



Historical Articles

## Astatine – The Elusive One

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**Abstract.** Astatine has proven, since its isolation by Corson, Mackenzie and Segre in 1940 to be an element with a fascinating history with respect to its discovery, confirmation and naming. It has also proven to have an interesting set of physical and chemical properties as well as isotopes of significant note. This element also has several applications of note as well as a captivating chemistry and the question whether it is or is not diatomic.

**Keywords.** Astatine, discovery/naming, properties of the element, applications of the element, chemistry of the element.

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### INTRODUCTION

Since 1922, a search for Element 85 [eka-iodine as per Dmitri Mendeleev's terminology] was underway involving the efforts of many scientists world-wide (1). Claims to the discovery [and confirmation] of the element were made by many individuals until the isolation of the element by Dale Corson, Kenneth MacKenzie and Emilio Segrè.<sup>1</sup> Significant discussion about the element's name also occurs in this paper. In addition, this paper also notes selected physical and chemical properties of astatine as well as its many known isotopes. An introduction to the applications of astatine follows and the chemistry of the element is then explored with a final examination made of whether or not astatine is diatomic.

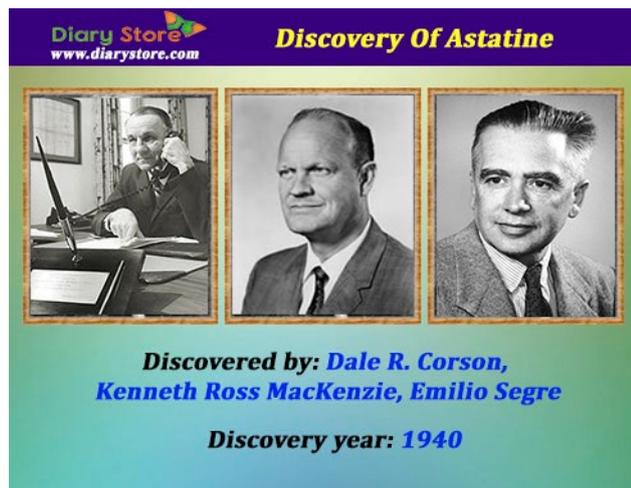
### DISCOVERY AND NAMING

#### *Early Efforts*

Reports of the search for element 85, termed eka-iodine by Mendeleev, go back to the year 1922.<sup>1</sup> In that year, Frederick Loring, working with numerical analysis, was led to believe that the element either did not exist or existed in very minute quantities.<sup>2,3</sup> A few years later, in X-ray experiments looking for element 87, he and his co-worker John Gerald Frederick Druce

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<sup>1</sup> A picture announcing the discovery of astatine is seen in Figure 1. This image is from [www.diarystore.com](http://www.diarystore.com).



**Figure 1.** An announcement of the discovery of the element astatine.

reported promising results suggesting the presence of both element 87 and element 85.<sup>4</sup> Many other individuals worked trying to isolate and characterize eka-iodine without noteworthy success during the 1920's and 1930's in the UK, Germany, British India, Denmark, France and Switzerland.

As the 1930's drew to a close, Horia Hulubei and Yvette Cauchois reported X-ray wavelengths for three spectral lines of eka-iodine in the emission spectra of radon that were closely related to Henry Moseley's predicted positions.<sup>1,5</sup> This was followed up by Manuel Valadares, who in the middle of World War II in Italy, repeated Hulubei's work with a large sample of Rn and saw new characteristic lines of element 85.<sup>6</sup>

In the time period 1942-1944, Berta Karlik and Traude Bernert noted the detection of  $\alpha$  particles and element 85 with a mass of 218. Karlik and Bernert were also involved in the discovery of naturally occurring astatine in 1942.<sup>6-13</sup> Even though these [and other] aforementioned scientists did stellar work, they were publishing their research in journals that were not well known. A good number of them also did not have available journal articles from the United States due to, in large part, World War II.

