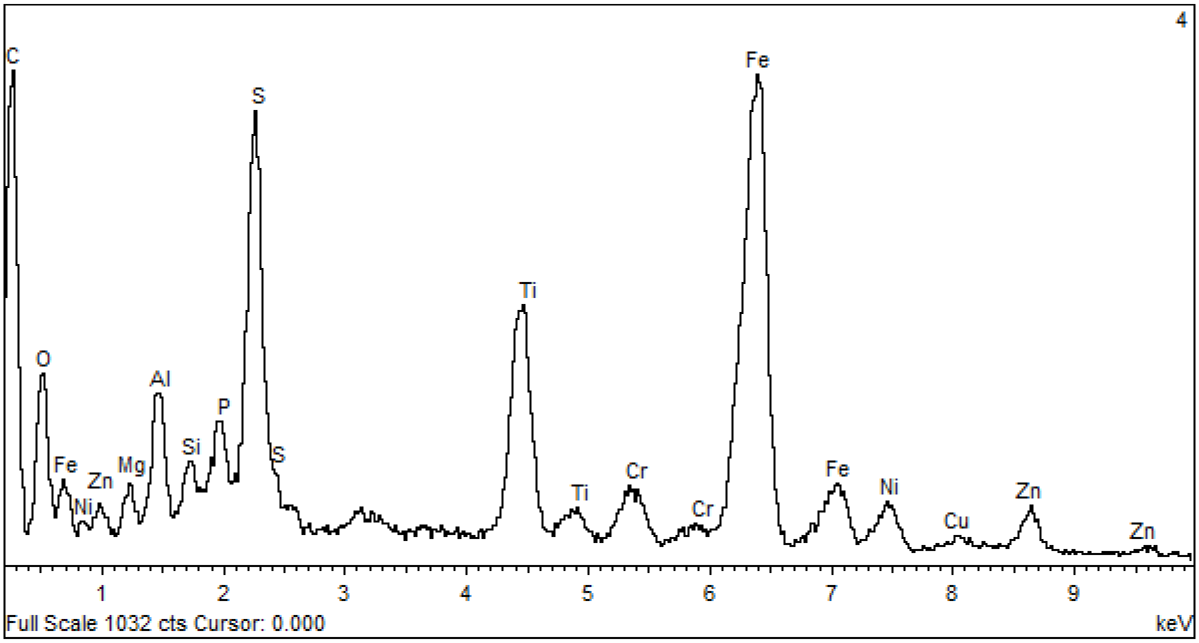
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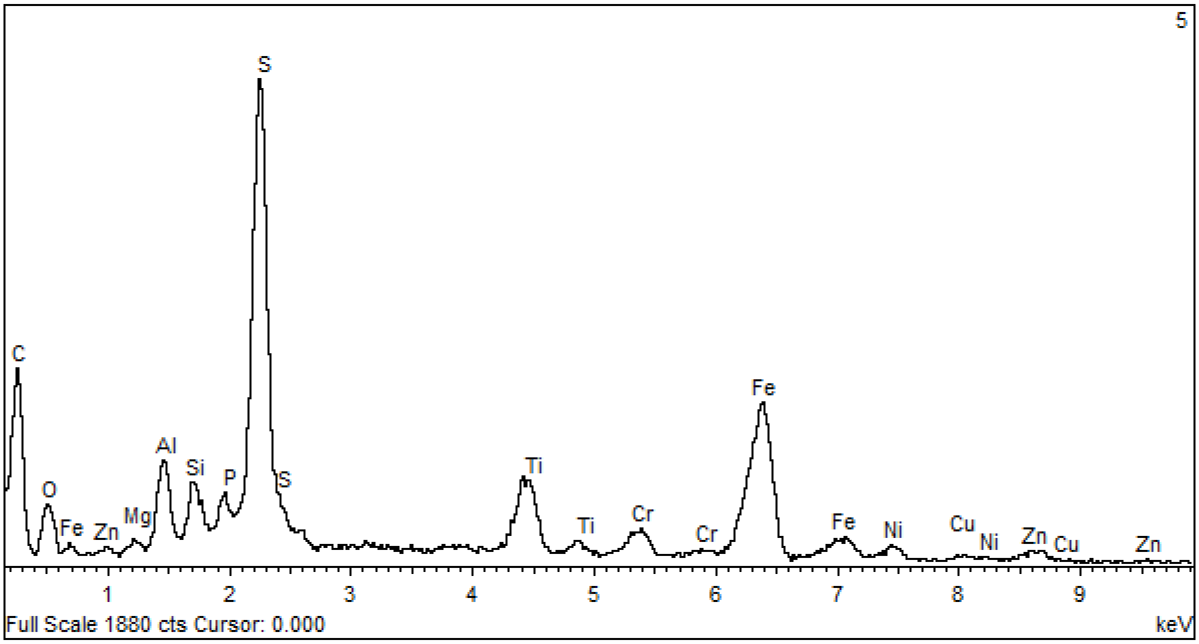
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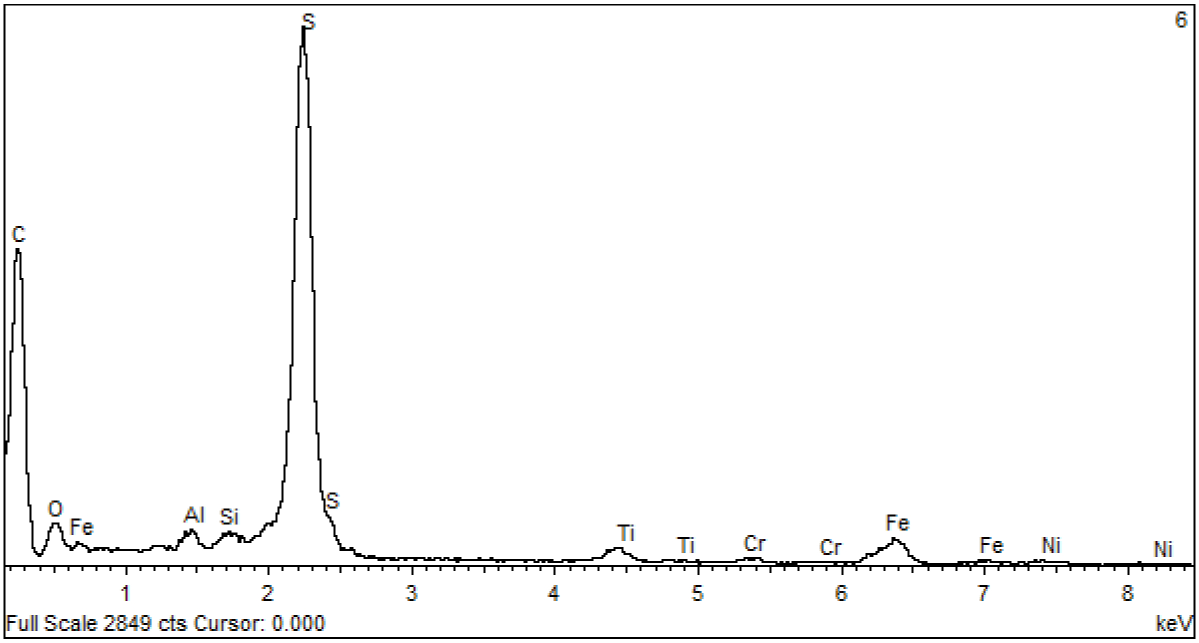
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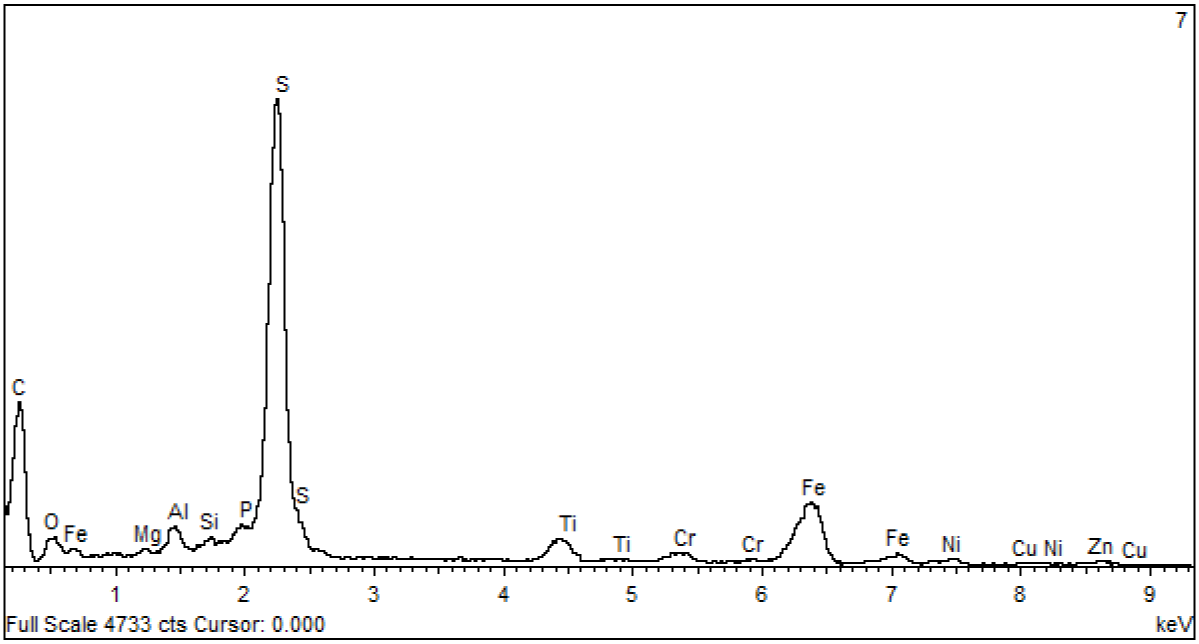
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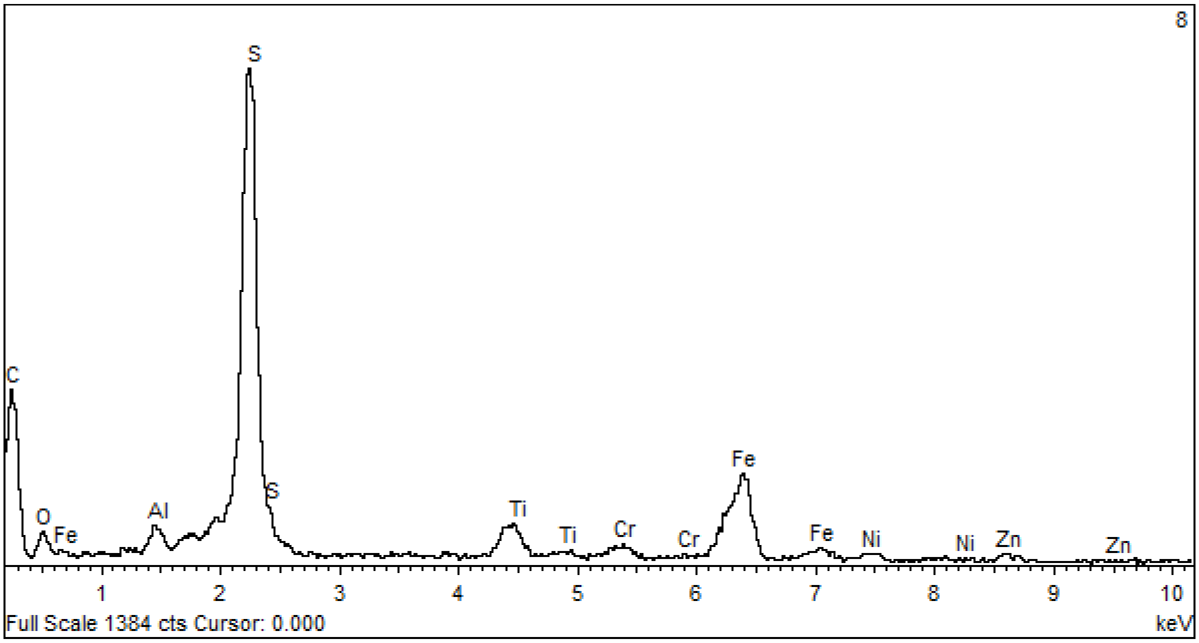
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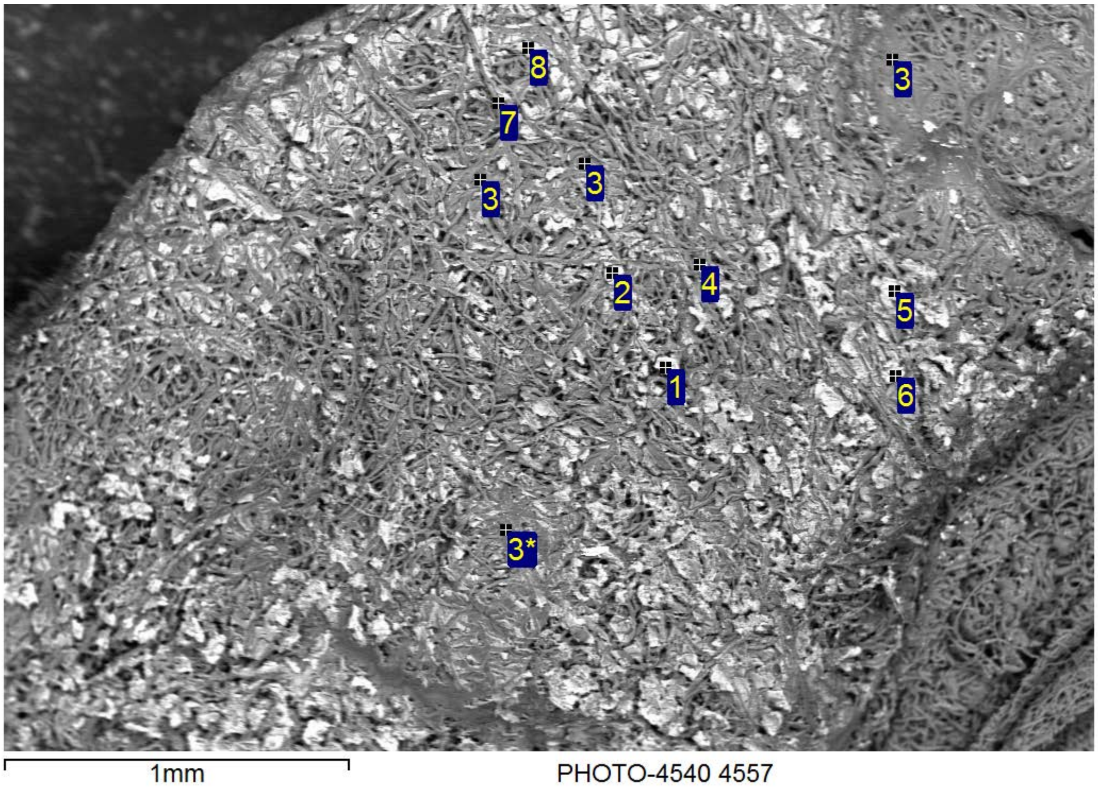
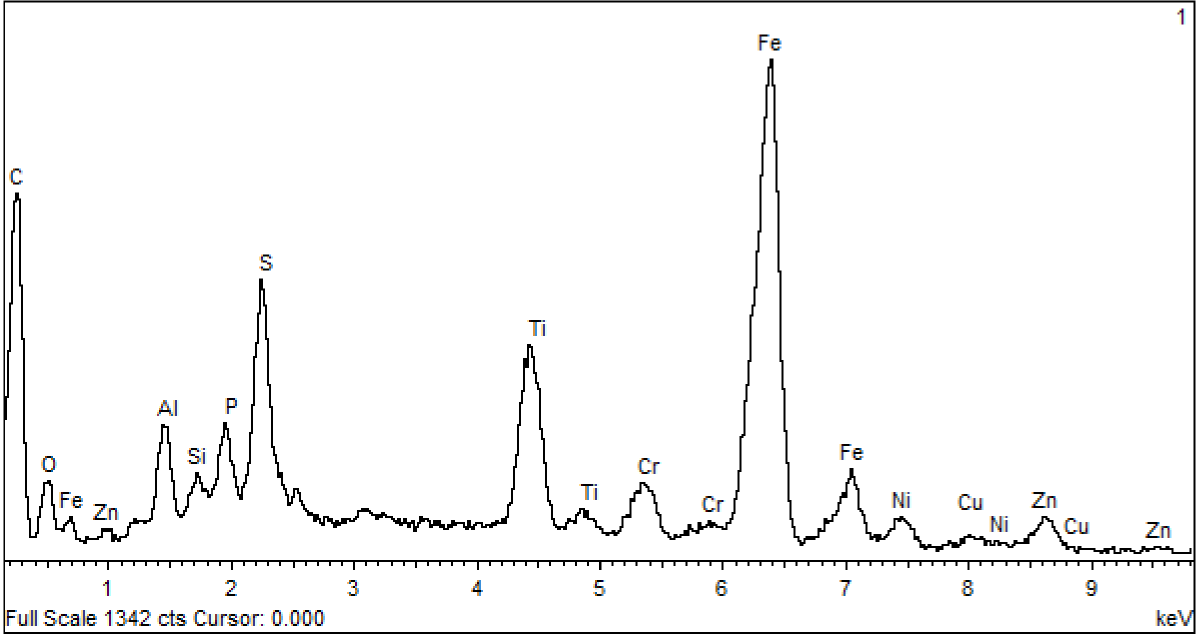
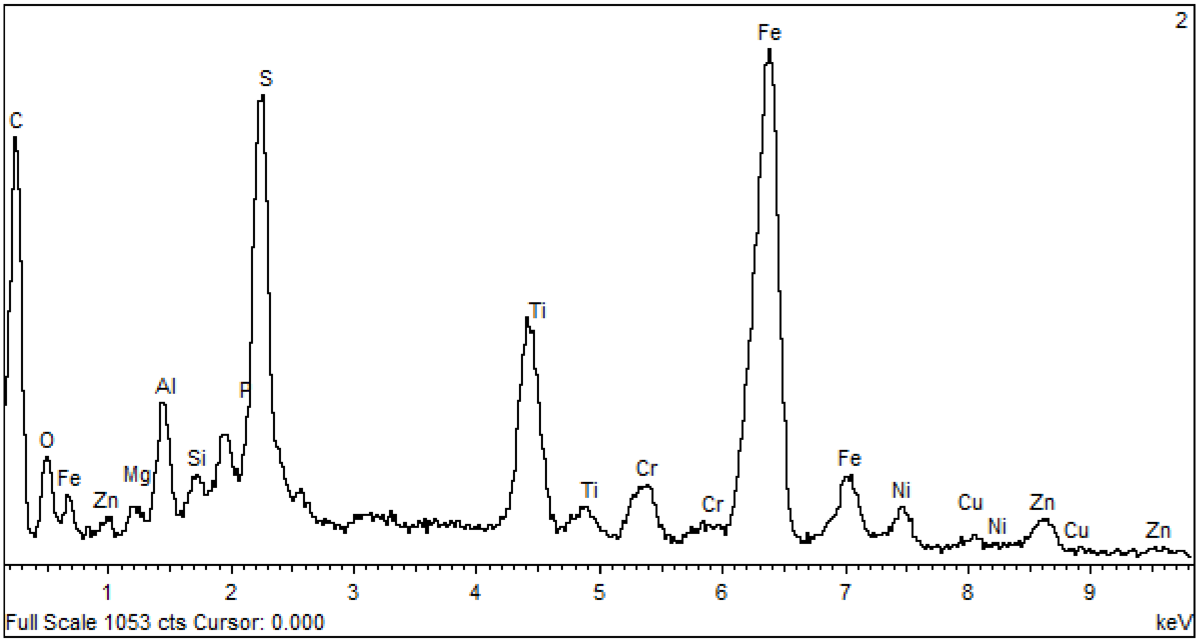


