Nanometric particles of high economic value in coal fire region: opportunities for social improvement

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Abstract

Spontaneous coal combustion in the La Guajira coals was studied for the presence of carbon nanophases (e.g., carbon nanotubes), occurrence of rare earth elements (REEs) in them, and the probable mechanisms for concentration of these rare compounds. For this purpose, various techniques such as scanning electron microscopy (SEM), Field-emission SEM, transmission electron microscopy (TEM), high-resolution TEM, and focused ion beam (FIB) were used. The development and alteration of the nanoparticles by geo-processes during the early modification periods of coal combustion were explored. Certain types of carbon nanophases and REE compounds may constitute nanominerals and ultra-fine particles accumulated in the coal peat. Assemblages of these nanophases (crystalline and amorphous compounds), predominantly the clay-monazite relationship and its connection to tonsteins in the coal combustion zones in the east region of the coal mines studied in this work, indicate that the coal area was subjected to REE concentration. The carbon nanophases contained several potential hazardous elements (PHEs), including, arsenic, bromine, cadmium, chlorine, fluorine, mercury, and other PHEs. While carbon nanotubes have been known to be produced from spontaneous combustion of coal of varying ranks, the present work is the first report on the naturally occurring REEs and carbon nanophases in the Colombian coal mining area.

Keywords: Rare earth elements; Carbon nanotubes; Spontaneous coal combustion; Advanced analytical approach; Colombian coals

1. Introduction

Globally, coal mining and subsequent coal fires are responsible for air, soil and water (e.g. gases, sulfuric acid, hazardous inorganic elements, polycyclic aromatic hydrocarbons) pollution involving, in part, a large amount of particulate matter which affects human health (Zheng et al., 2019; Hower et al., 2013; Ribeiro et al., 2010; Oliveira et al., 2019a,b,,d, 2018a,b, 2017, 2014; Gasparotto et al., 2018; Landim et al., 2018; Schneider et al., 2016). The International Agency for Research on Cancer (IARC), an agency operating under the World Health Organization (WHO), has cataloged outdoor air contaminants as the principal (Group 1) carcinogens affecting health (IARC, 2013). In addition to the many man-made threats to the atmosphere, self-combustion of coal also needs further scientific exploration (Kríbek et al., 2017; Garcia et al., 2014; Agudelo-Castañeda et al., 2017, 2016). The heterogeneity of a coal fire requires a more interdisciplinary approach to its local and global assessments (Dias et al., 2014). The coal-burning area studied in this work is located in the Department of La Guajira in northeastern Colombia between the areas of Albania, Barrancas, and Hatonuevo (Oliveira et al., 2019b). It is a combination of Wayúu ethnic settlements, a smaller Afro-Colombian population and rustic farming societies.

On the other hand, rare earth elements (REEs) and carbon nanoparticles (CNPs) are vital to the modern society as they are used in high-tech industry and a variety of consumer goods such as computers, cell phones, catalysis, fluorescent lighting, permanent magnets, medical devices and advanced defense technology (Dai et al., 2018; Liang et al., 2020). However, there is a sharp discrepancy between the high demand for and low production of REEs due to the limited availability of raw materials and feasible resources. Acute shortage of REEs has aroused concerns and has stimulated scientific research and technological developments for the recovery of REEs from secondary sources (Haberl et al., 2018). The REEs (including lanthanides and yttrium) are labeled as “critical materials” because of their importance for modern economy and the potential risks of supply disruptions (Liu et al., 2020). Over the recent years, there are growing interests in developing cost effective and environmentally friendly techniques for domestic recovery of REEs (Park et al., 2017). Coal combustion products have been suggested to be a promising REEs source (Dai and Finkelman, 2018).

Coal for power generation alone is not sustainable as in most countries mining has diminished due to the high cost of recoveries, depleted reserves and competition from natural gas and renewable energy sources (Nordin et al., 2018; Dias et al., 2014; Duarte et al., 2019). On the other hand, in Colombia, even though it is a Latin American country with the largest coal extraction industry, there is no coal power plant to generate enough electricity to supply a medium-sized city (between 500 and 1 million inhabitants). Since, the Colombian social impacts (as shown in Figure1) and health of people have been of the most concern to scientists (Guerrero-Castilla et al., 2019; Caballero-Gallardo et al., 2015). The present study aims to demonstrate that even in areas where self-combustion of coal occurs, NPs that are of high economic value can be found. The government and local people can exploit such areas where there are no concessions for large multinational coal mining.

