

THEORETICAL PREDICTIONS OF THE CHEMISTRY OF SUPERHEAVY ELEMENTS

Continuation of the Periodic Table up to $Z=184^*$

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I. Introduction

When Mendeleef set up the Periodic System a hundred years ago, only 63 elements were known. With the help of the systematics of this Table, predictions of the chemical behavior of the unknown elements were made, and indeed, in the next 30 years most of them were found. With the exemption of

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a few radioactive elements, the chemist lived for a long period with a somewhat closed system of known elements.

Even this did not change when Seaborg and his collaborators¹ produced in the last 30 years most of the Transuranium elements. Until now the elements up to $Z = 105$ are discovered. From a chemical point of view, the elements $Z = 90$ to $Z = 103$ form the actinide series in a way as it already was suggested by Seaborg in 1945. The properties of these $5f$ transition elements has been investigated in the last 20 years and the results are summarized in recent reviews by Seaborg^{1,2}, Cunningham³, and Asprey and Penneman.⁴ A most comprehensive review for the elements 101 to 105, the nuclear stability and chemical behavior of the forthcoming elements with respect to the year 1968 is given by Seaborg.⁵

In this article we summarize the research which has been done in the field of the prediction of the chemical behavior of the yet unknown elements beyond 105.

II. Predictions of Nuclear Stability and the Search for Superheavy Elements

The halflives of the Transuranium elements decrease rapidly with higher Z and, therefore, it was expected that there might be a limit in the detection of heavier elements which is mainly given by the time which is needed to detect them.

Recent developments in the calculations of nuclear stability suggest that in contrast to this behavior, nuclei with much higher proton numbers might exist with halflives up to many years.⁶⁻⁸ A region of relative large stability is expected between elements $Z = 110$ and 120 (which is called the first island of superheavy elements) and possibly a second near $Z = 164$ and/or near $Z = 184$ is predicted.* Especially two groups presented detailed data with

* Most of this research was summarized by Seaborg.⁵ A more recent summary of theoretical predictions for superheavy nuclei is given by Nix.⁹

predictions of halflives of superheavy nuclei against spontaneous fission and α -decay. In Fig. 1 the results of Nilsson et al⁶ for the first region of superheavy nuclei are shown. Very similar results are given by Grumann et al.⁸ In Fig. 2 their predictions are given for the second quasistable island near $Z = 164$. In both regions α -decay and fission will be mostly dominant so that the observation of α particles as well as the fission products will be the main method to detect these elements. In both regions halflives against β -decay and electron capture have been calculated as well but they lead only to small changes within these results.⁸

If these predictions are true (within a factor of $10^{\pm 6}$) we are again in the situation that only a part of the Periodic Table is known. Again the questions concerning their existence, their chemical behavior and, if possible, their location in the Periodic Table arise.

Because of the predicted long lifetimes especially in the first region of superheavy nuclei, many groups tried to find these elements in small amounts in nature. A summary of this research is given by Seaborg¹⁰ and by Hermann and Seyb.¹¹ Some reports^{12,13} are quite optimistic but up to now no real evidence for elements with $Z > 105$ is given. So it seems that these elements have to be synthesized by using appropriate accelerators to bombard target elements of relatively high atomic number with beams of heavy ions. Hopefully, superheavy nuclei will be produced by a direct fission process, a fission-fusion process or by a transfer process of a heavy particle in heavy ion scattering. Many of those heavy ion accelerators are under construction in the world but none of them has started yet with experiments. Therefore, recently a report of Marinov et al¹³ about the existence of $Z = 112$ was most interesting. They utilized a very high energetic proton-beam to create such highly energetic tungsten atoms by elastic scattering that these possibly could have fissioned to superheavy elements. After chemical separation, they proved the existence of a new element in observing an unknown α -line as

well as fission fragments. Their argument that $Z = 112$ was created is based on the prediction that this element would be chemically very similar to mercury.

Other experimental proofs of the existence of superheavy elements could be done by observation of their x-ray spectra, mass spectroscopy, optical spectroscopy or even by pure chemical methods.

Under these aspects the knowledge of the chemistry of superheavy elements is not only interesting but most important.