Note: Number with an asterisk (*) indicates the actual data point. Otherwise, they are representative. Representative EDS spectra are similar enough to each other that they are essentially identical with minor variations in peak heights, but not identified elements.

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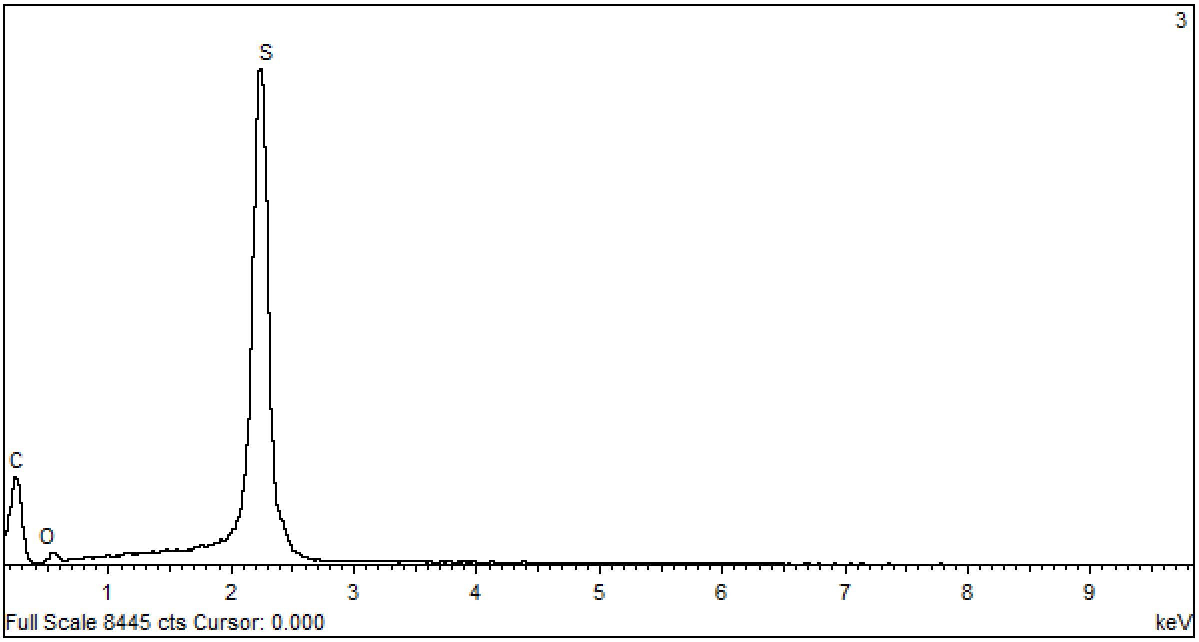
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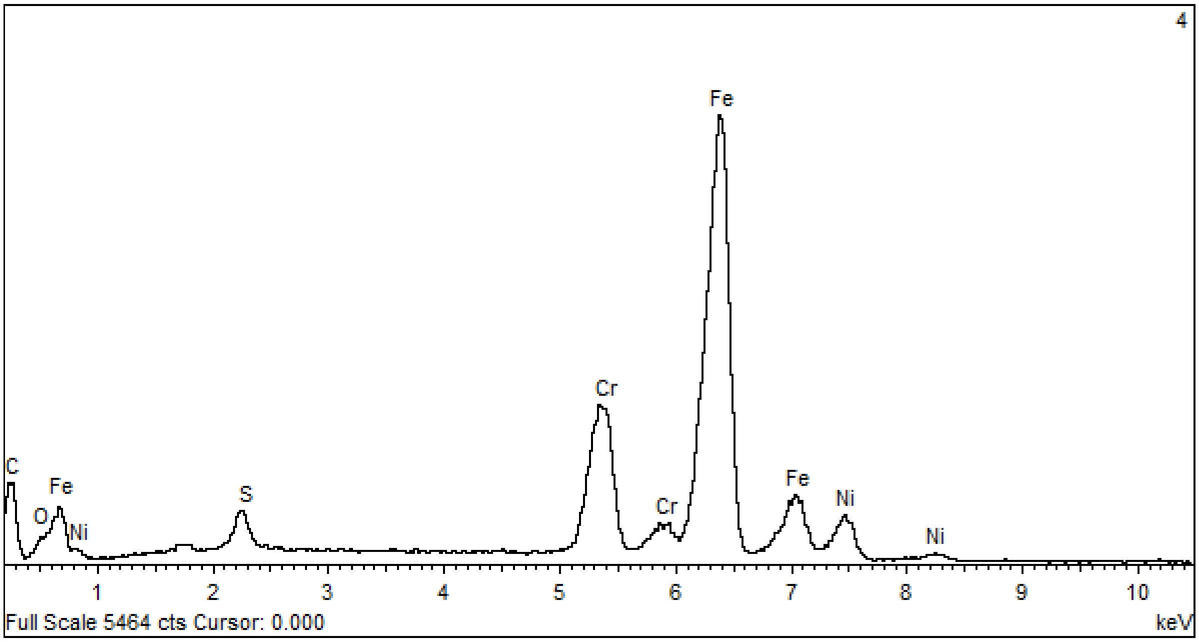
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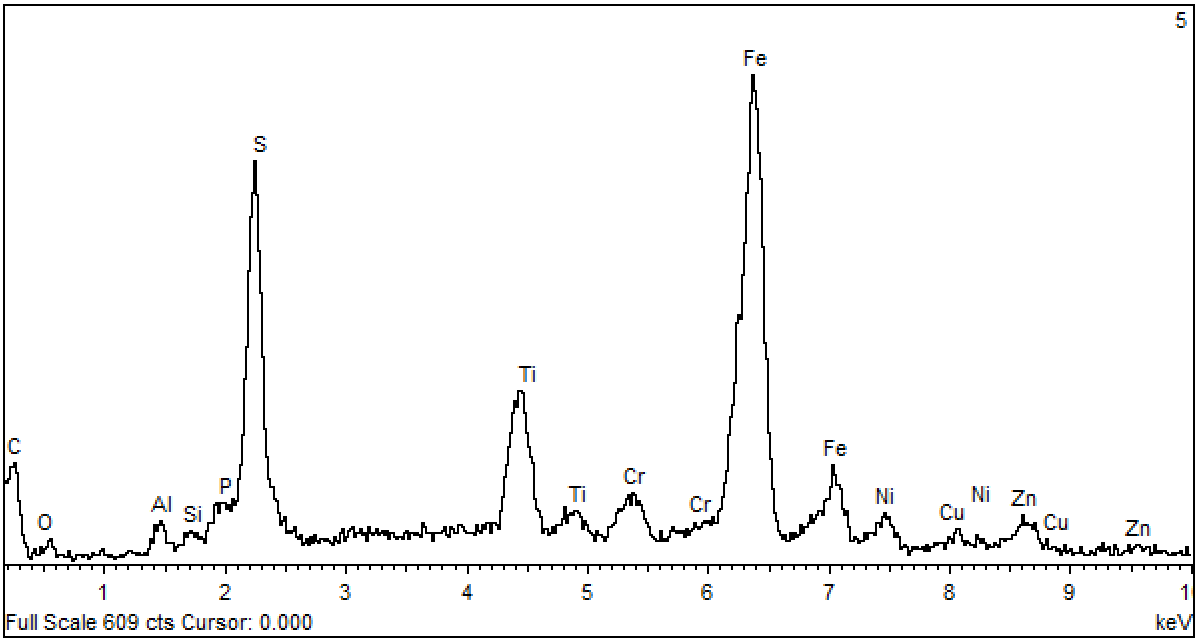
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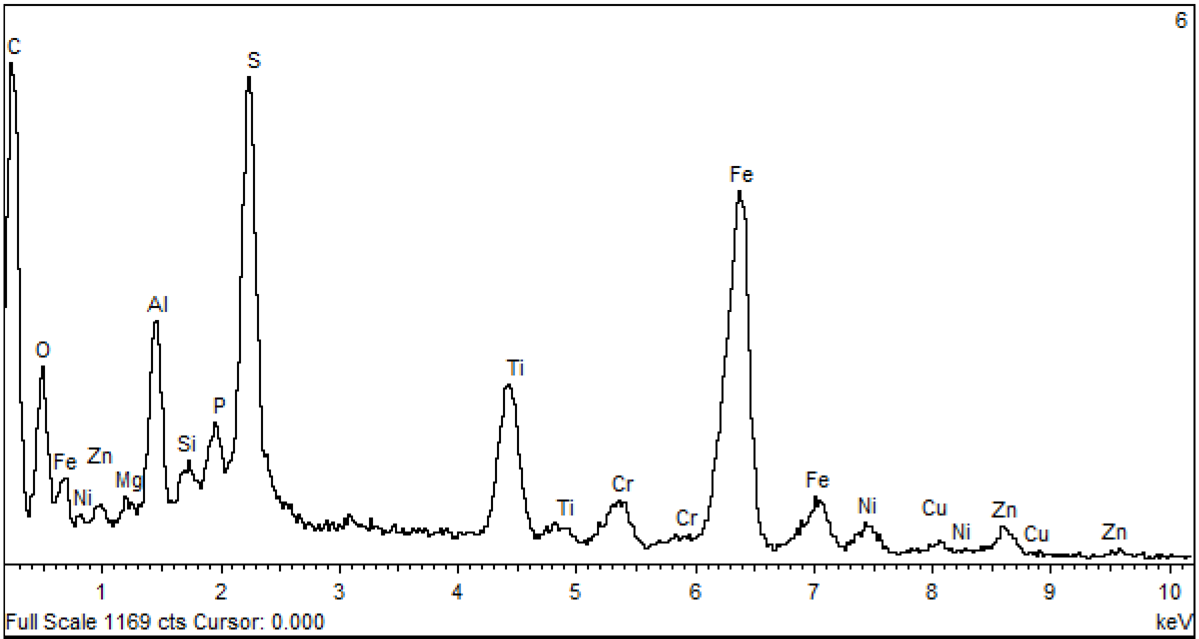
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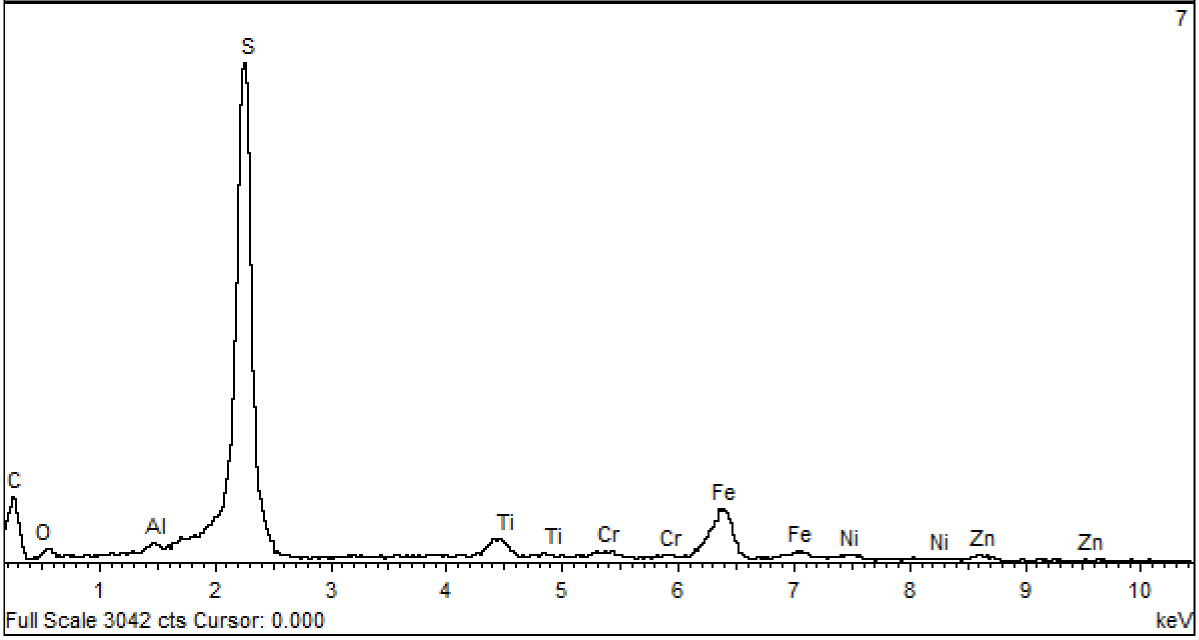
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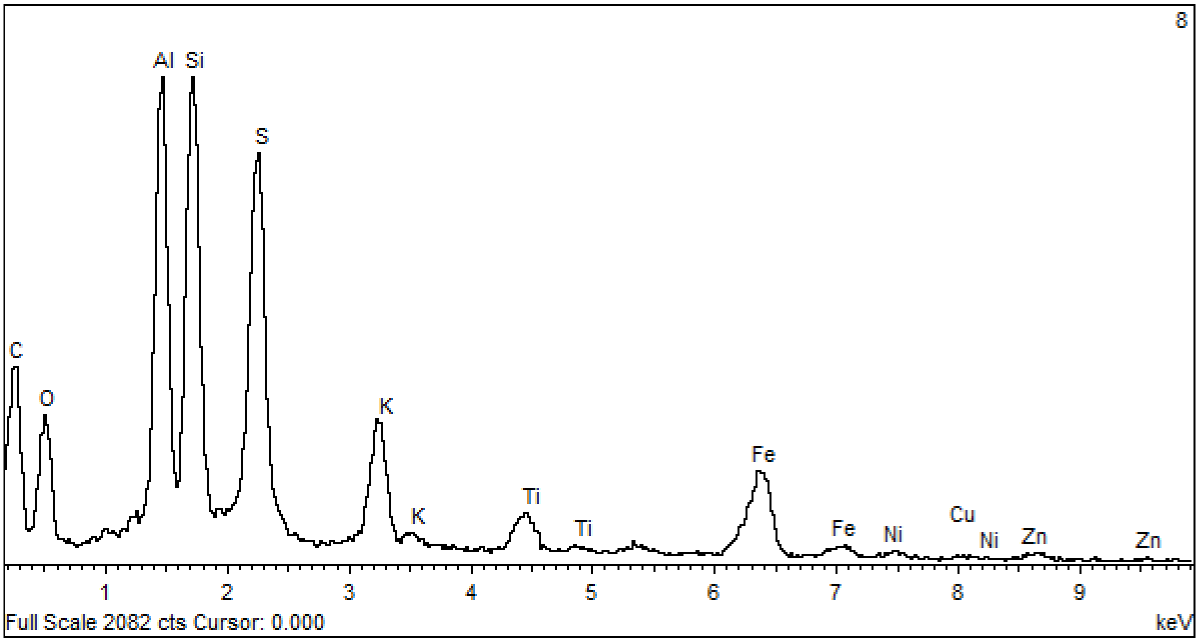
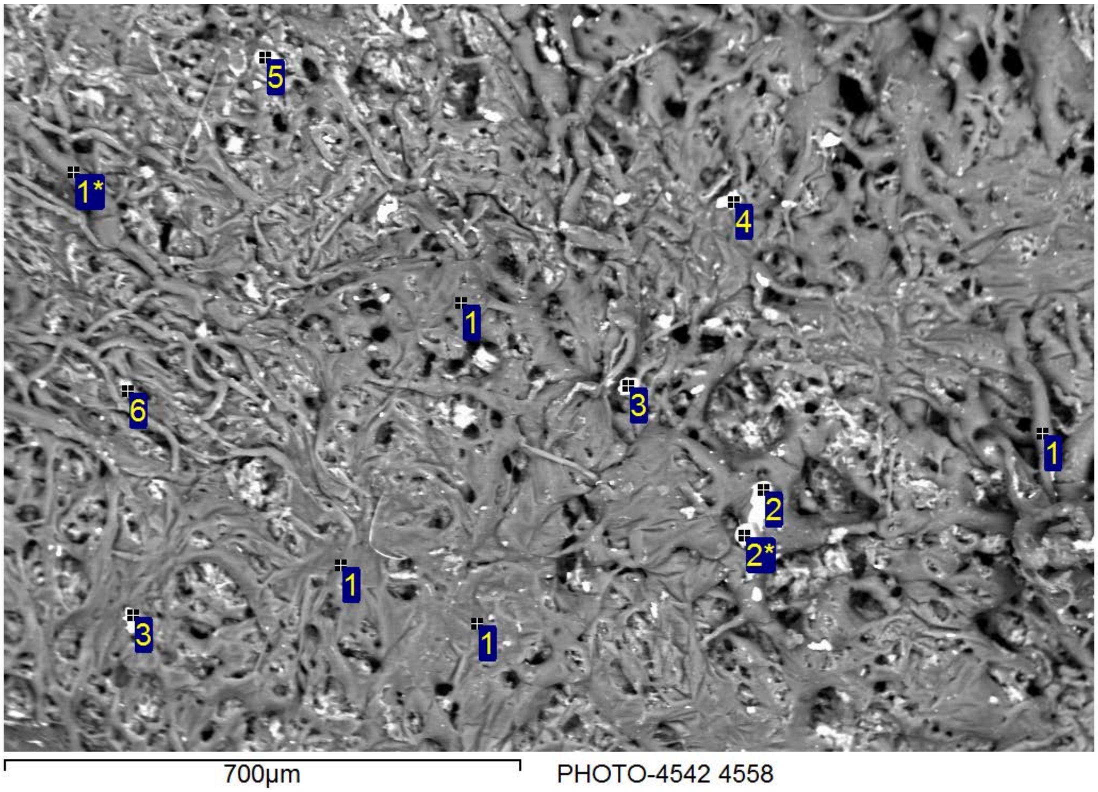
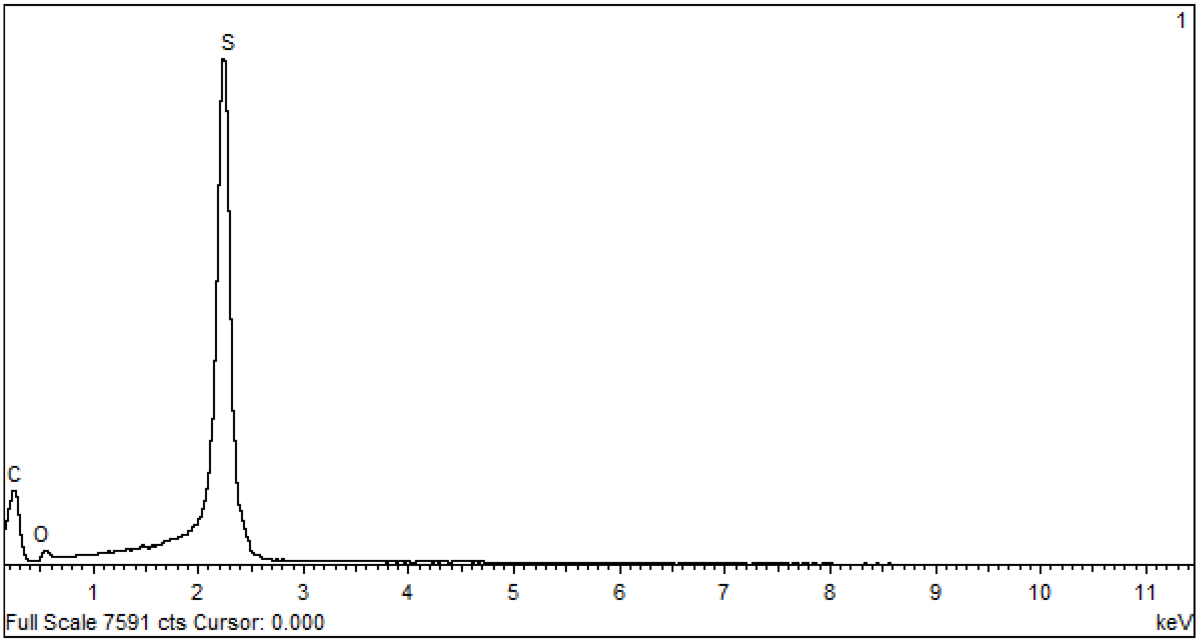
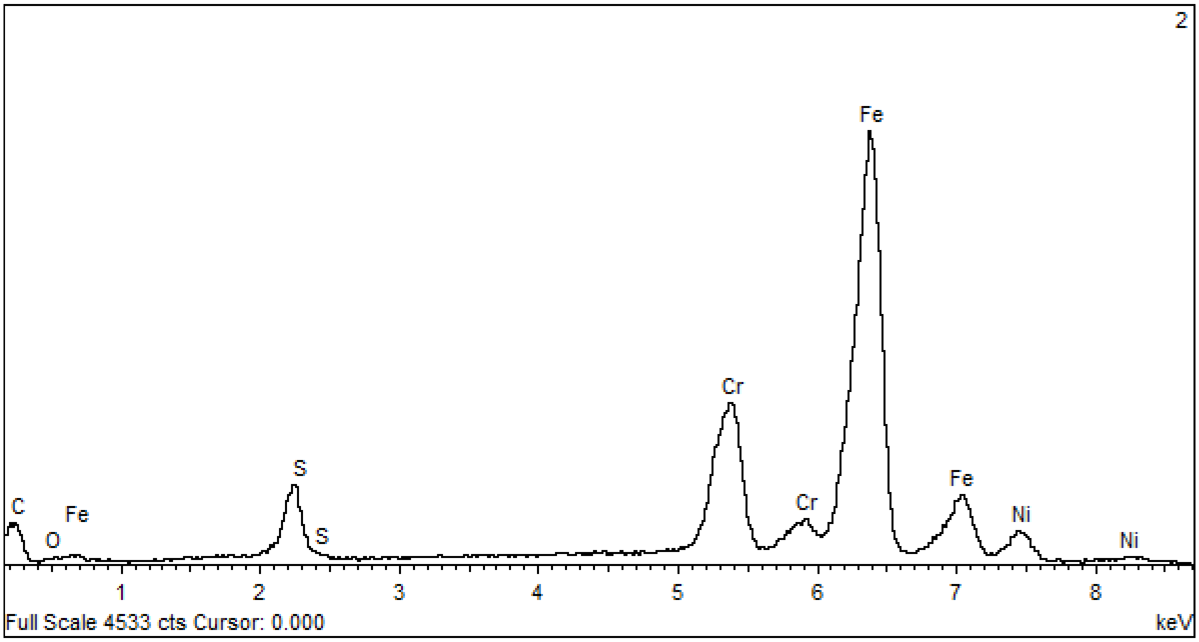
keV