#### *Element 85 Discovery*

In 1939 with construction nearing completion on the Radiation Laboratory at the University of California at Berkeley, Segrè postulated that adding an  $\alpha$  particle at 32 MeV to Bi-209 would produce an element with atomic number 85.<sup>14</sup> This work was done by Robert Cornog and

Corson who, with the assistance of MacKenzie, saw a number of forms of radiation, including the emission of  $\alpha$ ,  $\gamma$ , and X-rays and also low energy electrons all having a half-life of about 7.5 hours.<sup>15-19</sup>

This group was also able to perform chemical analyses on the "material" and track its radioactivity. As a result, Corson, MacKenzie and Segrè, in large part, are regarded as the actual discoverers of Element 85. A great deal more is written about the search to find this element in an excellent article written by Brett Thornton and Shawn Burdette.<sup>1</sup>

#### *Confirmation of Discovery*

Even with the announcement of the discovery of astatine in 1940, confirmation was still needed. This was done by Joseph Hamilton and Mayo Soley who put a minimum amount of the new element [with a half-life of approximately 7.5 hours prepared according to the method of Corson, MacKenzie and Segrè] into the food of a guinea pig. Given that eka-iodine lay directly beneath iodine in the periodic table, it was logical to assume that this newly found element would have similar properties to those of iodine. After several hours of digestion, the iodine-craving thyroid gland of the guinea pig had filtered and concentrated this new element. Therefore, confirmation of the discovery of this new element was, in part, forever ascribed to the digestive system and thyroid gland of a guinea pig.<sup>20,21</sup>

#### *Naming the element*

From the initial tentative claim of the discovery of eka-iodine in 1925 by Loring to the actual discovery of the element in 1940 by Corson, MacKenzie and Segrè, a variety of names were proposed for this new element. In 1931, Fred Allison used a later discredited magneto-optic method to attempt to discover eka-iodine at the Alabama Polytechnic Institute [now known as Auburn University]. Using this method, he tested sea water, hydrohalic acids, apatite and Brazilian monazite sand; a material that was a source of rare earth minerals.<sup>22</sup>

In work reported in 1932, he suggested the name Alabamine (Ab later changing to Am) to honor the state where the work was done.<sup>22</sup> Allison also described, in his paper, chemical tests that unfortunately depended on the magneto-optic method. This method, and Allison's claim to have discovered Alabamine, were shown by Herbert MacPherson to be due to imperfections in Allison's equipment.<sup>23</sup> Though Alabamine was no more, its name and symbol stayed in textbooks and reference

works until 1947; in addition the name Alabamine, surprisingly and Allison, as its discoverer, were still listed in the 1991 *Concise Columbia Encyclopedia*.<sup>5</sup>

In 1937, an Indian chemist named De read about Allison's efforts and proceeded to study Travancore monazite sand which he believed to contain a significant amount of eka-iodine.<sup>1</sup> After performing a series of chemical tests on the monazite, De noted the presence of a black, sublimable substance that he identified as eka-iodine; he named it Dakin likely in honor of Dacca [now known as Dhaka, the capital of Bangladesh].<sup>24,25</sup>

Hulubei and Cauchois, in 1939, were examining the radioactivity of radon in the hope of seeing eka-iodine. In 1936 they had observed a line that corresponded with where the  $K_{\alpha 1}$  line for eka-iodine was expected. In 1939, they reported two further X-ray lines that were consistent with predictions from Moseley's law and the presence of eka-iodine and Hulubei announced the discovery of the element. This new data came about from experiments that used higher resolutions than their 1936 work and thus led to enhanced confidence that they had discovered eka-iodine.<sup>14</sup>

In 1944 Hulubei wrote up his work and that of others who had been looking for this new element. This summary included a description of six X-ray lines that were thought to be due to natural radioactive decay. He suggested, as a name, "dor" which was taken from Romanian word for "longing" and given that the world was at war and "longing for peace".

Unfortunately, there were many reasons why Hulubei and Cauchois did not receive due credit for their independent discovery of element 85. Reasons for this omission were: the statement of Karlik and others that Hulubei and Cauchois's sample size was too small and that there were interferences from other elements in the spectra of Hulubei and Cauchois; other work on Element 85 had been refuted even though Hulubei and Cauchois's work had not been refuted; wartime scientific communications were generally poor, Hulubei had mistakenly claimed discovery of element 87 and that their work was rejected by Paneth in 1947 who noted that the work of Hulubei and Cauchois did not have sufficient means to characterize discovery of element 85.<sup>1</sup> In addition, Hulubei's discovery only received experimental confirmation from the work of Valadares in Italy; the one and only country outside of France that did so confirm Hulubei's work.