Given this scenario, the present study aims to demonstrate that the presence of high economic value NPs can be a more profitable alternative to mining in Colombia. As such, coal mining is not only expected to provide cheap and profitable energy to other countries but can also be used as a source for extraction of NPs which can add value to coal mining waste treatment. In this study, an investigation on the fundamental occurrences of active coal mining in Colombia has been done in order to assess the motivations for future studies for extracting NPs from these mines and offer new insights into a bulk geochemical process.

2. Colombian Coal Mining Areas Studied and Coal Aspects

Forests are the key players in the global carbon cycle because they store up to 30 to 40% of terrestrial carbon (Rumpel, 2019). The net carbon balance in the sedimentary deposits in forests is driven by natural fires, which produce large carbon emissions and are necessary to maintain the productivity and biodiversity of these forests (Rumpel, 2019). Coal deposit areas also considered as carbon sinks, mainly because they accumulate large amounts of carbon in the form of coal of different ranks, such as anthracite, bituminous, etc. (De Groot et al., 2013). Advances in micro and nanoscale analyses, as well as experimental approaches, are improving the characterization of these bio-signatures and restricting abiotic processes when combined with the geological context (Kronbauer et al., 2013; Ribeiro et al., 2013a,b). In this context, a re-evaluation of the evidence of early coal formation is challenging but essential in order to develop an understanding of the origin and evolution of life, both on Earth and beyond it (Cerqueira et al., 2011, 2012). After all, by specifically investigating CNPs and rare earth elements in coal, rather than just quantifying, we can greatly improve our mechanistic understanding of how fire affects the ability of forests to act as a carbon sink on a wide scale (Walker et al., 2019).

The zone that has been investigated in this work is a Colombian coal mining zone where a coal fire has been revealed. These fires are multifaceted assemblies that include a mixture of organic and inorganic complexes that contribute to the geochemistry of O_2 and dispersion of NPs of a wide range of sizes. A large air flux can distinctly disperse temperature. However, for NPs, air depletion permits an increasing build-up of heat. Several coal seams in the coal fire zones were sampled, basically at the same time and under equivalent weather conditions so as to reduce environment differences. Forty-three illustrative in-situ coal fire samples were sampled from different La Guajira coal zones (Figure 1) during March 2017, December 2017 and May 2018. Further details on the sampling can be found in a previous study (Civeira et al., 2016a ; Oliveira et al., 2019b).

Colombia is currently the world's fourth largest thermal coal mining country. The major coal-bearing mines are in the Paleocene Cerrejón Formation, situated in the area of La Guajira (Figure 1), the Paleocene Los Cuervos Formation located in the zones of Cesar and Norte de Santander, and the upper Maastrichtian to Paleocene Guaduas Formation, located in the zones of Cundinamarca and Boyacá. Several authors have reported that the geological and petrographical nature of the studied area was formed in deltaic and intermediate environments (Quetame and Sarmiento, 2004; Bayona et al., 2004) and had a significant impact on the region (Guzman, 1991). The geology and petrographic features of the studied coal areas differ significantly with coal ranks alternating from lignite to bituminous and anthracite (López and Ward, 2008).

3. Materials and Methods

The studied samples from different areas near the exhaust were removed from the coal mine drainages and topsoil. The color phases (reddish, yellowish, and whitish) will not be discussed in this study as other authors, who have evaluated the environmental contamination of the studied area previously, have discussed them (Oliveira et al., 2019b). In the present study, we focus on the non-superficial phases of the samples, aiming to find the NPs that are the most resistant to spontaneous coal combustion.

Several traditional methods, such as Particle-Induced X-ray Emission (PIXE), X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), X-ray fluorescence spectrometer (XRF), Raman spectroscopy (RS), inductively coupled plasma atomic emission spectrometry (ICP-AES) for major chemical elements and inductively coupled plasma mass spectrometry (ICP-MS) for trace chemical elements are available for the assessment of the simple morphology and proportion of chemical elements in such samples (Gredilla et al., 2019, 2017; León-Mejía et al., 2016, 2018; Sehn et al., 2016; De Vallejuelo et al., 2017; Sindelar et al., 2014; Martinello et al., 2014; Arenas-Lago et al., 2013, 2014). However, these methods do not offer evidence of the ultra-fine/nano-structure or the geochemical configuration of a single ultrafine particle (Civeira et al., 2016b; Cutruneo et al., 2014; Nordin et al., 2018).