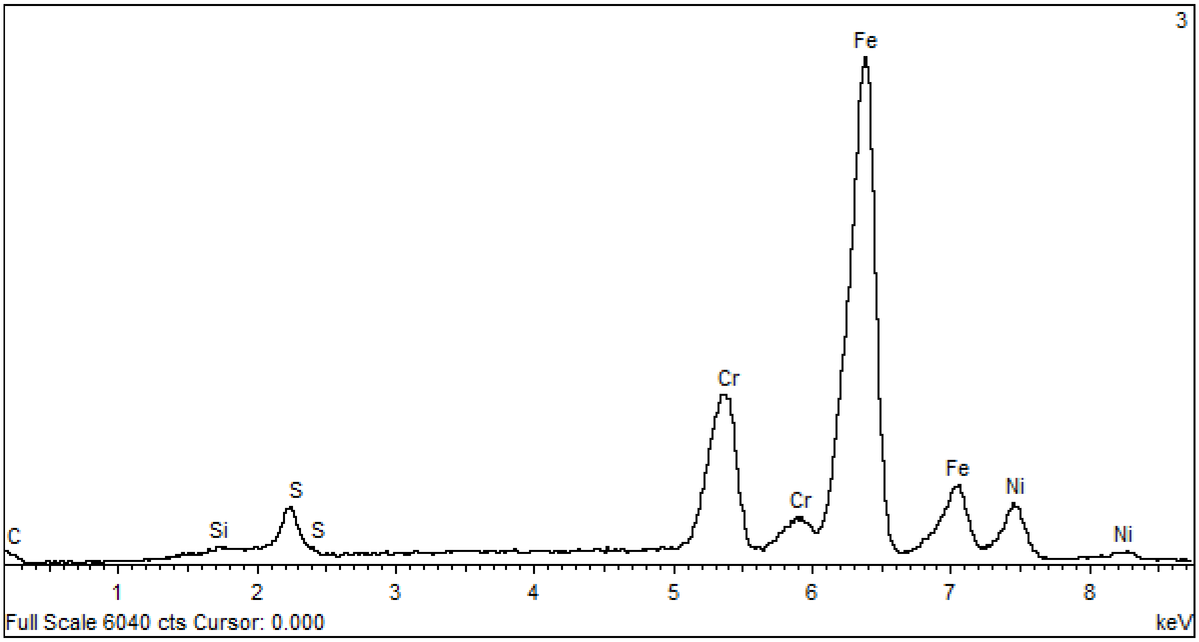


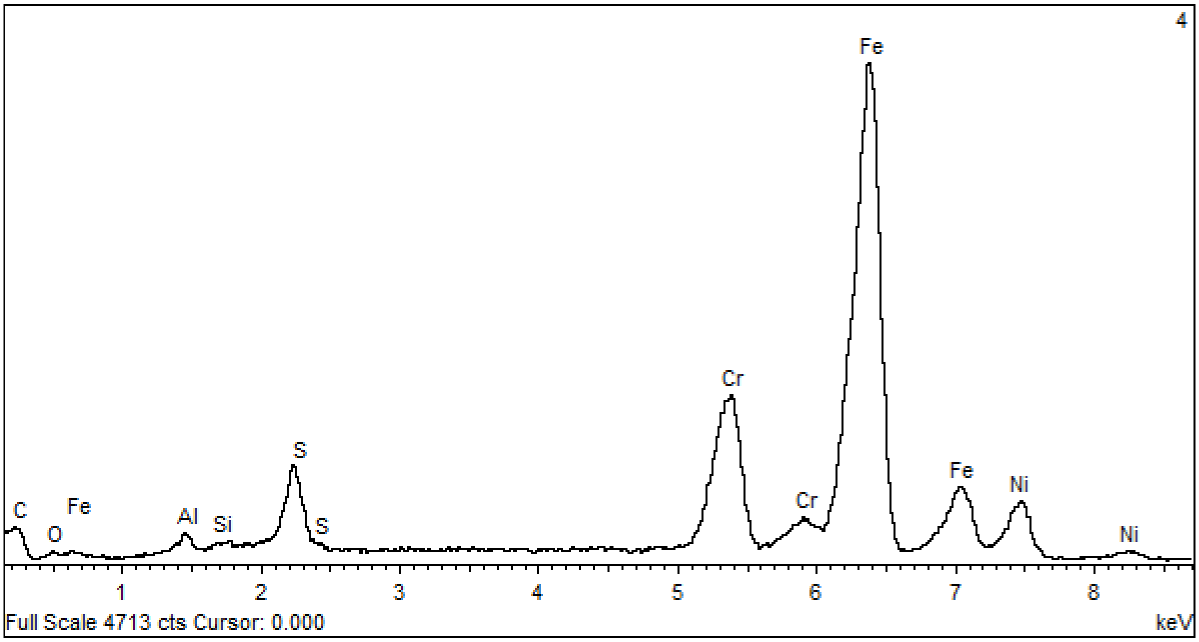


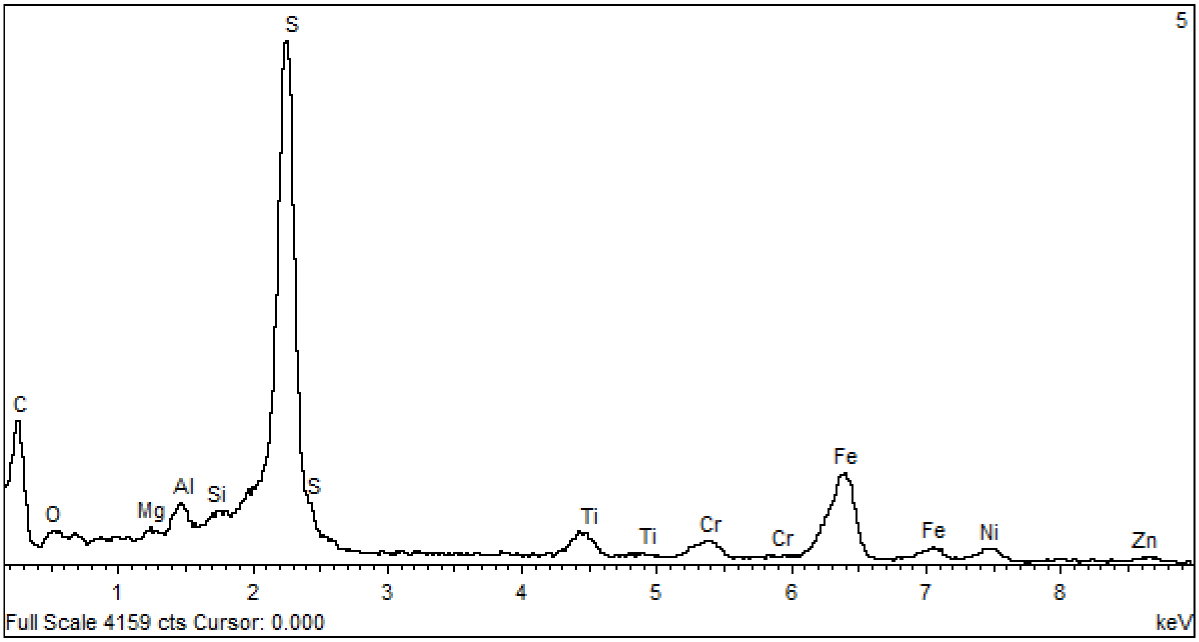
Note: Number with an asterisk (*) indicates the actual data point. Otherwise, they are representative. Representative EDS spectra are similar enough to each other that they are essentially identical with minor variations in peak heights, but not identified elements.