III. Physical Basis for Predicting the Chemical Behavior of Superheavy Elements

The chemical property of an element depends strongly on the outer electronic configuration and the filled electron shell concept of quantum mechanics gives rise to the periodicity of chemical properties that forms the basis for the concept of the Periodic Table.

With this experimentally found result, the first predictions of chemical properties of unknown elements were made and the success was great. From a more modern standpoint of view, this method worked so well because we now know that the quantum mechanics of the outer electrons in the first part of the Periodic System is very well described by the nonrelativistic theory with LS coupling and large energetic gaps between the different shells. Also the influence of the inner electron shells is rather small so that the outer electron configurations are very similar in the same chemical group but different periods. Nevertheless, one should not forget that there are differences even between two elements from the same chemical group. These differences can be explained mostly as the effect of slightly different size, changed ionization potential and different energies between analogous shells. If we come to higher elements at the end of the Periodic Table, the number of shells increases, the binding energy of the last electron decreases, a competition between different shells takes place and the influence of the inner electrons

becomes more significant. This rather complex behavior becomes even more complicated because relativistic effects begin to be important and the coupling between angular momenta of the electrons changes character to intermediate or even j-j coupling.

The best example for this complex structure and its connection to the chemical behavior is the fact that in lanthanum, a d-electronic state is occupied before the 4f shell starts to fill. But although the atomic configuration of most other lanthanides is pure $4f^n 6s^2$, they form mainly trivalent ions because in the +2 ionization state, the configuration is $4f^{n-1} 5d$ and the 5d electron can readily be oxidized.

The main change due to relativistic effects is the splitting of all shells with $l \neq 0$ into two subshells with $j = l + 1/2$ and $j = l - 1/2$. This means that e.g. the p electrons split into the $p_{1/2}$ subshell with 2 electrons and the $p_{3/2}$ subshell with 4 electrons. How large this effect will be can be seen in Fig. 3 where the energy eigenvalues of the $p_{1/2}$ and $p_{3/2}$ shell as a function of Z for the series of group IV elements is plotted. In general, spin orbit splitting increases with approximately the fourth power of Z. The spin-orbital stabilization of the $np_{1/2}$ states plays a dominant role in Lawrencium and elements beyond. This altogether means that a quantum mechanical calculation using a relativistic Hamiltonian becomes indispensable to get credible predictions of the chemical behavior of superheavy elements.

Unfortunately, it is impossible up to now to perform exact calculations of chemical compounds, even really exact calculations of the n-body problem of an atom or ion are not possible. Nevertheless, the more or less approximate calculations which are possible at the moment, lead to quite accurate predictions of most stable electron configurations for atoms and ions as well as ionization energies, and binding energies together with their wavefunctions. Those methods can be summarized under the general name of Hartree-Fock calculations with different approximations made for convenience. In Table 1 the

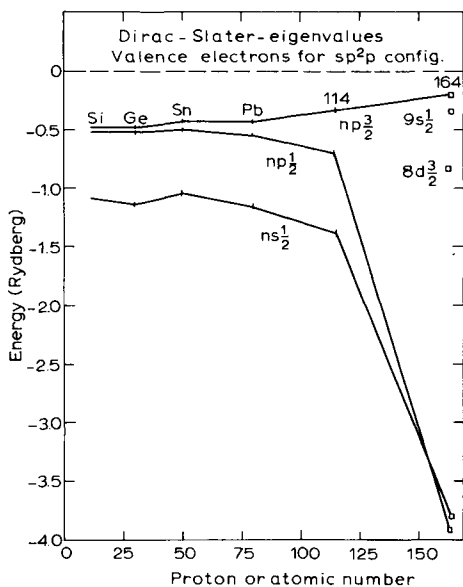


Fig. 3 Comparison of Dirac-Slater eigenvalues for IV A elements. This illustrates the very strong dependence on atomic number of the spin-orbit splitting between the $np_{1/2}$ and $np_{3/2}$ states. Levels for $8d_{3/2}$ and $9s_{1/2}$ are also indicated for element E164.