The study of single NPs can offer evidence of the geochemical development of the size of the assemblage and geochemical configuration of NPs (Saikia et al., 2015, 2014). Other spontaneous coal combustion studies on NPs have concentrated on their organization based on the size and geochemical structure of the whole NPs masses and only moderately based on the size, shape and geochemical configuration of individual NPs (Dalmora et al., 2016). Thus, considering the above mentioned aspects, field emission scanning electron microscopy (FE-SEM) combined with energy dispersive X-ray spectrometry (EDS) was applied to study the NPs and the precise size for other particles that have been described in previous works (Civeira et al., 2016c; Ramos et al., 2017; Quispe et al., 2012). They were initially investigated by a Nikon® SMZ645® stereoscopic optical microscope. Grain mounts for conducting studies under light microscopy were made with Cargille® Meltmount®. Photographs were taken with a Nikon® Labophot2-Pol® optical microscope having 10x, 20x and 40x objective lenses.

The microphotographs of complex mixtures were obtained by using a Nikon® Digital Sight DS-SM® camera. The refractive guides were determined by Cargille® Certified Refractive Index Liquids Series A and B for non-fibrous mixtures. The complex conformations of the detected NPs consisting of minerals and/or amorphous phases were investigated. This analytical approach is suitable for investigating the involvement of REEs and carbon nanotubes within the detected NPs as there are elements with high atomic numbers appearing in the bright zones of the image (Silva et al., 2011 a, b, c) while some with low atomic numbers appear in the dark-field zones (Silva et al., 2011 d, e). Thus, pictures of the NPs and precise sizes of 189 NPs were acquired and the findings have been summarized in the results and discussion section. The superficial geochemical configuration of the NPs was acquired by EDS (coupled with FE-SEM and H-TEM) from which the chemical elements were identified and that included Al, Ca, Fe, K, Mg, Mn, Na, P, S, Si, Ti, Zn and trace elements(Sánchez-Peña et al., 2018; Silva et al., 2009a,b; Wilcox et al., 2015).

4. Results and Discussion

In order to find the NPs that are most resistant to spontaneous coal combustion, numerous carbonaceous amorphous phases were measured. The principal NPs were amorphous NPs, organic coal phases that have an intact structure, clays containing a mixture of complex amorphous phases, isolated sulfate crystals and altered carbonaceous NPs, often including over 95% of the studied residual solids. These samples changed the geological composition of the environment in and nearby the studied coal area. At the Colombian coal mine areas studied in this work, considerable mineralization occurring due to an alteration in the coal itself caused by spontaneous coal fire has been noted (Oliveira et al., 2019). In addition, the high-temperature pyrometamorphism due to coal fires is one of the most widely recognized features (Ciesielczuk et al., 2014). Several particles containing REEs and carbon nanotubes were found to be generated by the thermal disintegration of aluminum silicates, carbonaceous matter, carbonates, and other oxides at approximately 800–1400 °C (Saxby, 2000). Similar results were reported in this work (e,g, Figures 2, 3, 4, 5). This is in agreement with elucidations specified for Latin American coals by previous authors (Dias et al., 2014; Oliveira et al., 2019).

The morphology of the REE-bearing ultra-fine particles and NPs is illustrated in Figure 2 of the supplementary material. It should be noted that the identified NPs are normally asymmetrical in form and typically cluster to form a mass. Additionally, more than 60% of REE particles were amorphous; individual minerals were rare. The geochemical configuration of several REE crystals mixed with amorphous phases, for example, monazite (Figure 2) and hydrated clays, may change during the sample investigation or when subjected to an electron beam vacuum. The observation of a great variety of these NP phases implies that they control the superficial area during the combustion of coal and the heterogeneous effects play an important role in the atmosphere, waters, and topsoil contamination. As illustrated in Figure 2, different forms of REEs were found in the particles detected in the study area. Several studies report that monazite is one of the main forms of REEs found in coal deposits. Monazite usually occurs in association with Al-Si-clays owing to the high temperatures of coal fires. Such clays may undergo chemical and morphological degradation, thus justifying the observation of Al, Si, K, and Mg during EDS analysis, as exemplified in Figure 2. In addition, high temperatures and other environmental factors (such as sulfuric acid formation) during coal combustion can fragment the original monazite particles leading to the formation of nanoparticles of size between 5 and 120 nm. Such a monazite degradation process has been previously reported in another study (Silva et al., 2010) in which hot sulfuric acid was added to a phosphate rock containing monazite, which justifies the interpretation of the REE particles in the present study. The dehydration of clays (e.g., kaolinite) depends on the temperature of the coal fire. The FE-SEM analysis established the moderately multifaceted behavior of these clays in coal burned zones. This fact is directly associated with the observation of NPs and ultrafine particles containing REEs.