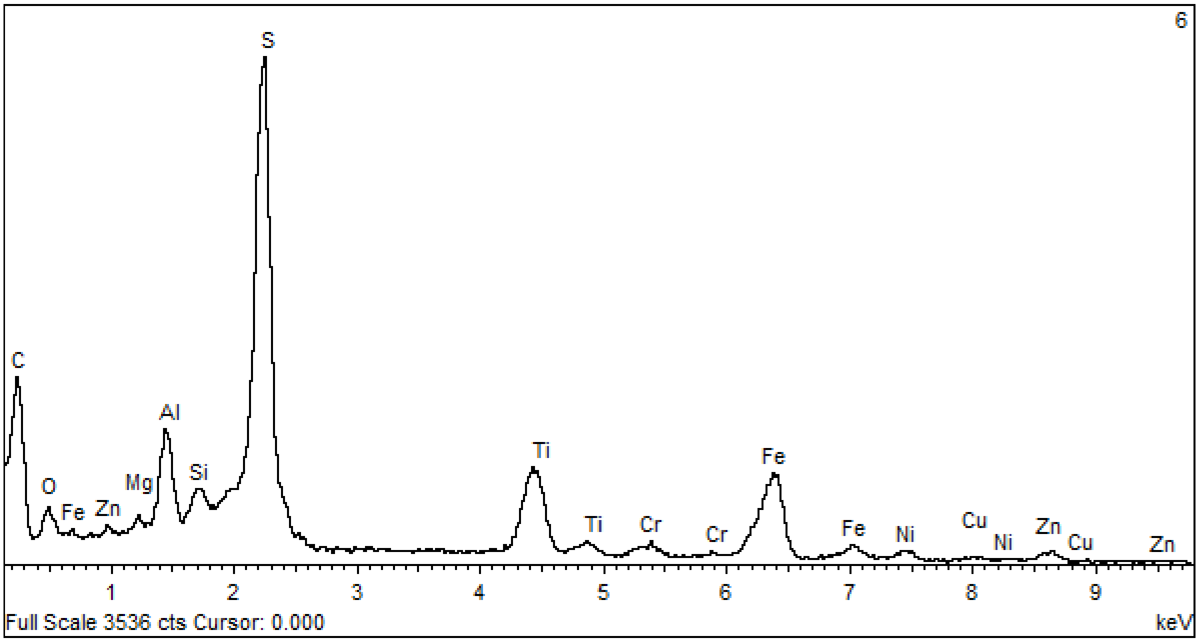

1
S
C
O
1
2
3
4
5
6
7
8
9
10
11
Full Scale 7591 cts Cursor: 0.000
keV


2
Fe
Cr
S
Cr
Fe
Ni
C
Fe
O
S
Ni
1
2
3
4
5
6
7
8
Full Scale 4533 cts Cursor: 0.000
keV

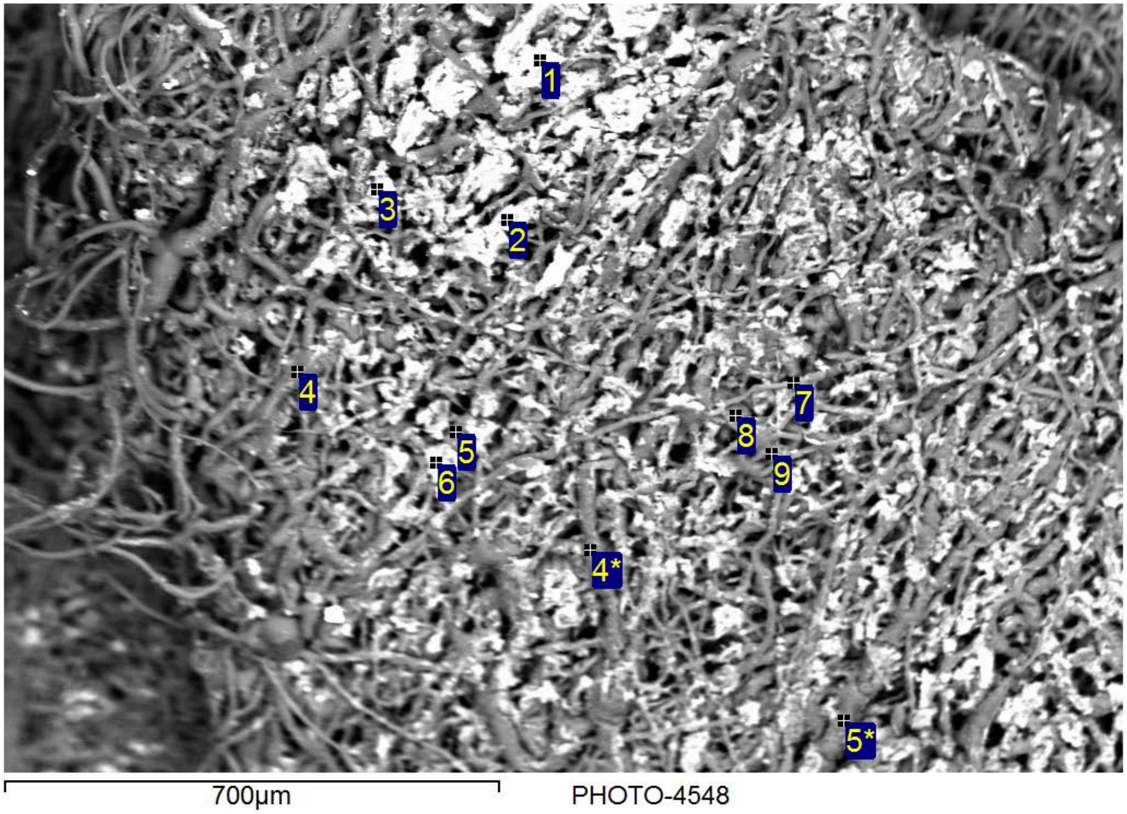

3
Fe
Cr
S
Cr
Fe
Ni
C
Si
S
Ni
1
2
3
4
5
6
7
8
Full Scale 6040 cts Cursor: 0.000
keV


4
Fe
Cr
S
C
Fe
Al
O
Si
S
Cr
Fe
Ni
Ni
1
2
3
4
5
6
7
8
Full Scale 4713 cts Cursor: 0.000
keV

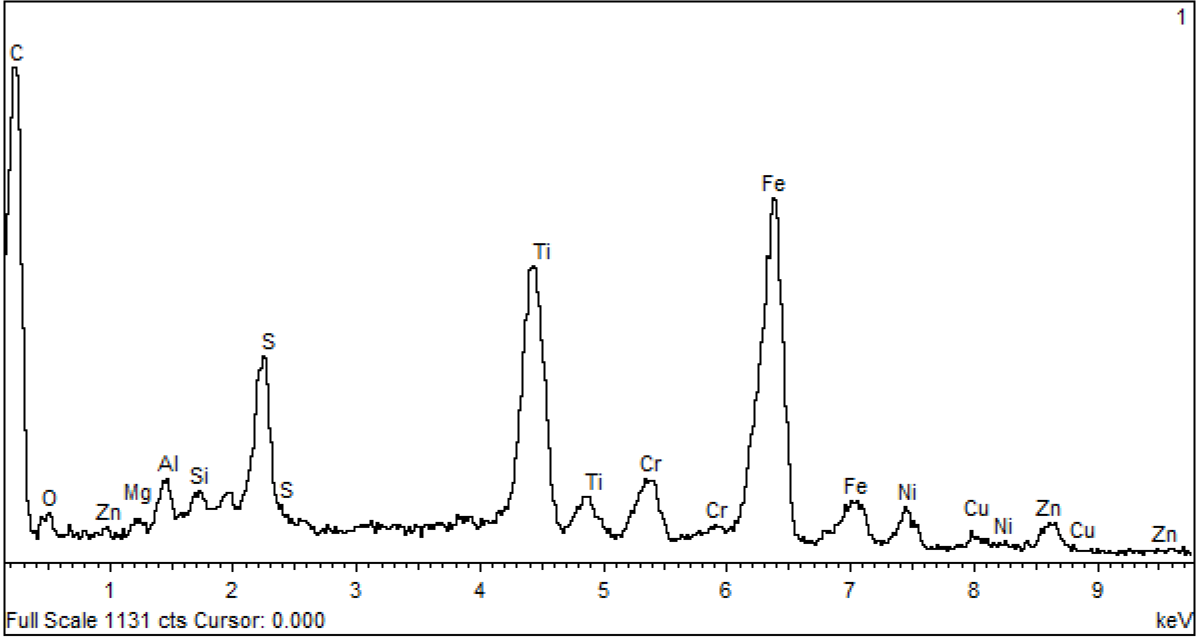

5
S
C
O
Mg
Al
Si
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Zn
1
2
3
4
5
6
7
8
Full Scale 4159 cts Cursor: 0.000
keV

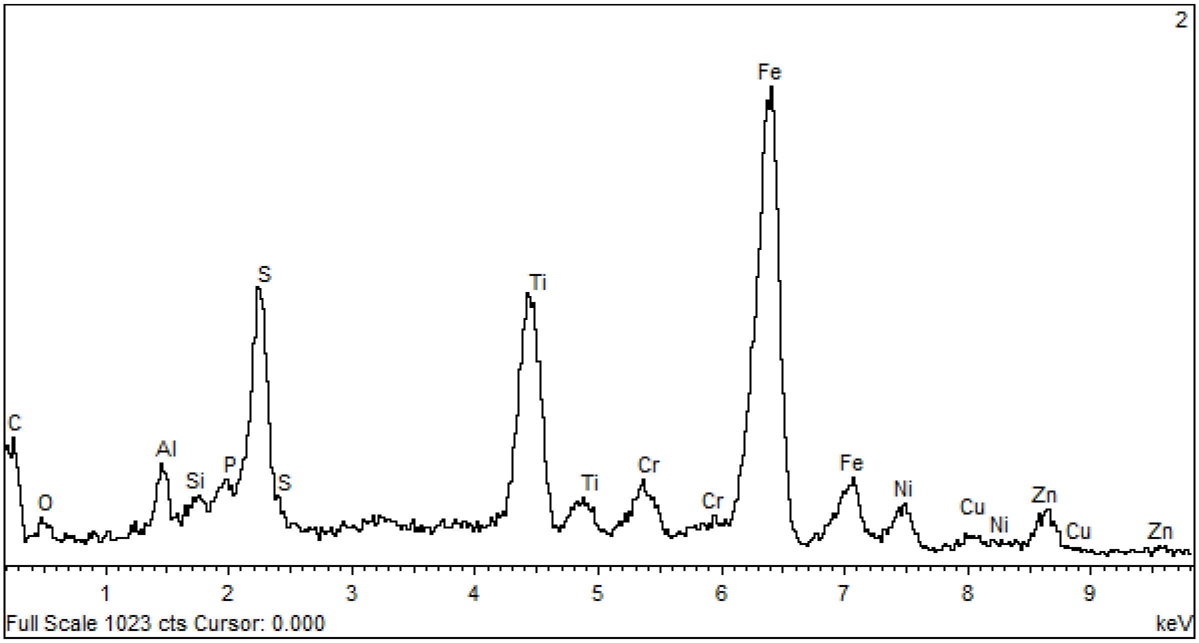

6
S
C
Al
O
Fe
Zn
Mg
Si
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Ni
Zn
Cu
Zn
1
2
3
4
5
6
7
8
9
Full Scale 3536 cts Cursor: 0.000
keV

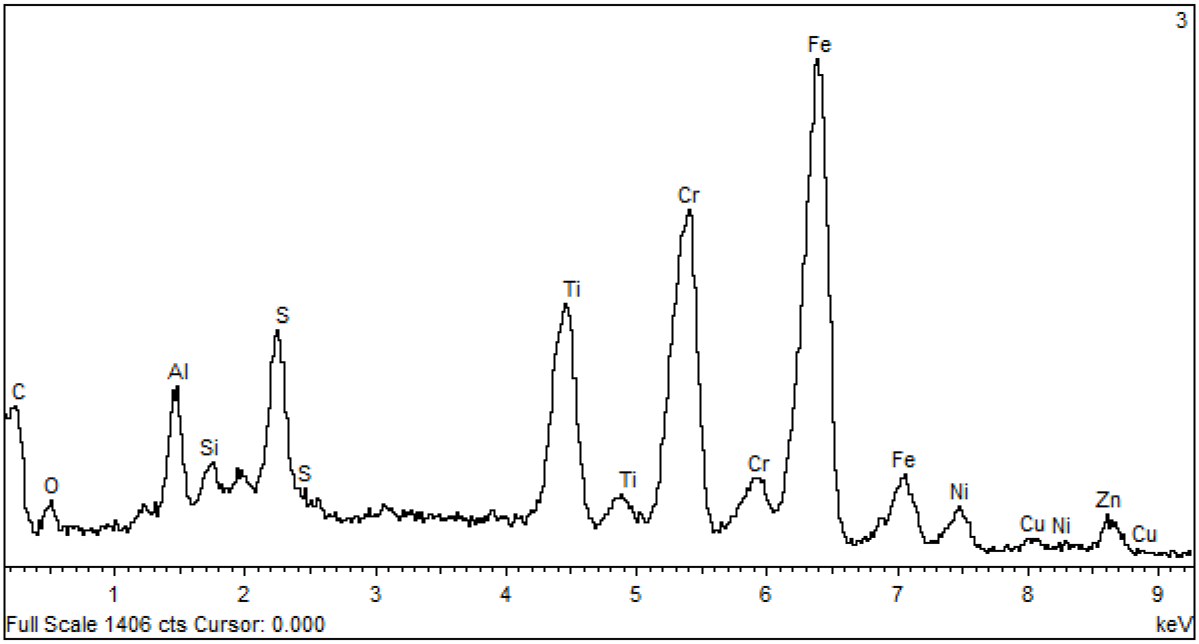
SEM images of the Mid-section of the SE Coalescer removed on April 2015
SE_MID_Slice 1 300318881

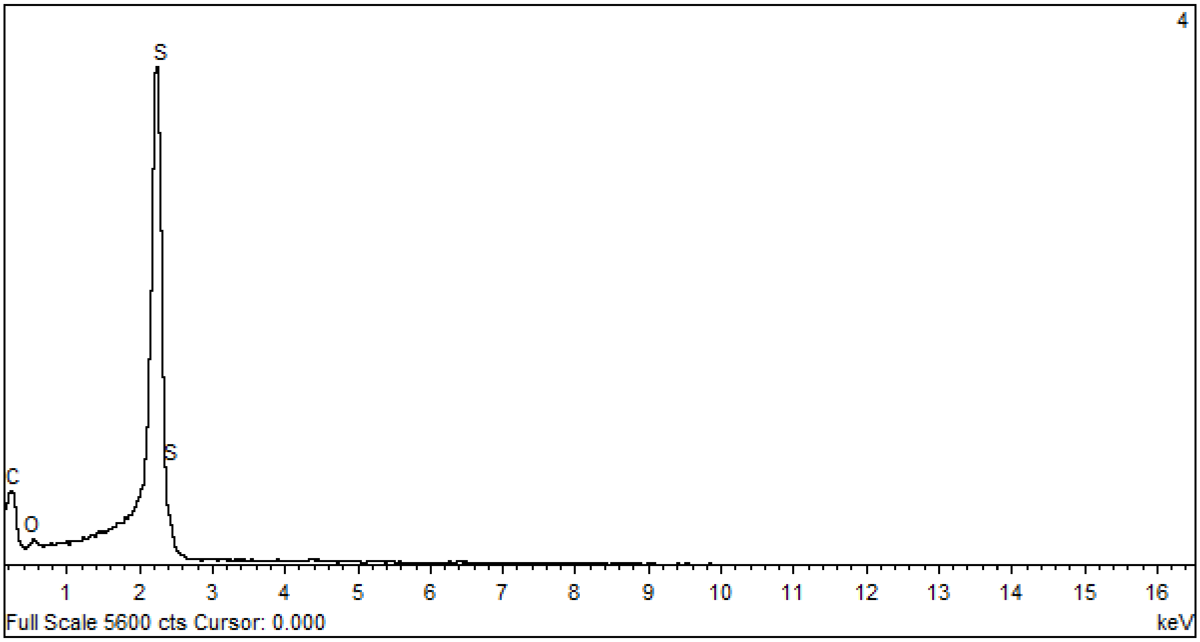


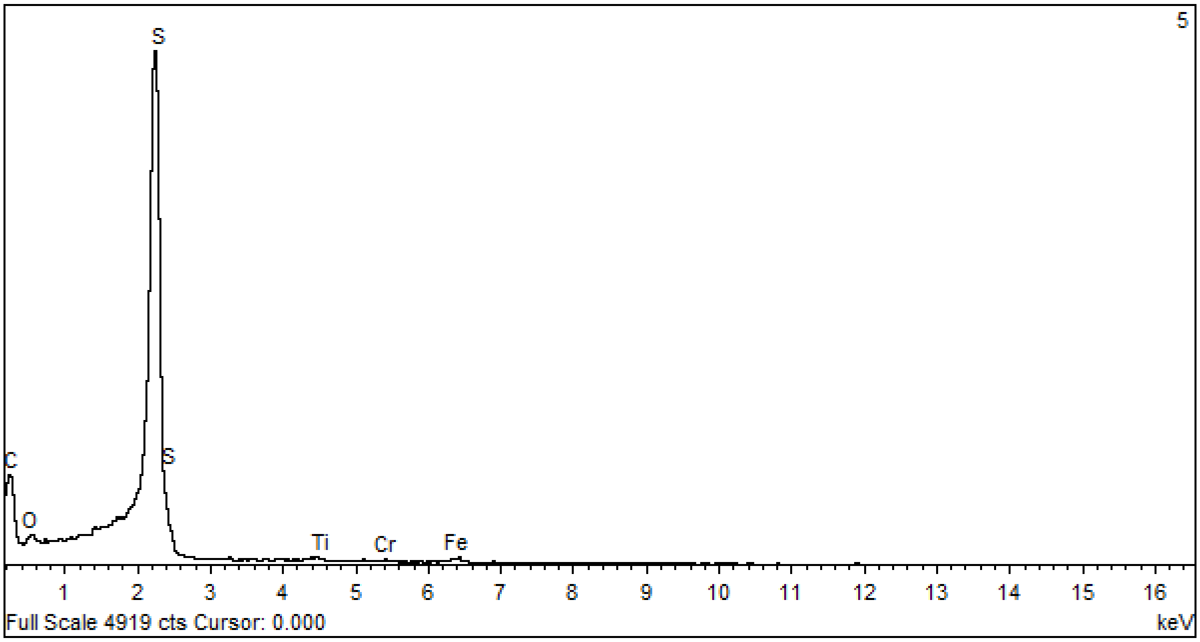
Note: Number with an asterisk (*) indicates the actual data point. Otherwise, they are representative. Representative EDS spectra are similar enough to each other that they are essentially identical with minor variations in peak heights, but not identified elements.