An additional significant claim of discovery of element 85 came from the work of the Swiss physicist Walter Minder.<sup>26</sup> In 1940 he said he saw an extremely weak  $\beta$  decay of radium. In this work, a couple of ionization chambers were attached to an electrometer. Chemi-

cal tests confirmed, in Minder's opinion, a new element which he named helvetium with the symbol Hv. In 1942 Minder repeated his work and announced the "discovery" [with his British colleague Alice Leigh-Smith] of anglohelvetium. However, this work was not reproducible and discussion of it faded rapidly into obscurity.

As a result, credit for discovering eka-iodine went to the team of Corson, MacKenzie and Segrè and not to Hulubei and Cauchois. Now with the definitive discovery of element 85, a name had to be chosen. Given that this element is so rare, only having been discovered as naturally occurring by Karlik and Bernert in 1942,<sup>9,13</sup> and elusive as far as its discovery was so concerned, the Greek word *astatos* meaning unstable seemed appropriate.<sup>27</sup>

Element 85 now had a name - Astatine!

In closing this section, it is significant to note the total amount of naturally occurring astatine on planet Earth might be as little as a few hundred milligrams to, at a maximum, perhaps 30 grams.<sup>27</sup> This amount of astatine is solely due to the decay of thorium and uranium present in the Earth's crust.<sup>28</sup> Thus, astatine is the rarest naturally occurring element in the periodic table.<sup>20</sup>

#### DISCOVERY/CONFIRMATION OF ASTATINE AND SCIENTIST NATIONALITY

With any new scientific discovery, there is of course great joy, satisfaction and a sense of accomplishment. Though these feelings were likely true for Corson, MacKenzie and Segrè, the same was not true for others involved in the search for element 85. This story showed that nationalistic [and even regional as seen with the work of Allison] prejudices strongly influenced who was given credit for the discovery/confirmation of this member of the periodic table.

#### PROPERTIES

With the discovery, confirmation and naming done for the element astatine, characterization was next. Some of this work is reported in the NUBASE evaluation of nuclear and decay properties as compiled from archived information by Audi et al.<sup>29</sup> Work was reported on mass excess, half-life, probability of alpha decay and electron capture of the element. In addition, reports on astatine in nature came from the efforts of Karlik and Bernert who studied the  $\alpha$ -activity from the thorium decay series<sup>9,13</sup> and also found a small number of previously unreported 8.4 MeV  $\alpha$ -particles related to a short-lived

At-215 isotope.<sup>11</sup> One other significant work on naturally occurring astatine showed that At-219 was present in U-235 ores in minute amounts.<sup>30,31</sup>

Astatine is quite difficult to work with – if one were to have a visible piece of the element, the element would immediately be vaporized due to heat generated by radioactive decay. It has not yet been determined if this issue in exploring the element can be overcome by appropriate cooling.<sup>32</sup> As a result, most physical properties of astatine are not known with any degree of certainty. It is also interesting that astatine may only be found in uranium 235 ores, nuclear facilities and/or research laboratories. Otherwise, the element is undetectable.

### Physical Properties

Some of the physical properties of astatine are seen in Table 1.<sup>33,34</sup> Based on the colors of the halogen group elements it is likely that a hypothetical solid astatine may be very dark in color, perhaps even black.<sup>32</sup> Of all the natural halogens, astatine is the least chemically reactive and may even exhibit metallic properties – perhaps even being a superconductor.<sup>35,36</sup> A good number of these tabulated properties are theoretical.