An abundant quantity of ultrafine carbonaceous NPs containing halogens (e.g., fluorine, Figure 3) was detected in the Colombian spontaneous coal combustion. These NPs may combine with the ambient air. Figure 3 shows a wide variety of carbon nanotubes with various diameters and lengths. This can be better observed from Table 1 in which, besides the dimensional characteristics, the main chemical elements associated with such carbonaceous NPs are also given. It is important to mention that no halogen-containing particles were crystalline indicating that such elements were present as amorphous phases associated with carbonaceous matter and/or phosphate minerals. More importantly, only carbon nanotubes were detected in the blackest samples. The samples that were in large numbers and showed yellowish as well as reddish color did not contain such NPs. Although the NPs were detected in more than 95% of the samples, carbon nanotubes were detected in only 16 of the 43 samples. This implies that in the samples collected superficially in the previous study (Oliveira et al., 2019), no carbon nanotubes were detected, probably because in such samples sublimates generated from the decomposition of the coal components are formed. Therefore, carbon nanotubes occur only in samples collected from areas below a depth of 30 cm, as is the case in the present study. Depending on the Colombian coal's self-combustion conditions, the studied samples also have the environment required for spontaneous burning but do not essentially have self-combustion properties. Thus, the possibility of coal oxidation is moderately high in the area. For a better understanding of the occurrence of carbon nanotubes, further studies in which coal self-combustion does not occur need to be carried out (Deng *et al.,* 2018).

Referring to Table 1 and the results reported by Oliveira et al., 2019, it can be confirmed that samples CF 1, CF 17 and CF 38, being the closest samples where mullite and salammoniac were detected, indicate that carbon nanotubes were formed due to higher temperatures, as these two minerals are formed only at temperatures above 900 ^{o}C. This indicates that in the area under study, the geochemistry of carbon nanotube formation is associated with REE phases. Several previous authors have studied the geochemical speciation and physical dispersal of REEs in coal and coal by-products in order to develop effective approaches for REEs extraction (Taggart et al., 2018; Liu et al., 2019). Datas from those works exposed that REE-bearing phases might undergo varied degrees of speciation change and re-distribution during coal combustion. For example, REE-bearing phases (e.g., monazite and zircon) displayed morphological characteristics of spherical shape (Liu et al., 2019) and size reduction (Hood et al., 2017) as observed by scanning electron microscopy (SEM). In addition to the occurrence of REE-bearing particles encapsulated in aluminosilicate glass phase (e.g., Hower et al., 2018), REEs were also found to be dispersed throughout glass, likely resulting from the decomposition of REE-bearing phases and mobilization into glass (Liu et al., 2019). Moreover, most REEs are generally present at trivalent oxidation state, while Ce can occur as both Ce(III) and Ce(IV). Using micro X-ray absorption spectroscopy (μ-XAS), Stuckman et al. (2018) found 10-μm Ce(IV) oxides and partially oxidized Ce-bearing particles in CA, which might have resulted from the decomposition and oxidation of REE-bearing phases during coal combustion.

The results presented in Table 1 motivate further extraction studies, after all, the detected carbon nanotubes adsorb various hazardous elements, including highly volatile elements such as Hg, As, F, Br, among others. The datas from this work, if REEs-organic complexes, REEs carbonates, and REEs-bearing carbonates are present in coal fire area, they would decompose and Ce would oxidize during coal spontaneous combustion, and finally occur as REEs oxides or REE-bearing oxides. Such findings are consistent with previous studies on REEs speciation in CA at both micro and bulk scales. At the micro scale, REEs oxides (Montross et al., 2018) and REEs-bearing lime in coal ashes (Liu et al., 2019) have been observed using SEM-EDS. At the bulk scale, REEs-organic complexes are estimated to account for ~25% of total REEs in coal (Lin et al., 2017), therefore, it is reasonable to expect that REEs oxides would be a significant fraction in CA if the abovementioned transformations occur during coal combustion.

The significant alteration in the coal configuration and geology, i.e., the assembly of the samples studied from the spontaneous coal combustion zones, are simply identified from FE-SEM and H-TEM analyses as “oxidation rims” along with coal fire-grain margins or cracks and openings. Numerous carbonaceous NPs containing Cr, Na, and Si (Figure 4) have also been detected in the studied zones and cracks containing hot gas and water discharge. These data are from the sub-surface coal combustion and carbonaceous topsoil debris.

4.1 Environmental Considerations

The study on the relationship of the complex NPs found from the Colombian mines due to the spontaneous combustion of coal needs a multifaceted interdisciplinary methodology and can be done in three steps: mapping, modeling, and inspection.