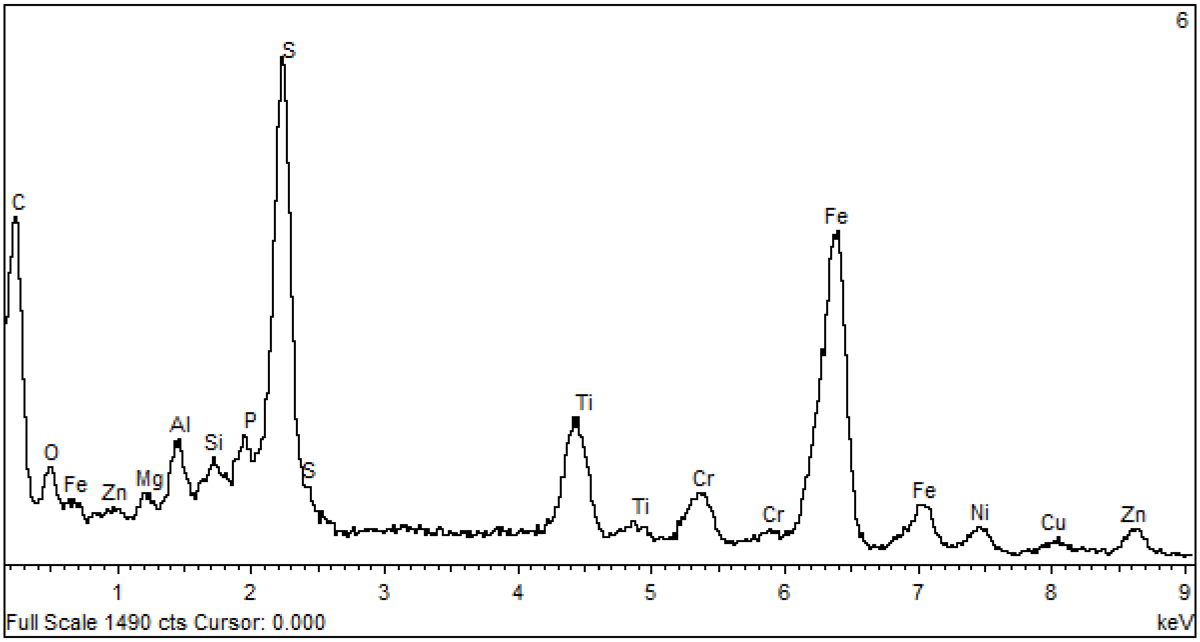


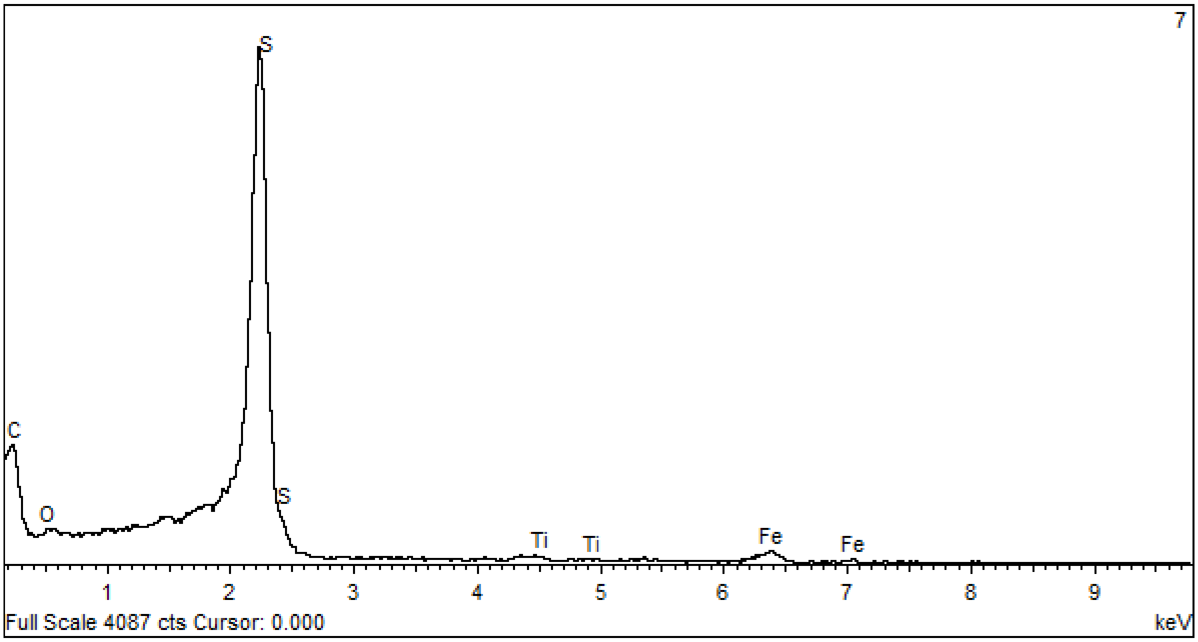

2
Fe
S
Ti
C
Al
Si
P
S
O
Ti
Cr
Cr
Fe
Ni
Cu
Ni
Zn
Cu
Zn
1
2
3
4
5
6
7
8
9
keV
Full Scale 1023 cts Cursor: 0.000

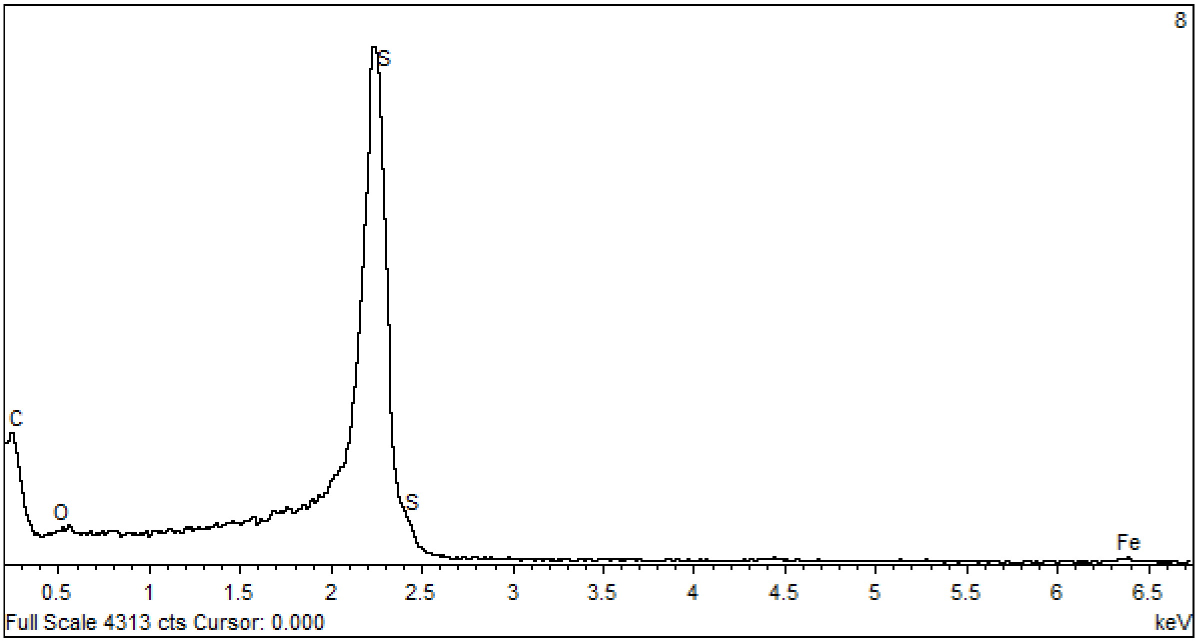

3
Fe
Cr
Ti
S
Al
C
Si
O
S
Ti
Cr
Fe
Ni
Cu
Ni
Zn
Cu
1
2
3
4
5
6
7
8
9
keV
Full Scale 1406 cts Cursor: 0.000

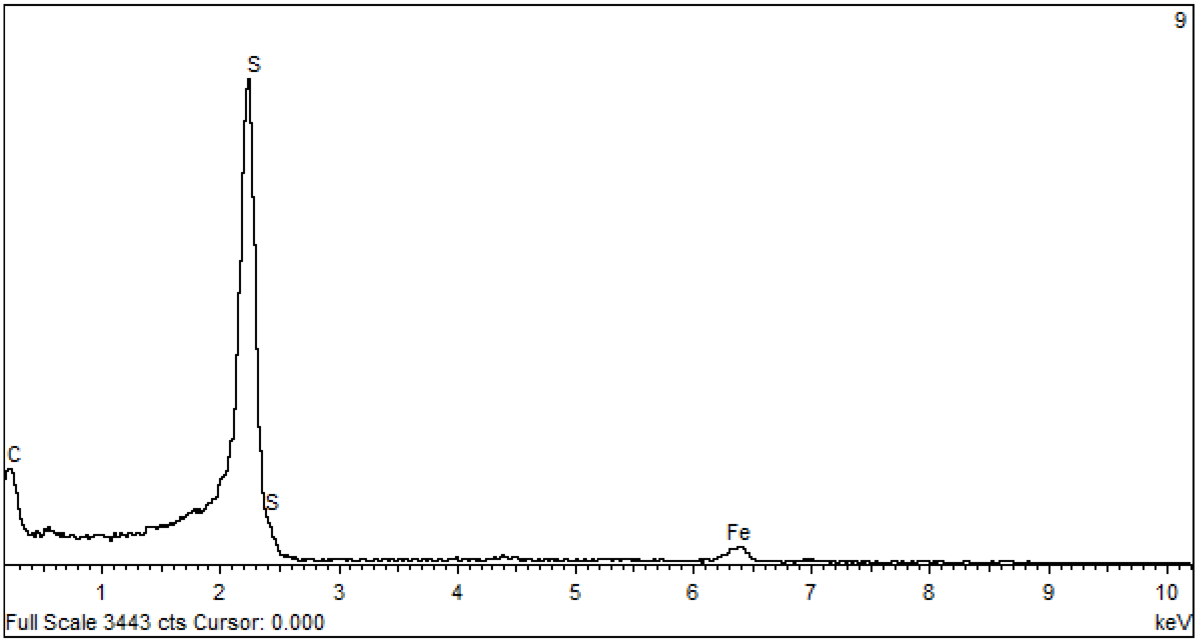

4
S
S
C
O
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16
Full Scale 5600 cts Cursor: 0.000
keV

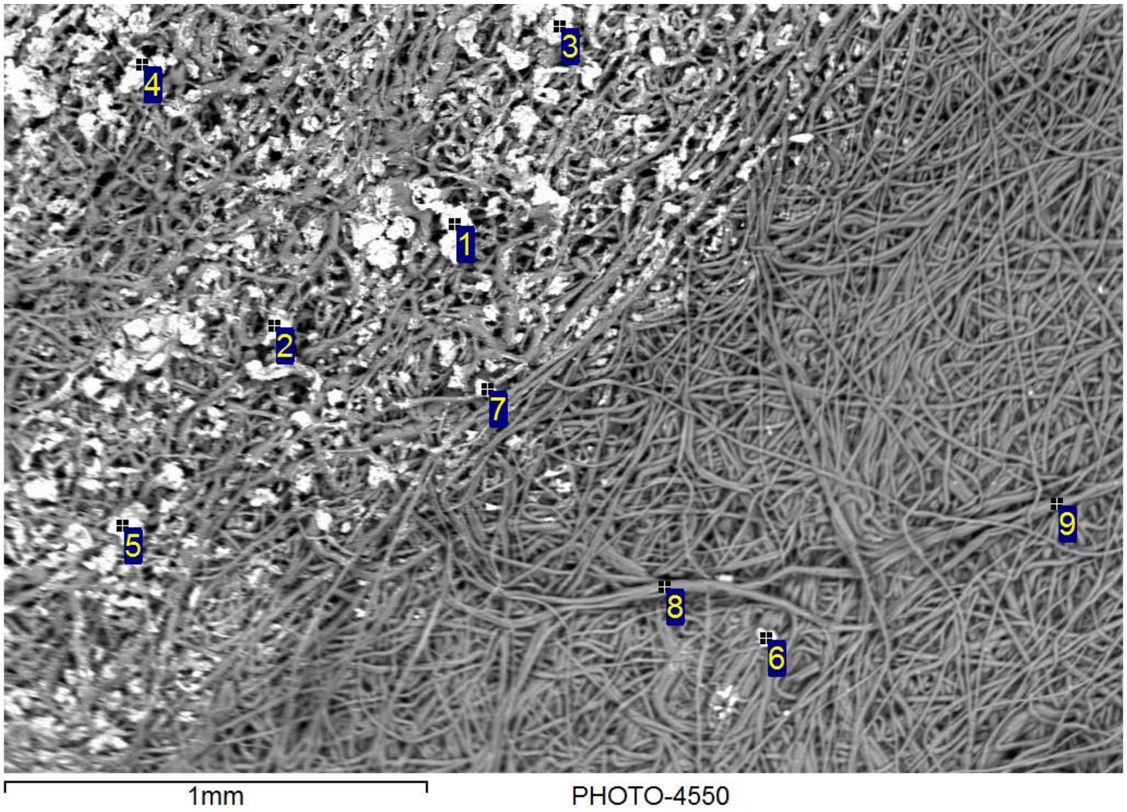

5
S
S
C
O
Ti
Cr
Fe
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16
Full Scale 4919 cts Cursor: 0.000
keV

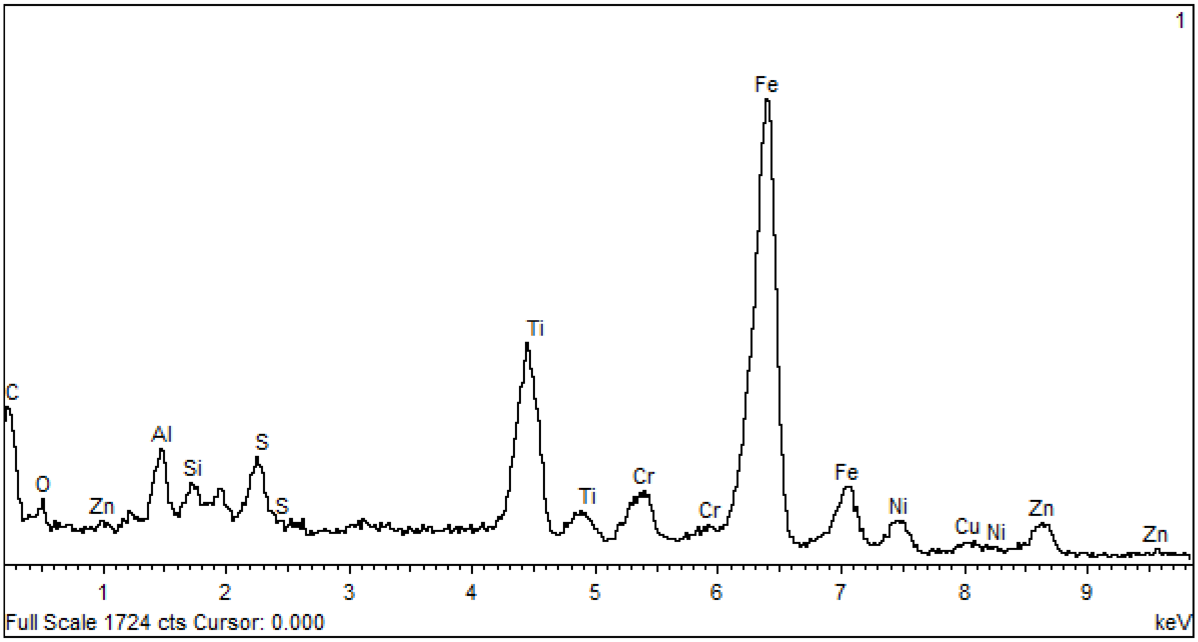

6
S
C
Fe
Ti
Al
P
O
Si
Fe
Zn
Mg
S
Cr
Ti
Cr
Fe
Ni
Cu
Zn
1
2
3
4
5
6
7
8
9
keV
Full Scale 1490 cts Cursor: 0.000

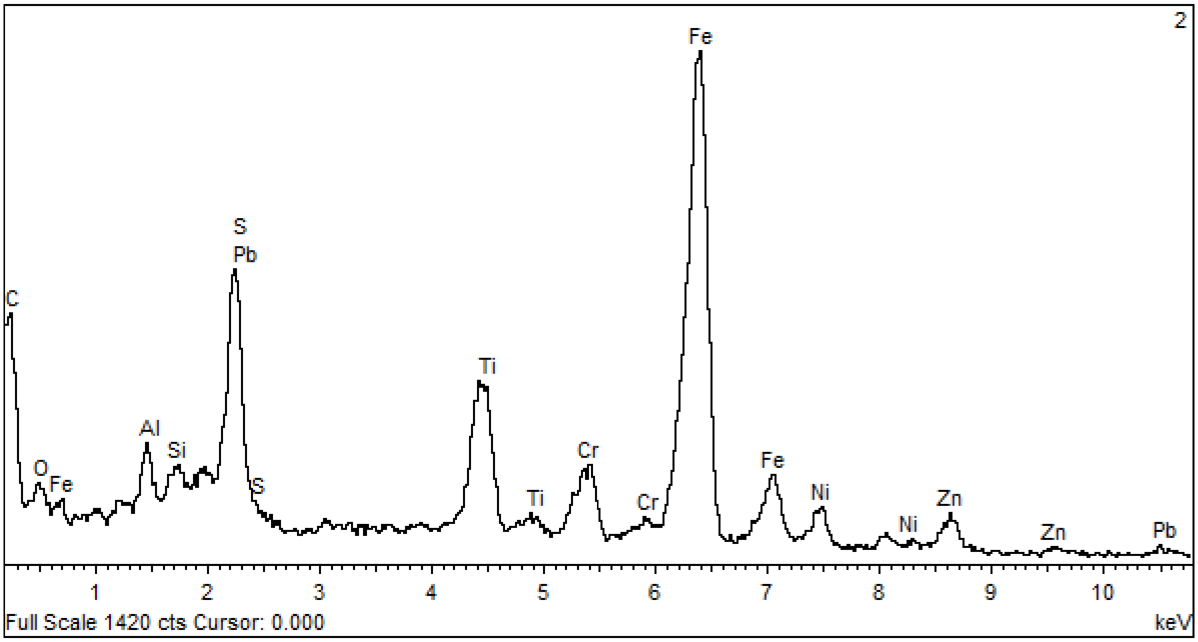

7
S
C
O
S
Ti
Ti
Fe
Fe
1
2
3
4
5
6
7