**Table 1.** Selected Physical Properties of Astatine.

Atomic number	85
Atomic mass	~210 grams/mole
State at 20°C	Solid (predicted)
Density (estimated)	6.35 – 6.50g/cm <sup>3</sup>
Boiling point	230-233°C (estimation)
Heat of fusion	~ 6 kJ/mol (estimation)
Electron configuration	[Xe] 4f <sup>14</sup> 5d <sup>10</sup> 6s <sup>2</sup> 6p <sup>5</sup>
Covalent radius	150 pm
Van Der Waals radius	202 pm
Crystal structure	fcc (predicted)

### Chemical Properties

Astatine has many possible oxidation states [-1, 1, 3, 5 and 7] with -1 and 1 being most common.<sup>33</sup> Only francium is less stable than astatine out of the first 101 elements in the periodic table. When astatine undergoes radioactive decay, the element decays into Bi, Po, Rn or other isotopes of astatine. Astatine has an electronegativity of 2.2 [on the revised Pauling scale] and a first ionization energy of 899 kJ/mol. The chemistry of astatine, difficult due to its short half-life, is “clouded by the extremely low concentrations at which astatine experiments have been conducted, and the possibility of reac-

tions with impurities, walls and filters, or radioactivity by-products, and other unwanted nano-scale interactions”.<sup>35</sup>

Of interest also is the fact that astatine is the least reactive of the halogens. In order to ensure a reaction, dilute solutions of astatine are mixed together with larger amounts of iodine. Iodine, acting as a carrier, ensures that there is sufficient material for such techniques as filtration and precipitation to be properly conducted.<sup>37-39</sup>

## ISOTOPES

Currently, astatine has 39 known isotopes with atomic masses ranging from 191 to 229. According to theoretical modeling, another 37 isotopes might exist.<sup>30,40</sup> Out of all these possible isotopes, not one is likely to be stable or even long-lived. The most stable isotope, known to date, is At-210 and it has a half-life of 8.1 hours. This isotope has as its primary decay mode  $\beta^+$  which gives the  $\alpha$  emitter Po-210.

Of all the possible isotopes, only five have half-lives greater than one hour and they are: At-207 (1.80 hr), At-208 (1.63 hr), At-209 (5.41 hr), At-210 (8.1 hr) and At-211 (7.21 hr). The shortest lived isotope found to date is At-213 has a half-life of 125 nanoseconds (29); this isotope undergoes  $\alpha$  decay to the extremely long-lived Bi-209 (half-life of  $1.9 \times 10^{19}$  yr).

## INTRODUCTION TO APPLICATIONS OF ASTATINE

With the discovery and initial characterization efforts on astatine somewhat complete, significant interest turned to potential applications of the element. Given astatine’s similar behavior to iodine in concentrating in the thyroid gland as found by Hamilton and Soley,<sup>21</sup> medical applications of the element and its isotopes became interesting. Later work showed that fairly small amounts of astatine could lead to significant changes of structure in the parathyroid gland and other peritracheal tissues.<sup>41</sup>

Further investigation of this take-up of astatine by bodily tissues was then made in guinea pigs, rats and monkeys; in fact the treatment of hyperthyroidism in human beings by astatine was proposed. This was substantiated by the results of tracer experiments where it was found that human beings accumulated astatine in their thyroid glands. Benefits of this “tendency” were noted to have to be weighed against hazards from exposure to the radiation of astatine.

### Actual Applications of Astatine

With the half-life of even the longest-lived astatine isotope only about 8 hours, finding any real world applications have been quite difficult, to say the least. At-210 decays quite readily into Po-210 which is notorious; it was used to kill Alexander Litvinenko in November 2006.<sup>42</sup> Fortunately though, At-211 is an isotope that has seen some use in nuclear medicine and may also be helpful in the future.

Much like I-131, At-211 does not emit high-energy  $\beta$ -particles, but instead emits  $\alpha$  particles and it is known as one of the few  $\alpha$ -emitting radionuclides considered for medical use;<sup>43</sup> most other  $\alpha$ -emitters cause severe damage to internal organs. A typical  $\alpha$ -particle released by At-211 may travel only 70 mm through bodily tissues; an average  $\beta$ -particle emitted by I-131 can travel more than 25 times farther in bodily tissues.<sup>44</sup> Given this short path length of  $\alpha$ -particles in the human body and their high energy (6.0 to 7.5 MeV), such particles are very effective at killing cells bound by carrier-targeting agents.<sup>45</sup>