Significant quantities of inorganic and organic NPs encompassing hazardous complexes might have been liberated over many years. Besides, the hazardous components such as organic gases, As, Br, Cd, Ce, F, Hg, La, Pb, Se and others (Table 1) might have also been released into the atmosphere and preserved within the detected nanoparticles by geological developments that lead to an incomplete trapping of the gaseous vapors.

The distribution of semi-volatile dangerous compounds in the studied coal combustion zone corresponds to that of the volatile compounds existent in the Colombian coals. It can consequently be incidental that the large quantities of volatile (organic complexes, halogens, S, Hg, and Se) as well as the non-volatile elements (e.g., Si, Ce, La, Pr, Nd) found in the studied zones may be due to the large quantities of trace compounds in the original coal.

The data from this study indicate that the health risk of people who work and live in this region is highly compromised. Therefore, greater government action is required to reduce such impacts. A good proposal on the basis of the results presented would be a profound financial investment in order to undertake REE and carbon nanotube extraction and purification studies, as these materials have a high economic.

With respect to the hazardous element (HEs) concentrations detected in this study, the HEs found in the coal area at higher temperatures, reported by Oliveira et al., (2019), had higher proportions with small diameters. It revealed that partial vaporization during spontaneous coal combustion was followed by subsequent condensation of the semi-volatile elements on the coal solid nuclei. The exact progression by which HEs convert from the vapor level to the condensed phase is important for evaluating the human hazards from the by-products of spontaneous coal combustion.

4.2 Social vision

Engrained coal benefits have blocked the progress of a sustained fair and just transition movement in La Guajira by weakening union power, creating several advocacy groups that construct positive images of coal, and portraying local environmental activists as radical outsiders (Lewin et al., 2018). This is the dilemma where can the movement attract locals while simultaneously bringing in non-local participants. Non-local activists provide vital resources and help strengthen networks for building a national movement. Yet, in order to truly attain a fair and just transition and to practice energy democracy, those who bear the social costs of energy transitions must be part of the movement (Veelen and van der Horst, 2018). Where the pathway to contention seems clear, the pathway for standing together and having a debate about La Guajira's future does not. In order to stem ecological protest that would challenge their political power, the coal industry constructed economic identities that were strongly pro-coal in their ideology. Bell and York (2010) argue that the US coal industry successfully engaged in cultural manipulation through the astroturf group, Friends of Coal, by appropriating iconography and tying the region's history and culture to coal. Pro-coal culture gains resonance through effective frames of energy security and casting environmentalists’ arguments. In other hand, To moderate the worldwide challenge of climate change, several countries must burn less coal. For example, In recent years, the share of U.S electricity generated by coal has fallen from nearly 50% to 33%. In genral the global reduction in coal use for generating power is especially notable because it has occurred without the worldwide imposing carbon pricing or a carbon tax (Cragg et al., 2013). In U.S the substitution away from coal is mainly due to adoption of fracking technology and some states sharply ratcheting up their renewable portfolio standards (Eye et al., 2020).

Colombia has encouragements to promote the growth of one of their key industries. Given the durability of housing capital and the built up social networks established in coal mining areas, its residents face both migration costs and asset losses if the demand for coal mining declines. Such individuals face a fundamental job retraining challenge that middle-aged workers who have worked in mines will have trouble transitioning to other works. Resident officials in coal zones are well aware that many of their constituents depend on the continuing viability of the coal industry. Local officials internalize the benefits of coal’s prolonged sunset but they ignore the social environmental costs associated with such implicit subsidies.

Given all the challenges encountered in La Guajira, we believe that carbon nanophases and REEs quantification, processing and extraction studies can be a medium-term solution for the population to obtain more resources that are used to reduce the impacts of coal mining.

5. Conclusions

The morphology and geochemical structure of several REEs and CNPs were described using advanced electron beam spectroscopy techniques. The NPs were typically asymmetrical and produced due to the coalescence of primary compounds. The generation of amorphous and crystalline NPs, which strongly depends on the temperature in the spontaneous coal combustion areas, was detected. Complex carbon nanophase assemblages (carbon nanotubes, graphene, and several amorphous phases), hazardous elements and several REE phases were observed in the samples. The studied coal fire areas presented a continuous alteration in the size of the NPs. The electron beam spectroscopy results confirmed the presence of amorphous carbon and several Al-Si-K-Mg-O-C NPs that are considered as dangerous elements.