8
9
keV
Full Scale 4087 cts Cursor: 0.000

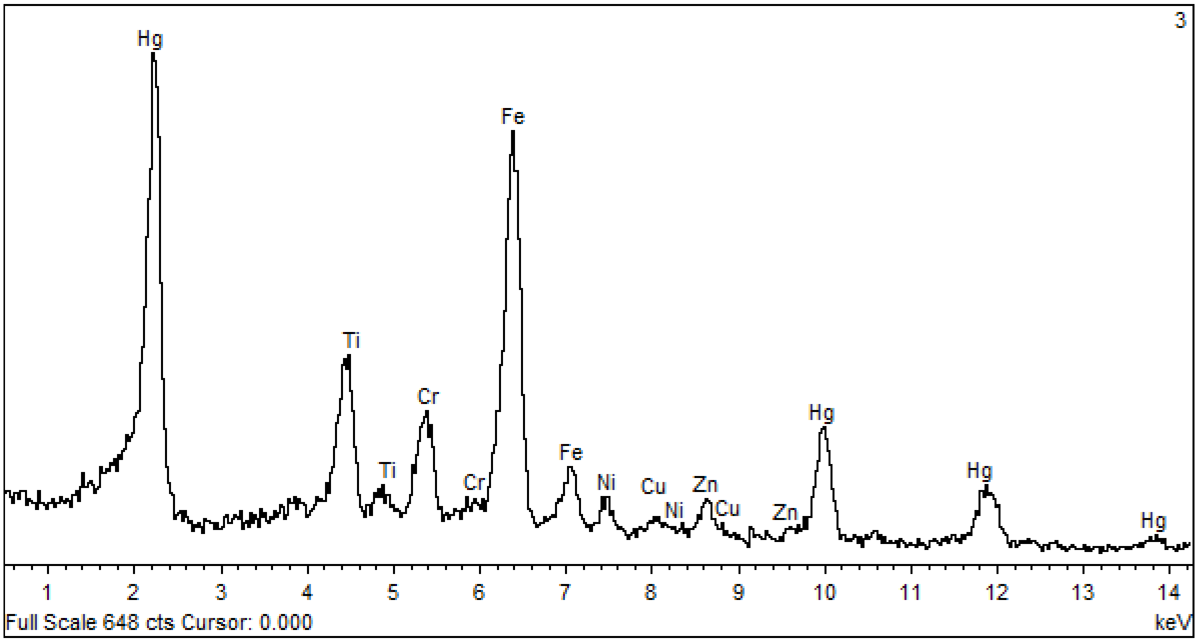

8
S
C
O
S
Fe
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5
5.5
6
6.5
Full Scale 4313 cts Cursor: 0.000
keV

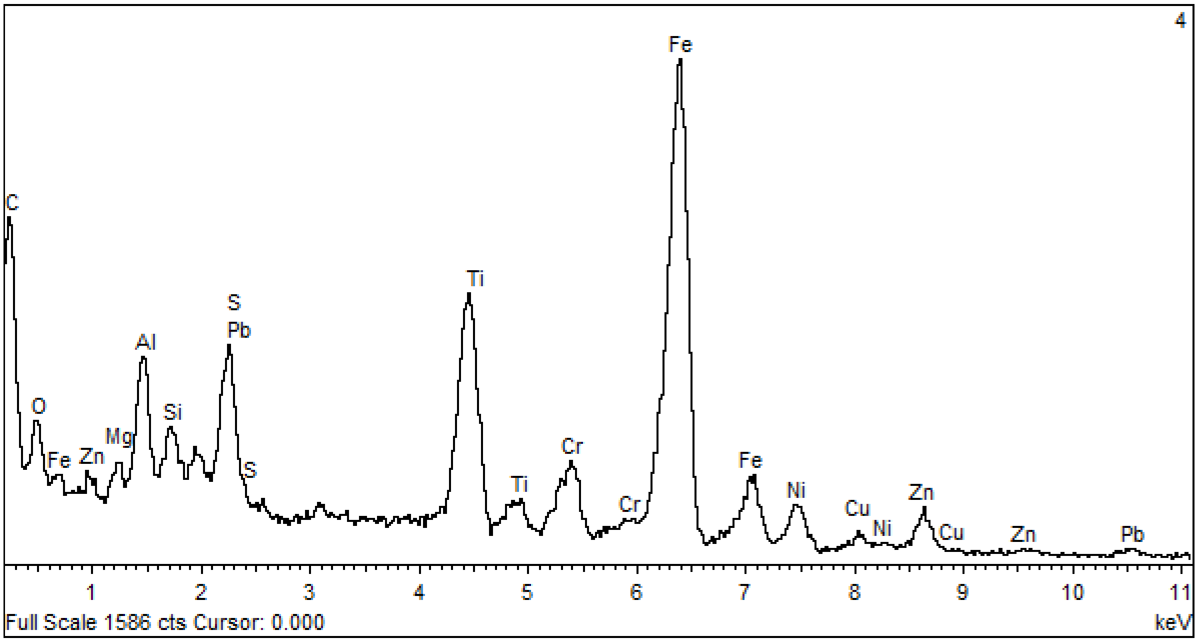

9
S
C
S
Fe
1
2
3
4
5
6
7
8
9
10
Full Scale 3443 cts Cursor: 0.000
keV

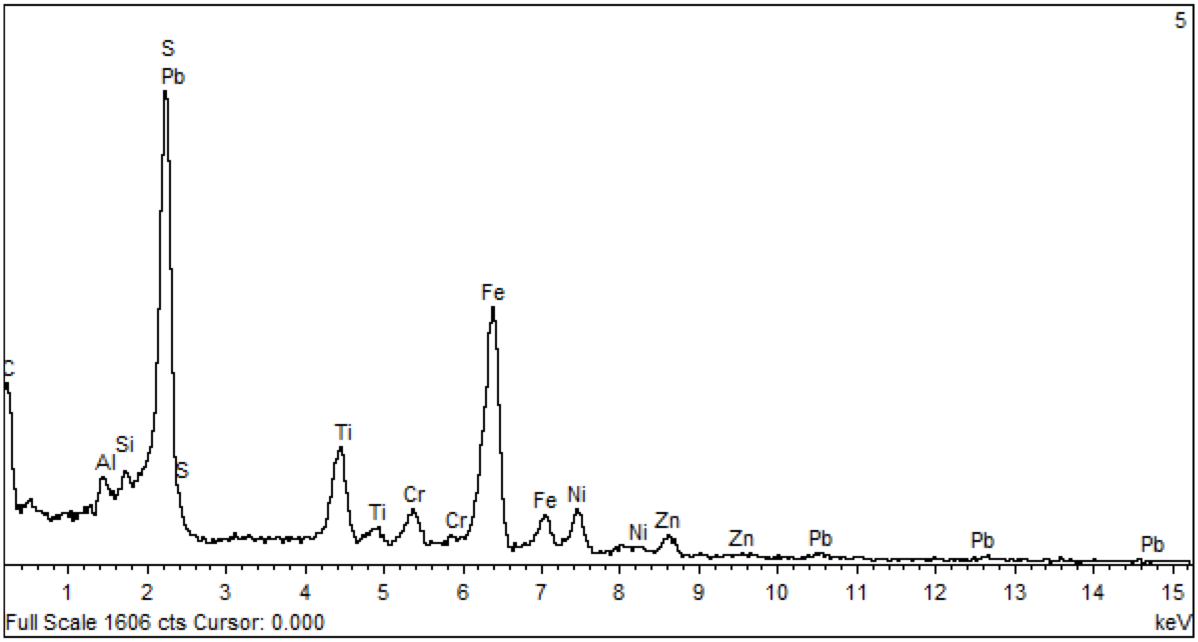

1
2
3
4
5
6
7
8
9
1mm
PHOTO-4550

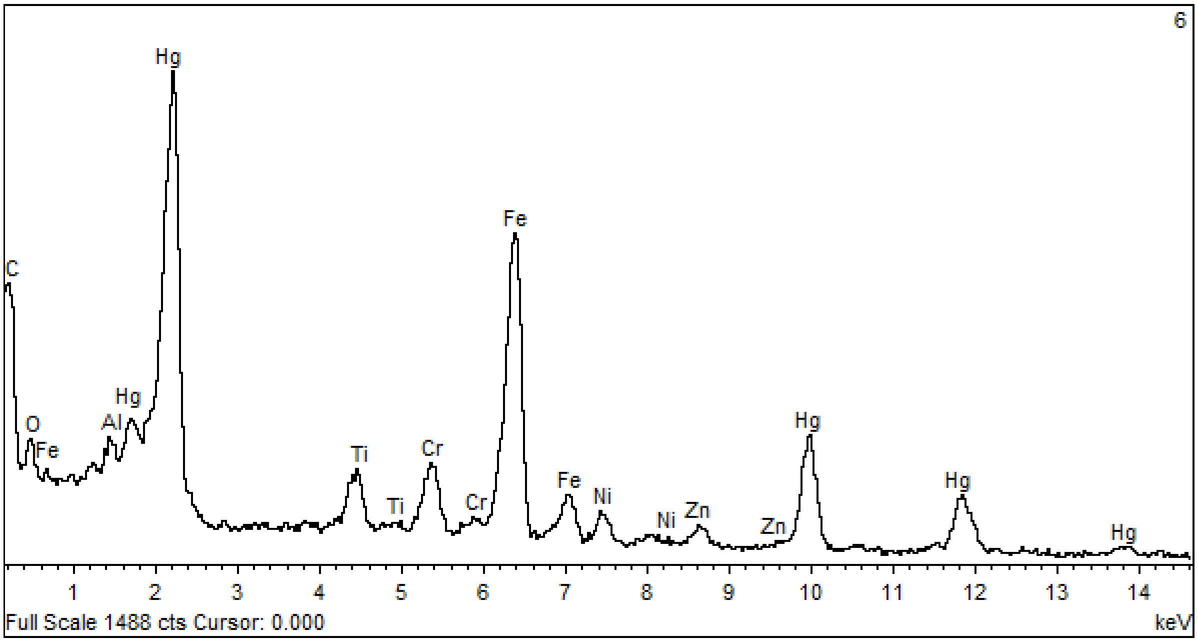

1
Fe
Ti
C
Al
Si
S
O
Zn
S
Ti
Cr
Cr
Fe
Ni
Cu
Ni
Zn
Zn
1
2
3
4
5
6
7
8
9
Full Scale 1724 cts Cursor: 0.000
keV

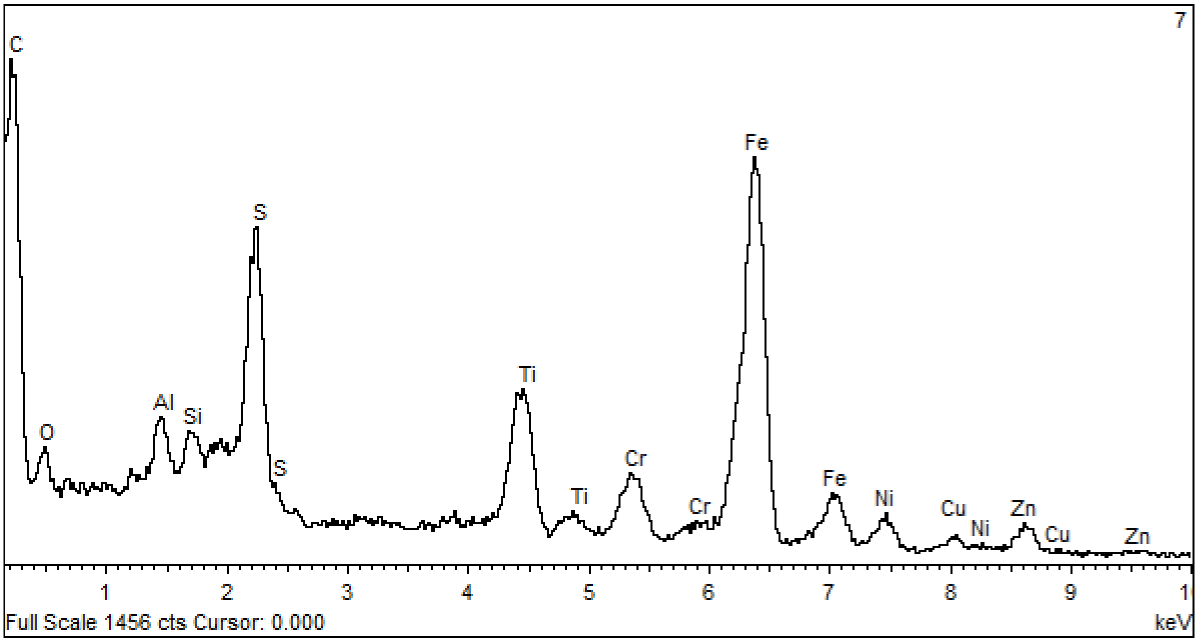

2
Fe
S
Pb
C
Ti
Al
Si
O
Fe
S
Cr
Ti
Cr
Fe
Ni
Ni
Zn
Zn
Pb
1
2
3
4
5
6
7
8
9
10
Full Scale 1420 cts Cursor: 0.000
keV

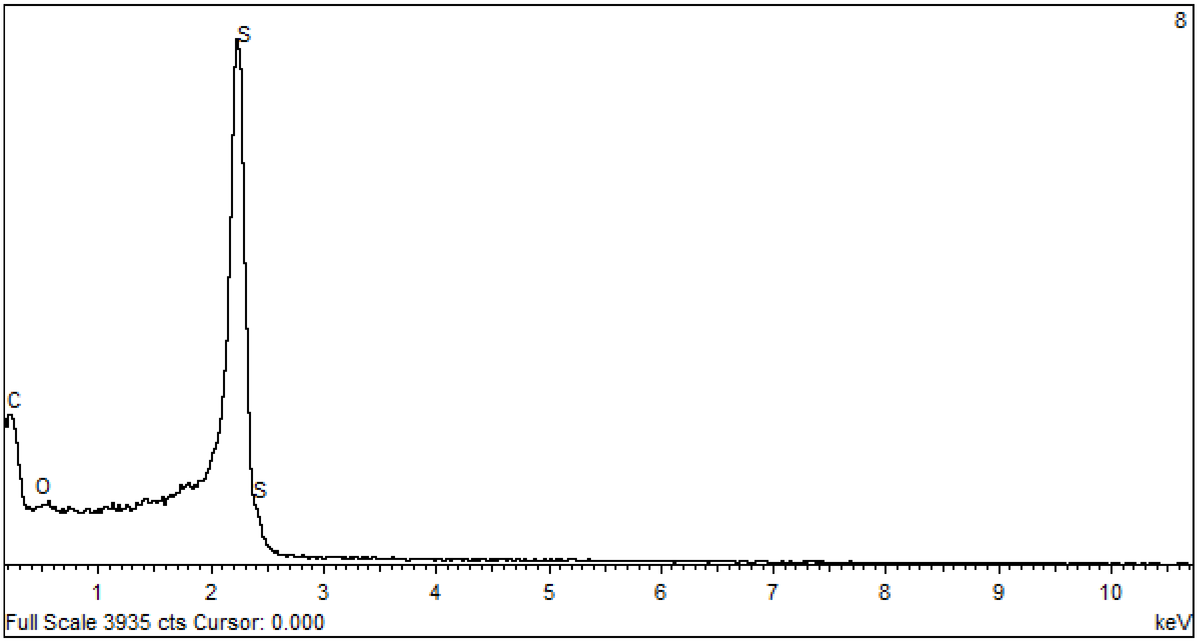

3
Hg
Fe
Ti
Cr
Hg
Ti
Cr
Fe
Ni
Cu
Zn
Ni
Cu
Zn
Hg
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Full Scale 648 cts Cursor: 0.000
keV