Tab. 1 The usual abbreviations and their explanation for the most common atomic calculations.

abbreviation	explanation
Hartree	non relativistic without exchange
Hartree-Fock-Slater (HFS)	non relativistic with Slaters exchange approximation
Hartree-Fock (HF)	non relativistic with exact exchange
Dirac-Slater (DS) or relativistic HFS	relativistic with Slaters exchange approximation
Dirac-Fock (DF)	relativistic with exact exchange

mostly used abbreviations for the different methods are summarized. The best summary and critical analysis of the present status of the relativistic methods is given by Grant.¹⁴

For light elements it is sufficient to solve in these procedures the nonrelativistic one-electron Schrödinger equation and assume the central field arises for a point nucleus. In the relativistic treatments, the single particle Dirac equation has to be solved to get the single particle energies and wavefunctions of all electrons in the central field of an extended nucleus as well as in the presence of the (N-1) electrons. The main difficulty arises by taking into account the so-called non-local exchange potential which is distinct for each orbital and, therefore, in the past, most calculations were done with an exchange approximation proposed by Slater.¹⁵ From the sum of the squares of the wavefunctions, which is the electron density, a new potential can be generated by use of the Poisson equation and from there, the wave equations can be solved again. When the new potential becomes equal to the (old) potential from the last iteration, the problem is said to be solved in a self-consistent way.

Because in the region of superheavy elements, the relativistic effects become large, nearly all calculations are done by solving the Dirac equation. The analytical solution of the Dirac equation for the energy eigenvalues of the electrons in the field of a point nucleus leads to nonphysical imaginary results for the $1s_{1/2}$ and $2p_{1/2}$ levels for $Z \cdot \alpha > 1$, where $\alpha = (137.038)^{-1}$ is the finestructure constant. This difficulty can be circumvented by taking into account the extended nucleus. Nevertheless this difficulty is shifted by this method only near to elements $Z = 175$ but does not disappear. Therefore, up until quite recently no calculation has been made to date for elements beyond.

Recently several discussions^{14,16} have appeared about how to estimate

ionization potentials IP and the accuracy of Koopman's theorem.¹⁷ Mostly one uses the adiabatic approximation in which it is assumed that sufficient time elapses during the removal of an electron for the other electrons to rearrange so that IP is given by the difference in total energy of the atomic species with N and with N-1 electrons.

There are as well some other effects which become non-negligible in real atoms with large atomic number which are only partly taken into account in the calculations; these are the quantum electrodynamical effects¹⁸ (vacuum polarization and vacuum fluctuation¹⁹) magnetic effects²⁰, correlation and retardation.²⁰ All these effects and their influence in the calculations are presently under investigation for heavy elements. So far, how large these effects are is not really known or whether possibly other unknown effects become important in the region where $Z\alpha > 1$. These open problems and their possible influence on the chemical behavior will be discussed shortly in section VI.

Nevertheless, if all the calculations are done carefully very little can be said about the chemical behavior of these unknown elements. Because there are only a few quantities which can be calculated directly, all other predictions can be done only in a combination of the very traditional (old) method of continuation of the trends of the interesting physical and chemical quantities in a chemical group into the unknown region with a comparison of the trend shown in the calculations, if this is possible. On the other hand, the calculations have to be checked in the known region of elements and possible systematical deviations determined from experiment will lead then to even better predictions of the interesting physical and chemical properties of superheavy elements.

IV. Chemical Basis for Predicting Chemical Behavior

The recent classification of ions with hard and soft Lewis acids by

Pearson²¹ has brought considerable order into inorganic chemistry. While a specific definition or scale of softness is not universally accepted, the general principles are clear. They can be used to predict a little more about what compounds or complex ions might be expected for the superheavy elements.

Hard Lewis acids are found among the small, highly charged ions such as Al^{+3} or La^{+3} which have low lying orbitals available for occupation - in general, these ions are not readily polarized. Soft Lewis acids tend to be large, easily polarized ions such as Ag^{+1} and frequently the state of ionization is low. In addition, they contain unshared pairs of electrons such as p or d electrons in their valence shell. The hard Lewis bases are ions such as F^- or OH^- which have small ionic radii and are characterized by high electronegativity, low polarizability and are difficult to oxidize. In comparison to these ions, I^- is more polarizable, acts as an electron donor and is, therefore, a softer Lewis base.