This short range effectiveness of At-211 can possibly reduce risk of exposure to other bodily tissues and perhaps then be used throughout the human body in treating cancer. This short half-life of At-211 and lesser penetrating ability is a definite advantage in situations where the “tumor burden is low and/or malignant cell populations are located in close proximity to essential normal tissues”.<sup>46</sup> Significant morbidity in cell culture models of cancers in humans have been achieved with from one to ten At-211 atoms bound per cell.<sup>47</sup>

There is though a problem with the low stability of astatine to aromatic carbon bonds *in vivo*.<sup>48</sup> Developing labelling reagents with more stable aromatic astatine-boron bonds has helped in lessening this difficulty and it is possible that further study with other elements may lead to additional scientific advances.

Astatine-based radiopharmaceuticals have seen a variety of obstacles during the 20<sup>th</sup> and 21<sup>st</sup> centuries. World War II brought such research to a halt for at least a decade. Experiments noted that a cancer-selective carrier would need to be developed and it was not until the 1970's that monoclonal antibodies became available for this purpose. Given that astatine tends to become detached from its compounds, is toxic and is also retained in the body [preferentially accumulating in the thyroid gland, lungs and spleen].<sup>47,48</sup> At needs to stay attached to its host molecule. Mitigating the effects of astatine induced radiolysis of labelling chemistry and carrier molecules is another area needing additional work.

### CHEMISTRY OF THE ELEMENT

The chemistry of astatine is typically done using masses of  $10^{-13}$  to  $10^{-8}$  grams of the element<sup>49</sup> or at extremely low concentrations. Unfortunately, this low-level method allows reactions with impurities, walls, filters and even radioactive byproducts.<sup>35</sup> A good number of astatine's properties have been observed in very dilute solutions typically at concentrations of less than  $1 \times 10^{-10}$  mol/L. Even though “Astatine ... [is] miserable to make and hell to work with,” as noted by Patricia Wallace Durbin,<sup>50</sup> it does undergo a variety of chemical reactions. The element has the ability to form compounds with metals; combine with hydrogen; interact with boron and carbon; react with oxygen and other chalcogens and also form compounds with chlorine, bromine and iodine.

Though there are only a few compounds of astatine with metals, astatides incorporating sodium, magnesium, palladium, silver, thallium and lead have been formed.<sup>51,52</sup> Given the extraordinarily limited amount of available astatine, estimations, by extrapolation, of the characteristics of AgAt and NaAt have been made based on other metal halides.<sup>53</sup>

Astatine also exhibits chemistry with boron and carbon. A variety of boron cage compounds have been made with At-B bonds stronger than At-C bonds.<sup>51,54</sup> The compounds carbon tetraastatide (CA<sub>4</sub>) and astatobenzene (C<sub>6</sub>H<sub>5</sub>At) have been prepared.<sup>51</sup> Astatobenzene (C<sub>6</sub>H<sub>5</sub>At) can also be oxidized to C<sub>6</sub>H<sub>5</sub>AtCl<sub>2</sub> by exposure to Cl<sub>2</sub>. This chlorinated compound can then be treated with a basic hypochlorite solution to give C<sub>6</sub>H<sub>5</sub>AtO<sub>2</sub>.<sup>55</sup> A perchlorate of astatine has also been prepared, [At (C<sub>5</sub>H<sub>5</sub>N)<sub>2</sub>] [ClO<sub>4</sub>] where the astatine is bonded to each nitrogen atom in the two rings of pyridine.<sup>56</sup>

Reactions of astatine also occur with oxygen and other chalcogens. With oxygen, there is evidence to support the existence of AtO<sup>•</sup>, AtO<sub>2</sub><sup>-</sup> and AtO<sup>+</sup> in aqueous solution.<sup>57,58</sup> An AtO<sub>3</sub><sup>-</sup> ion can be obtained by the oxidation of astatine with KOCl in a solution of KOH.<sup>59,60</sup> Further chemistry of this AtO<sub>3</sub><sup>-</sup> species has seen the formation of La(AtO<sub>3</sub>)<sub>3</sub> through the oxidation of astatine in a hot Na<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (sodium peroxodisulfate) solution.<sup>44</sup> The AtO<sub>4</sub><sup>-</sup> ion also exists; further oxidation of AtO<sub>3</sub><sup>-</sup> in a hot, basic solution of XeF<sub>2</sub> so gives this anion.<sup>61</sup> Substituting periodate (in either neutral or basic media) will also yield the AtO<sub>4</sub><sup>-</sup> ion – which is only stable in neutral or basic media.<sup>62,63</sup>