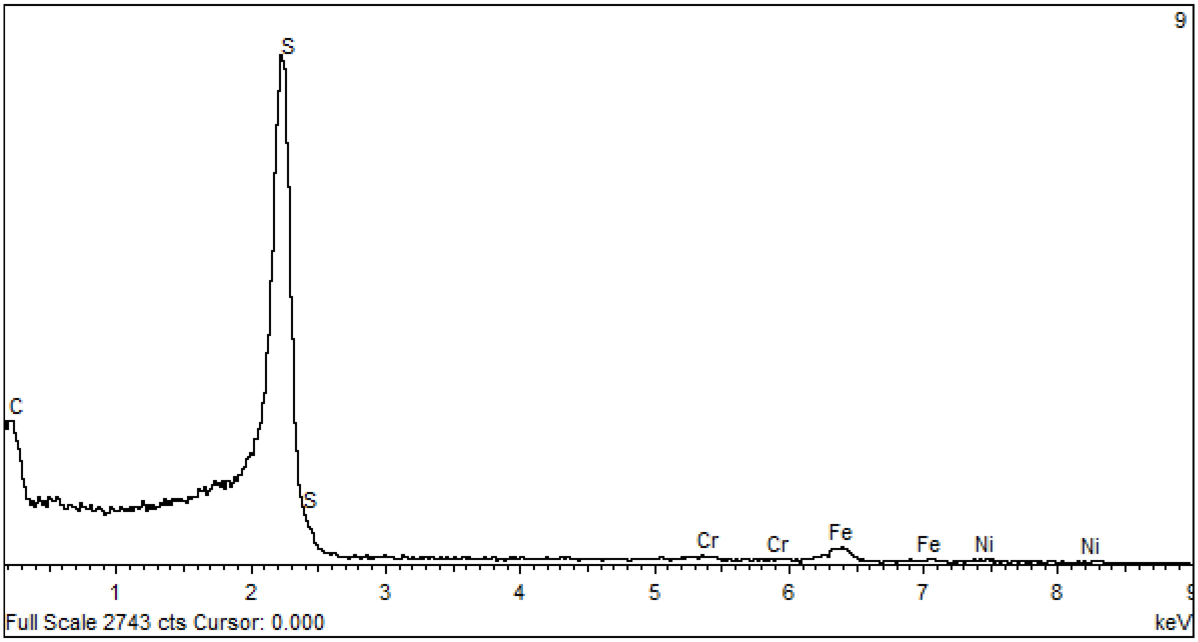

4
C
O
Fe
Zn
Mg
Al
Si
S
Pb
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Ni
Zn
Cu
Zn
Pb
1
2
3
4
5
6
7
8
9
10
11
Full Scale 1586 cts Cursor: 0.000
keV

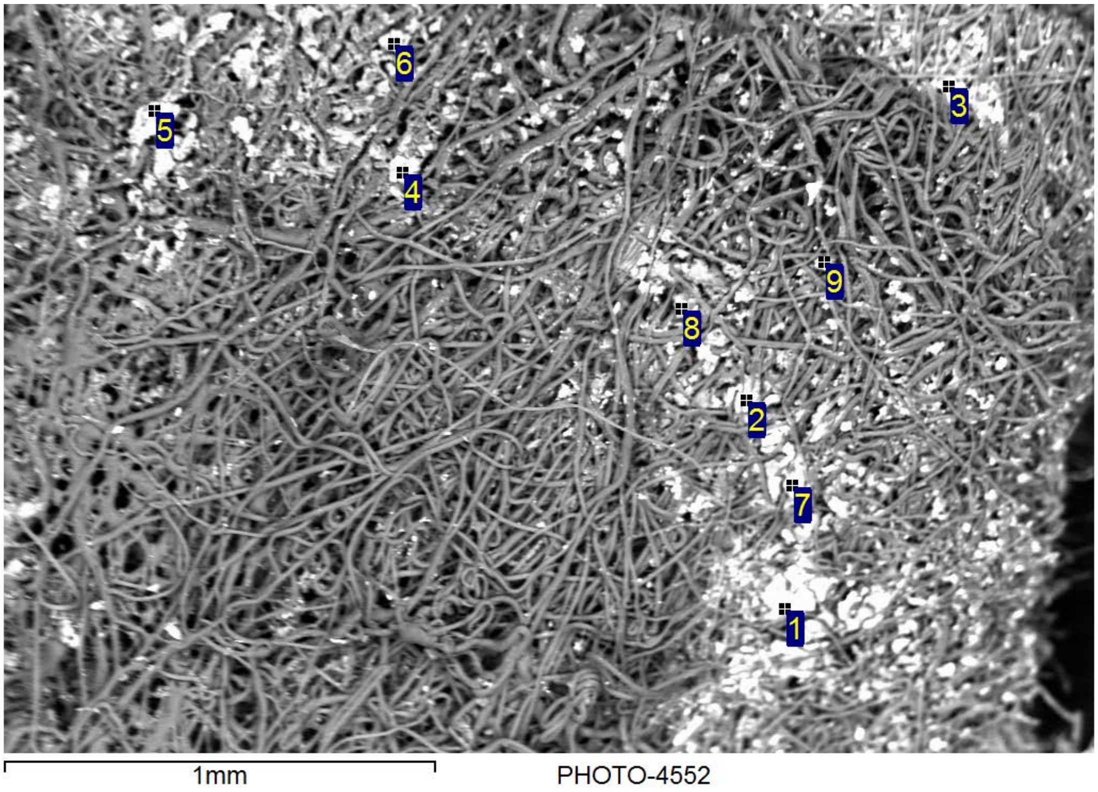

5
S
Pb
C
Al
Si
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Ni
Zn
Zn
Pb
Pb
Pb
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Full Scale 1606 cts Cursor: 0.000
keV

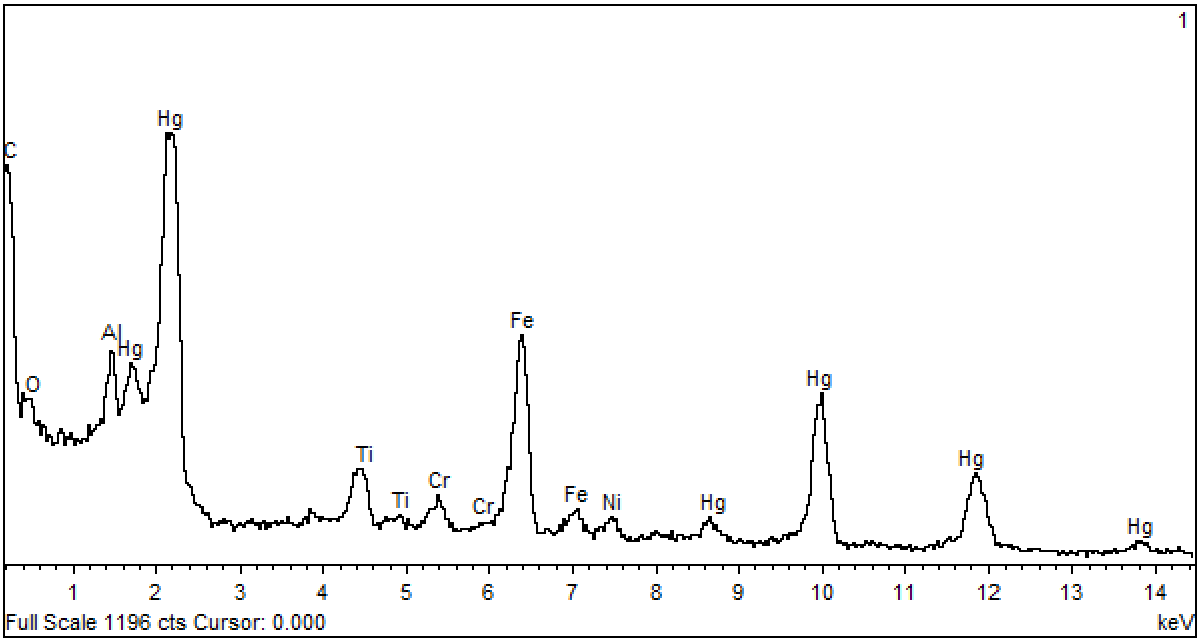

6
C
O
Fe
Al
Hg
Hg
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Ni
Zn
Zn
Hg
Hg
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Full Scale 1488 cts Cursor: 0.000
keV

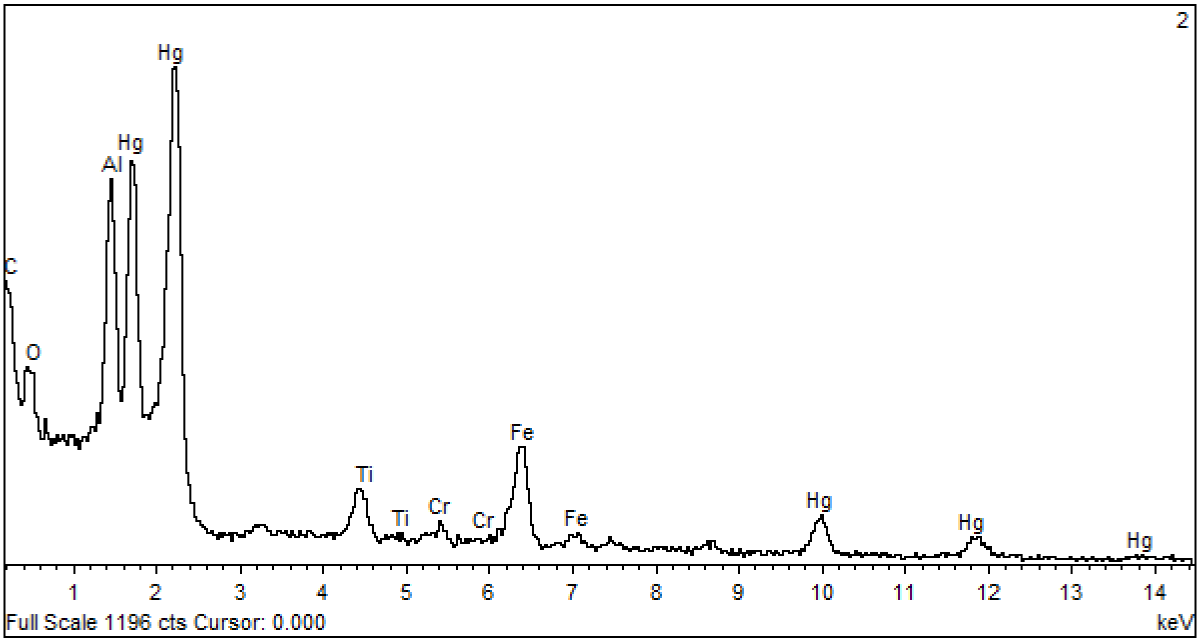

7
C
O
Al
Si
S
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Ni
Zn
Cu
Zn
1
2
3
4
5
6
7
8
9
1
Full Scale 1456 cts Cursor: 0.000
keV