This study provides an overview of the occurrence of REEs and CNP in spontaneous coal combustion in La Guajira, Colombia. It also presents a directional dataset for further scientific studies for investigating the connections between the organo-metallic NPs and other ecological issues in numerous spontaneous coal combustion zones. In addition, studying diverse samples of spontaneous coal combustion from a specific coal mining area may be of significance in understanding the geological structures of spontaneous coal combustion in other coal zones in Colombia. This study can be considered as a "door-opener" for future REE and CNP extraction studies.

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Figure captions

Figure 1: Studied coal fire area.

Figure 2: Detected REEs microcrystals and general EDS.

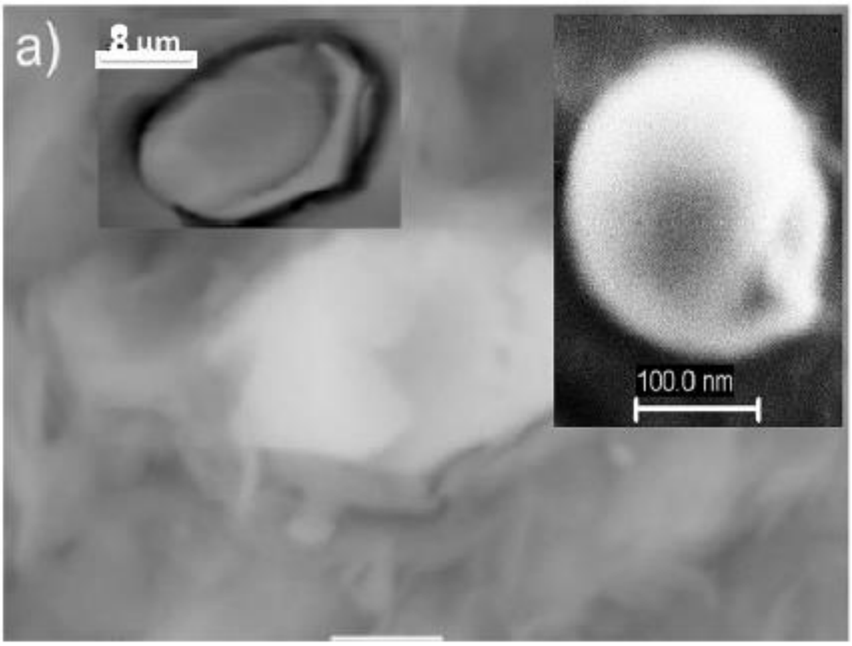


Figure 3: Carbon nanotubes and halogens association.

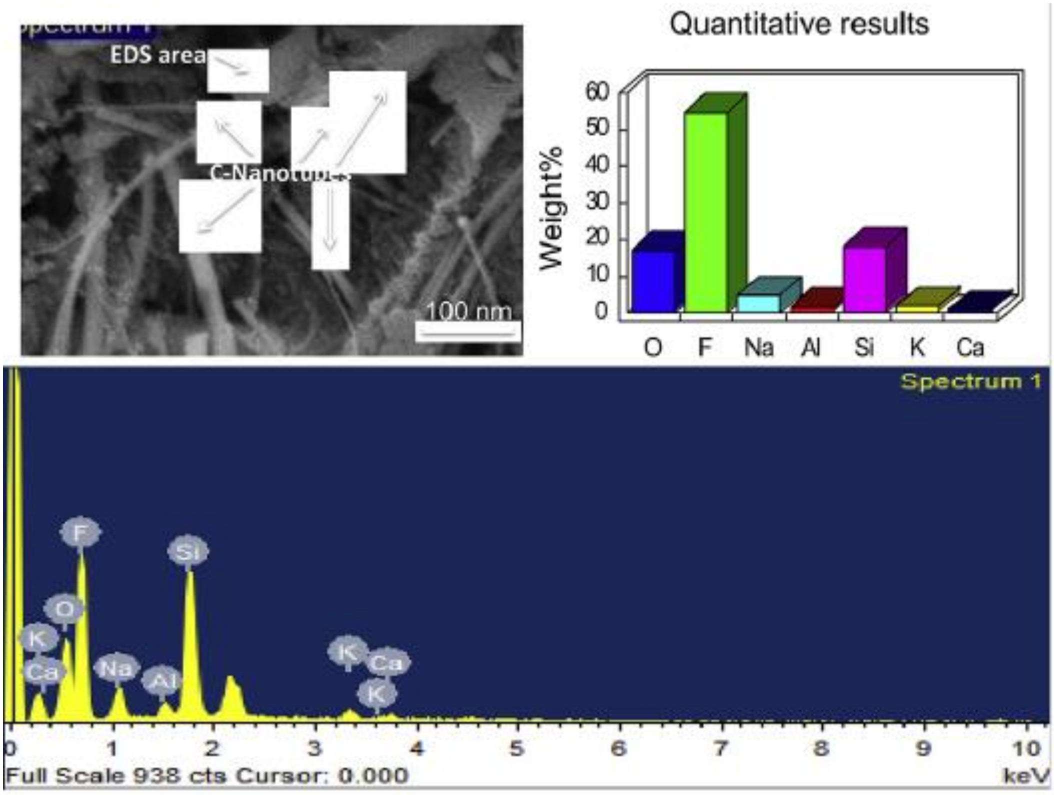


Figure 4: Complex organometallic nanoparticles.