Astatine can also form, with sulfur, the S<sub>7</sub>At<sup>+</sup> ion and the thiocyanate ion species At(CSN)<sub>2</sub><sup>-</sup>.<sup>64</sup> In addition, cationic astatine compounds with thiourea, thiourea derivatives and some N-acyl thioureas in aqueous solu-

tion have been reported.<sup>64</sup> Astatine also has the ability to form a selenourea coordination compound with selenium and a colloidal tellurium compound.<sup>64</sup>

Reactions of astatine in the vapor state with chlorine, bromine and iodine have produced the diatomic compounds AtCl, AtBr and AtI.<sup>65</sup> AtBr is also formed by interacting astatine with an iodine/iodine monobromide/bromide solution whereas AtI is prepared by reacting astatine with iodine/iodide solution in an aqueous media.<sup>66</sup> Excess of iodide or bromide could lead to the formation of  $\text{AtBr}_2^-$  and/or  $\text{AtI}_2^-$  ions or if in a chloride solution, species such as  $\text{AtBrCl}^-$  or  $\text{AtCl}_2^-$  can come about via equilibrium reactions with the chlorides.<sup>67</sup> Oxidation of At with  $\text{Cr}_2\text{O}_7^{2-}$  (while in a  $\text{HNO}_3$  solution) showed that when adding chloride a compound of either AtCl or AtOCl was produced; similarly, the ions  $\text{AtOCl}_2^-$  or  $\text{AtCl}_2^-$  may also be produced.<sup>65</sup> Polyhalides like  $\text{PdAtI}_2$ ,  $\text{CsAtI}_2$ ,  $\text{TlAtI}_2$  are directly known or have been presumed to be precipitated in chemical reactions.<sup>68-70</sup> The existence of  $\text{PbAtI}$  is also likely.<sup>71</sup> The possibility of the  $\text{IAtBr}^-$  ion, as determined by quantum mechanical calculations, was found by Galland to exist and even predominate in aqueous solutions.<sup>72</sup>

#### ASTATINE - DIATOMIC OR NOT?

All serious students of chemistry know the seven diatomic elements. Whether astatine is also diatomic is a matter of debate to this date. As an analog of iodine, it may have an orthorhombic crystalline structure composed of diatomic astatine molecules and also be a semiconductor.<sup>73</sup> Unfortunately, the structure of solid astatine is not known though some expect it to be a black solid with a metallic appearance.<sup>34,74</sup>

Takahashi, in work reported in 1986, described interactions of astatine with benzene, toluene and monochlorobenzene. In this study, the chemical bond between  $\text{At}_2$  is quoted to cleave in a two-step mechanism that worked for benzene and toluene but not for monochlorobenzene.<sup>75</sup> Additional work by Takahashi reported in 1991 looked deeper at the chemical behavior of astatine.<sup>76</sup> On the other hand, condensed astatine is calculated to behave like a metal at 1 atmosphere of pressure and might possibly be a superconductor but would be monoatomic.<sup>77</sup>

Simply enough, in the gas phase, astatine might be weakly diatomic but when it is solid it is the spin orbit coupling contribution of relativistic effects that weakens the covalent character of a bond in  $\text{At}_2$  and thus makes solid astatine monoatomic.<sup>77,78</sup> This also extends to the ability of At to be a halogen bond donor.<sup>79</sup>

#### CONCLUSION

The discovery [and confirmation of the existence] of the element astatine involved the efforts of many scientists and many years of work in Europe, India and the United States. These investigations gave the world the properties of astatine and its chemistry as well as some isolated but medically important applications of the element. The question of whether  $\text{At}_2$  exists was explored. All in all, element 85 has shown that even though it has an unstable nature, it is still a fascinating and vital member of the periodic table.

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