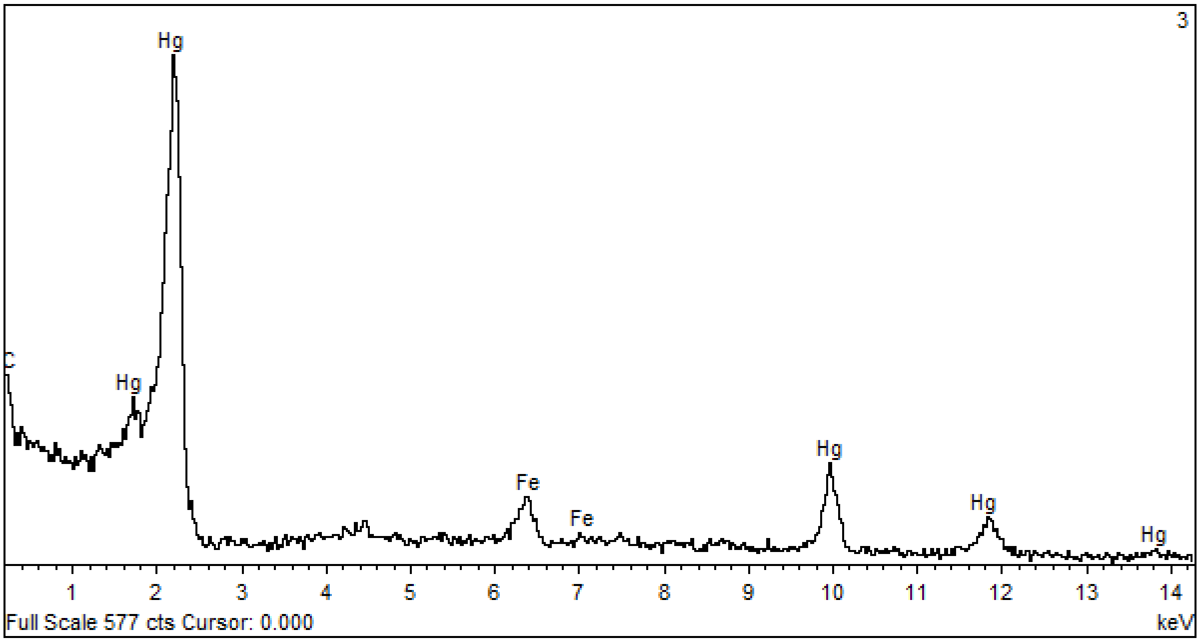

8
S
C
O
S
1
2
3
4
5
6
7
8
9
10
keV
Full Scale 3935 cts Cursor: 0.000

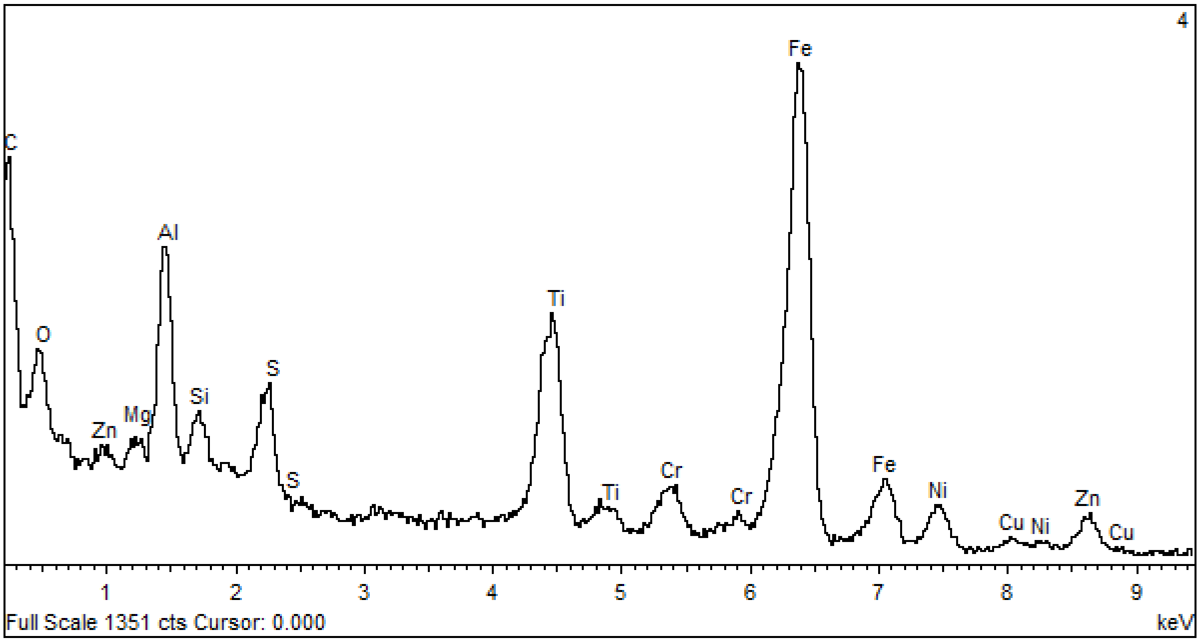

9
S
C
S
Cr
Cr
Fe
Fe
Ni
Ni
1
2
3
4
5
6
7
8
keV
Full Scale 2743 cts Cursor: 0.000

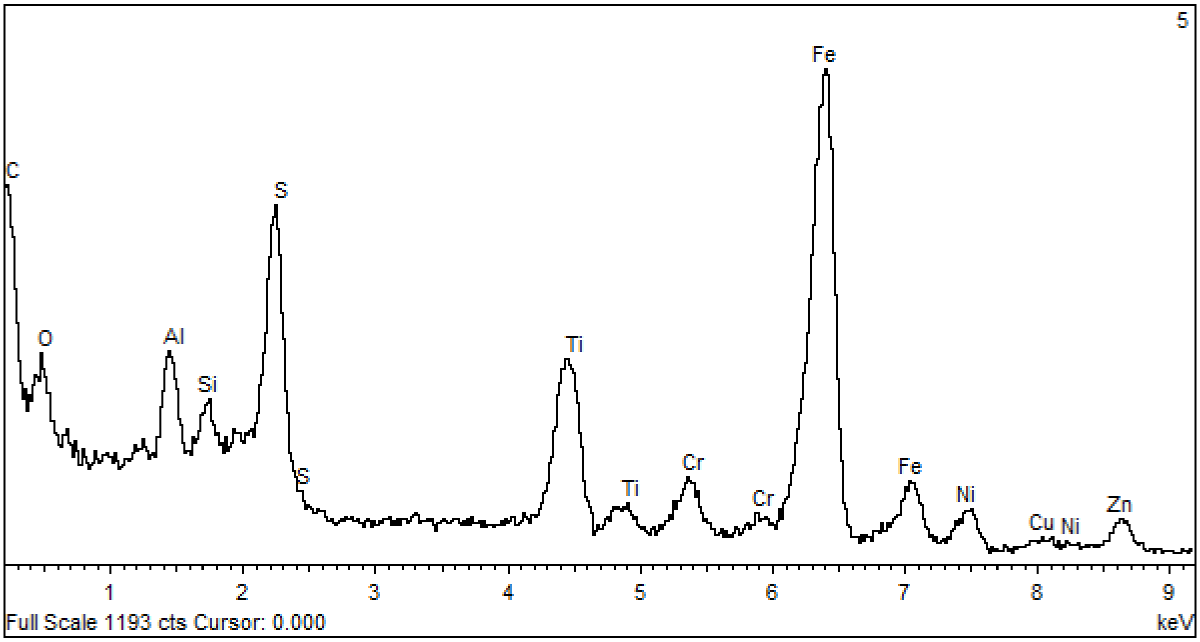

6
5
3
4
9
8
2
7
1
1mm
PHOTO-4552

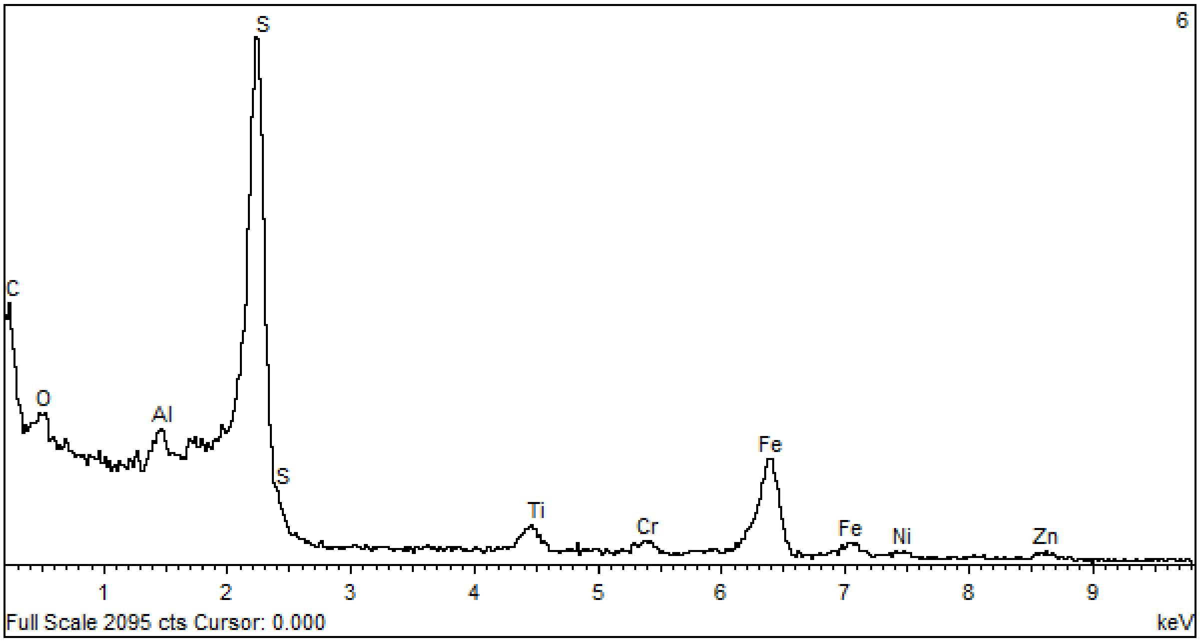

1
Hg
C
Al
Hg
O
Fe
Hg
Ti
Hg
Cr
Ti
Cr
Fe
Ni
Hg
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Full Scale 1196 cts Cursor: 0.000
keV

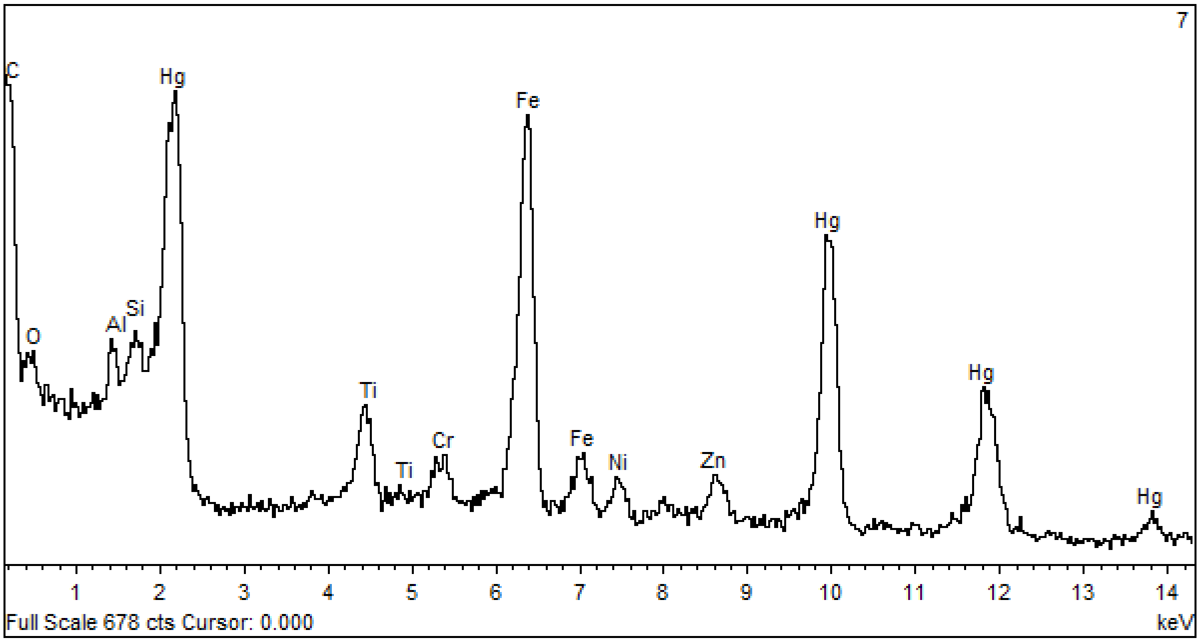

2
Hg
Hg
Al
C
O
Ti
Ti
Cr
Cr
Fe
Fe
Hg
Hg
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Full Scale 1196 cts Cursor: 0.000
keV

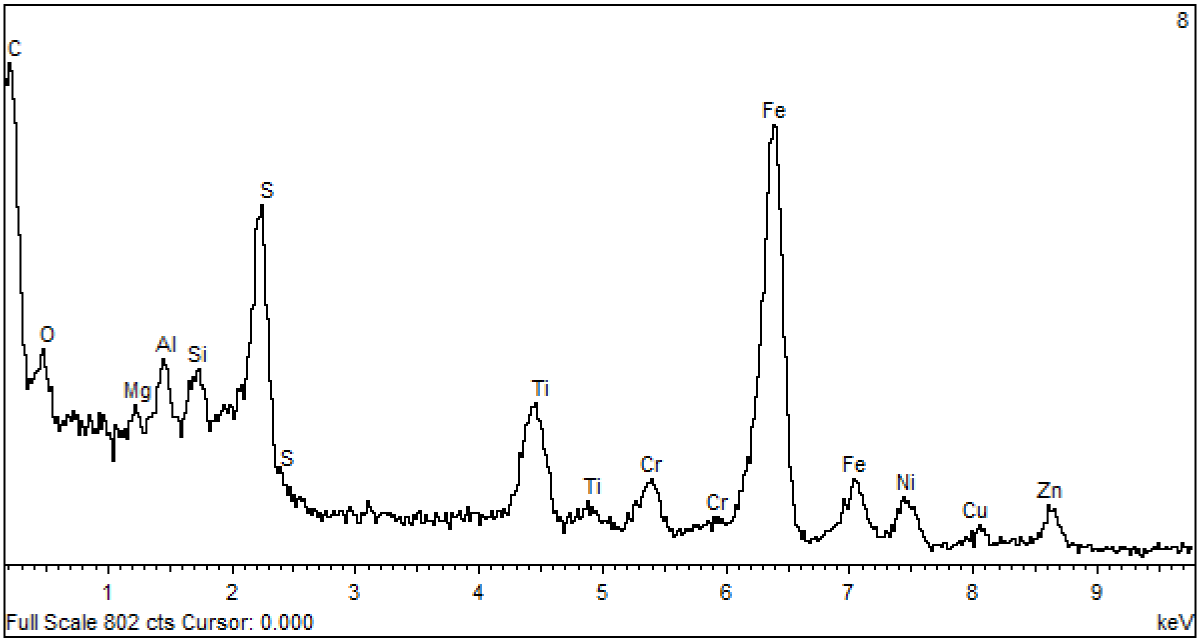

3
Hg
C
Hg
Fe
Fe
Hg
Hg
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Full Scale 577 cts Cursor: 0.000
keV

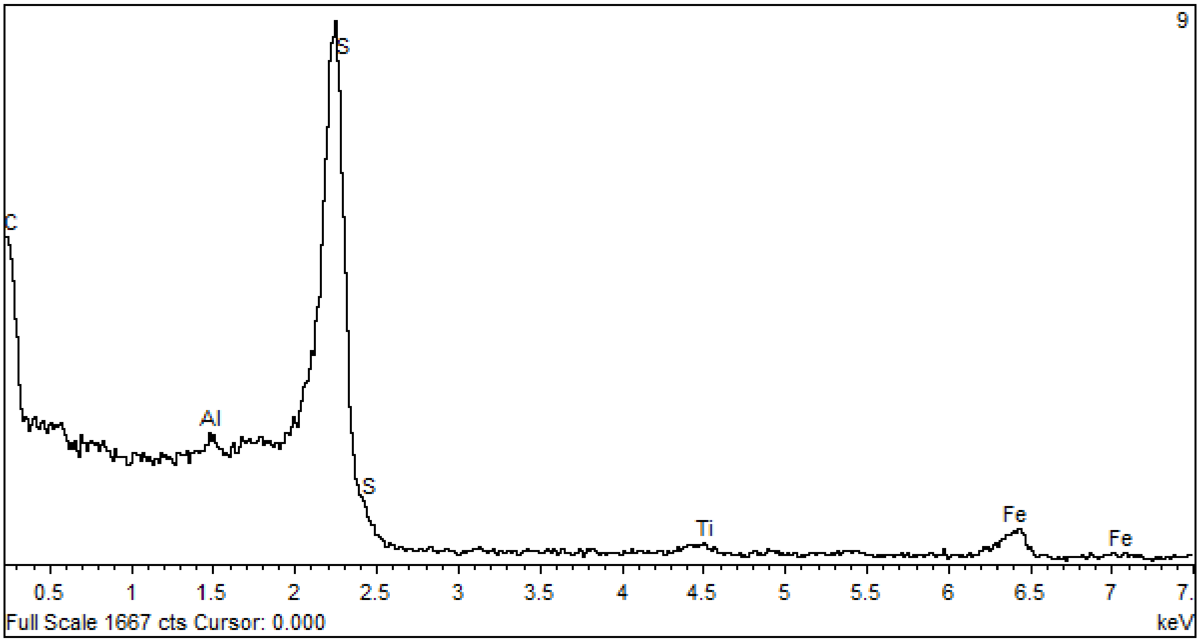

4
C
O
Zn
Mg
Al
Si
S
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Ni
Zn
Cu
1
2
3
4
5
6
7
8
9
Full Scale 1351 cts Cursor: 0.000
keV


5
C
O
Al
Si
S
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Ni
Zn
1
2
3
4
5
6
7
8
9
Full Scale 1193 cts Cursor: 0.000
keV


6
S
C
O
Al
S
Ti
Cr
Fe
Fe
Ni
Zn
1
2
3
4
5
6
7
8
9
keV
Full Scale 2095 cts Cursor: 0.000


7
C
Hg
Fe
Hg
Si
Al
O
Ti
Hg
Ti
Cr
Fe
Ni
Zn
Hg
1
2
3
4
5
6
7
8
9
10
11
12
13
14
keV
Full Scale 678 cts Cursor: 0.000


8
C
O
Mg
Al
Si
S
S
Ti
Ti
Cr
Cr
Fe
Fe
Ni
Cu
Zn
1
2
3
4
5
6
7
8
9
Full Scale 802 cts Cursor: 0.000
keV


9
S
C
Al
S
Ti
Fe
Fe
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5
5.5
6
6.5
7
7.
Full Scale 1667 cts Cursor: 0.000
keV

## Distribution:

T. B. Brown, 773-A
M. E. Cercy, 773-42A
D. A. Crowley, 773-43A
D. E. Dooley, 773-A
A. P. Fellinger. 773-42A
S. D. Fink, 773-A
C. C. Herman, 773-A
D. T. Hobbs, 773-A
E. N. Hoffman, 999-W
J. E. Hyatt, 773-A
K. M. Kostelnik, 773-42A
B. B. Looney, 773-42A
D. A. McGuire, 773-42A
T. O. Oliver, 773-42A
F. M. Pennebaker, 773-42A
G. N. Smoland, 773-42A
B. J. Wiedenman, 773-42A
W. R. Wilmarth, 773-A
Records Administration (EDWS)

E. A. Brass, 241-121H
C. K. Chiu, 704-27S
J. S. Contardi, 704-56H
A. G. Garrison, 241-121H
C. M. Santos, 241-152H
P. E. Fogelman, 241-121H
C. J. Scherman, 241-152H
K. M . Marra, 241-120H
B. A. Gifford, 704-56H
R. T. McNew, 766-H
E. J. Freed, 704-S
M. A. Rios-Armstrong
V. Jain, 766-H

P. R. Jackson, 703-46A
J. A. Crenshaw, 703-46A

T. B. Peters, 773-42A
F. F. Fondeur, 773-A
D. H. Jones, 773-A