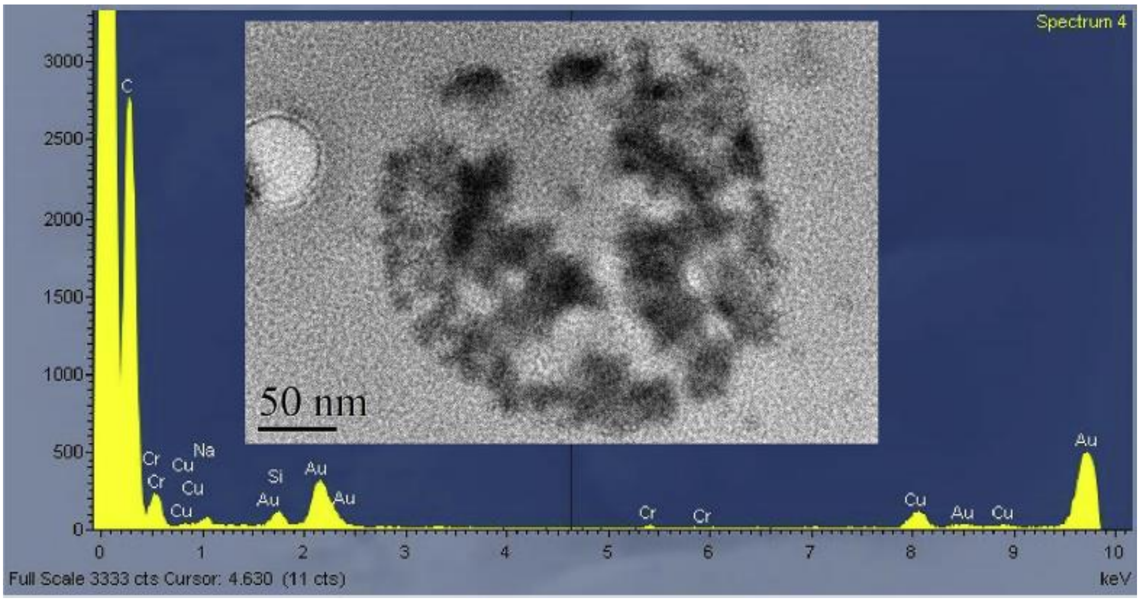


Figure 5: Carbon nanotubes with different crystallites.

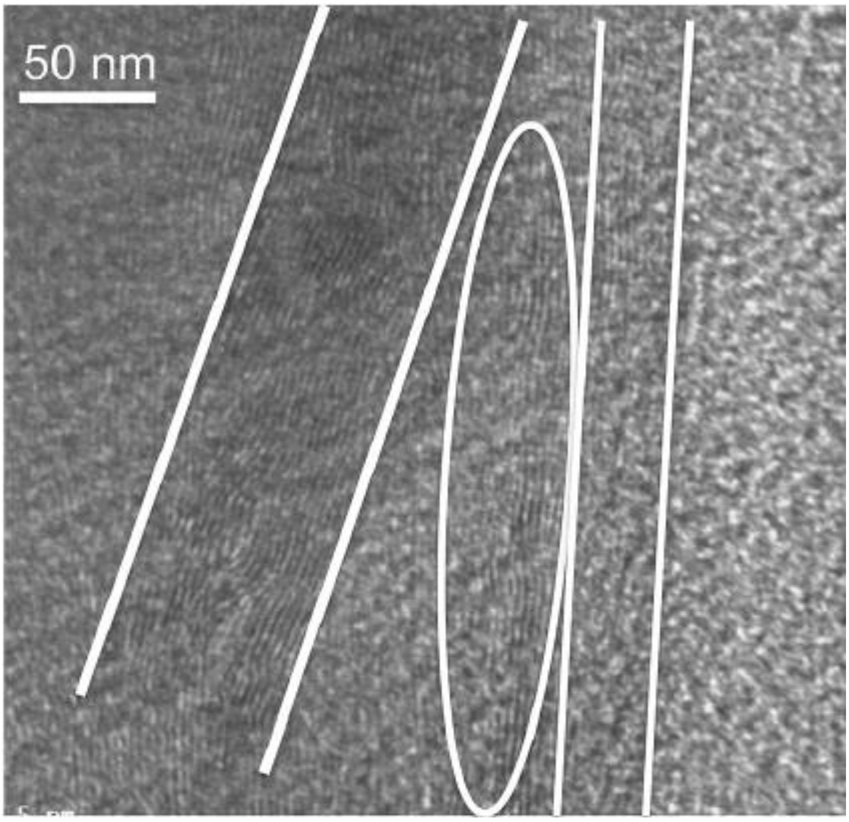
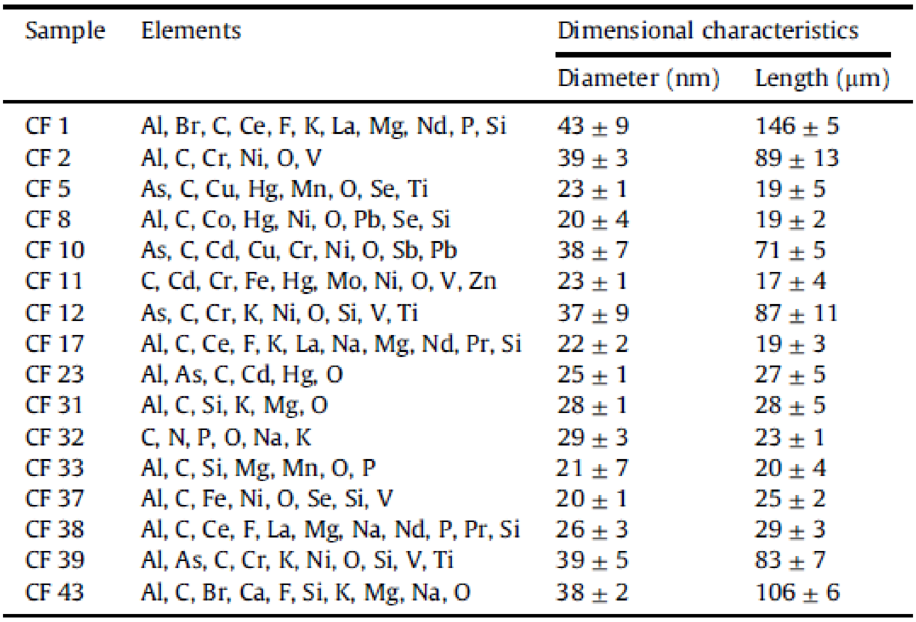


Table 01 - Dimensional characteristics of C-nanotubes and associated chemical elements present in sampled coal fire areas.



Sample	Elements	Dimensional characteristics	
		Diameter (nm)	Length (μm)
CF 1	Al, Br, C, Ce, F, K, La, Mg, Nd, P, Si	43 ± 9	146 ± 5
CF 2	Al, C, Cr, Ni, O, V	39 ± 3	89 ± 13
CF 5	As, C, Cu, Hg, Mn, O, Se, Ti	23 ± 1	19 ± 5
CF 8	Al, C, Co, Hg, Ni, O, Pb, Se, Si	20 ± 4	19 ± 2
CF 10	As, C, Cd, Cu, Cr, Ni, O, Sb, Pb	38 ± 7	71 ± 5
CF 11	C, Cd, Cr, Fe, Hg, Mo, Ni, O, V, Zn	23 ± 1	17 ± 4
CF 12	As, C, Cr, K, Ni, O, Si, V, Ti	37 ± 9	87 ± 11
CF 17	Al, C, Ce, F, K, La, Na, Mg, Nd, Pr, Si	22 ± 2	19 ± 3
CF 23	Al, As, C, Cd, Hg, O	25 ± 1	27 ± 5
CF 31	Al, C, Si, K, Mg, O	28 ± 1	28 ± 5
CF 32	C, N, P, O, Na, K	29 ± 3	23 ± 1
CF 33	Al, C, Si, Mg, Mn, O, P	21 ± 7	20 ± 4
CF 37	Al, C, Fe, Ni, O, Se, Si, V	20 ± 1	25 ± 2
CF 38	Al, C, Ce, F, La, Mg, Na, Nd, P, Pr, Si	26 ± 3	29 ± 3
CF 39	Al, As, C, Cr, K, Ni, O, Si, V, Ti	39 ± 5	83 ± 7
CF 43	Al, C, Br, Ca, F, Si, K, Mg, Na, O	38 ± 2	106 ± 6