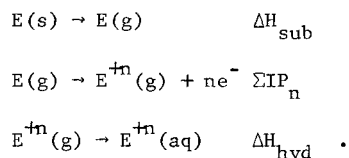
Pearson's Hard-Soft-Acid-Base (HSAB) principle is that hard acid-base combinations form readily and generally are ionic compounds. The other group of stable compounds and complex ions involves the interaction between soft acid and soft bases. For these, the bonding is primarily covalent with interpenetrating orbitals. The combination of hard acid to a soft base, or vice versa, have little stability.

Klopman²² defined the frontier orbitals for a base as the highest occupied orbitals of the donor atom or ions and as lowest unoccupied orbitals of the acid or acceptor ion. For elements with low atomic numbers, such orbitals are radially extended, such as the 3d orbitals. With increasing atomic number this feature is not guaranteed and orbitals which are not tightly bound may become buried beneath other competitive orbitals. The essential idea is that the frontier orbitals must overlap significantly in order that covalent bonding can occur in the soft-soft interactions.

The difference in the chemistry of the light and heavy actinides may be rationalized in this way. The early numbers beyond Thorium have unpaired d and f electrons available for forming covalent bonds and hence, for example, they readily form many complex ions and intermetallic compounds. Such ions are soft acids. Beyond Americum, the $5f$ electrons are not competitive and the closed shell of six ($5f\ 5/2$) electrons will not be readily available for bonding and only those f electrons with $j = 7/2$ are available. These tend to become buried radially as the atomic number increases and hence the divalent ions become relatively hard Lewis acids.

The intrinsic strength factors of any set of ions might be deduced from consideration of energy eigenvalues for gaseous species such as the free atoms and ions calculated in extenso by the groups at Los Alamos and Oak Ridge. However, strong solvent corrections must be made before any substantial progress can be made.

Water is a hard solvent with respect to both acids and bases; hydrated ions will be soluble in water if the desolvated ion is itself hard. Solvation of cations is of primary importance in determining an Electromotive Series. The value of the standard electrode potential can be estimated from the three partial reactions for an arbitrary element



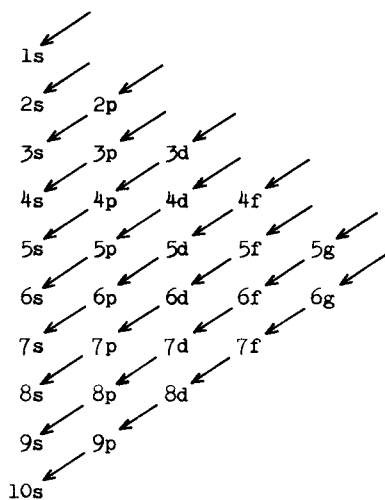
The heat of sublimation ΔH_{sub} and the sum of the several ionization potentials IP_n require the expenditure of energy and this is offset by heat of hydration ΔH_{hyd} . In general, soft metal ions will have low hydration energy and if ΔH_{hyd} is small, the metal will be unreactive and have low electrode potentials. Again, this argument might be applied to uranium

ions. While species like U^{+4} will be hydrated, because of their large ionic charge, if Cl^- ions are added to solution, they will displace the stronger water molecules and species like UCl_x^{++} will be found in solution. However, the tendency to form highly hydrated or ionic complexes will be greatly reduced for the higher actinide species. Two analytical expressions which measure the reactivity of ions with respect to water were given by Jørgensen.²³

V. Discussion of the Elements

The easiest and simplest overall prediction of the chemistry of super-heavy elements is just the normal continuation of the Periodic Table towards the very old systematics of the filling of the shells as it is shown in Table 2. According to this basis of prediction the Periodic System, with its continuation up to $Z = 168$, is shown in Fig. 4. This was given by

Tab. 2 Diagram showing the sequence of the filling of the electron shells in the Periodic Table. It is nearly correct, up until very far into the region of unknown superheavy elements. For comparison see Fig.4 and Fig.14.



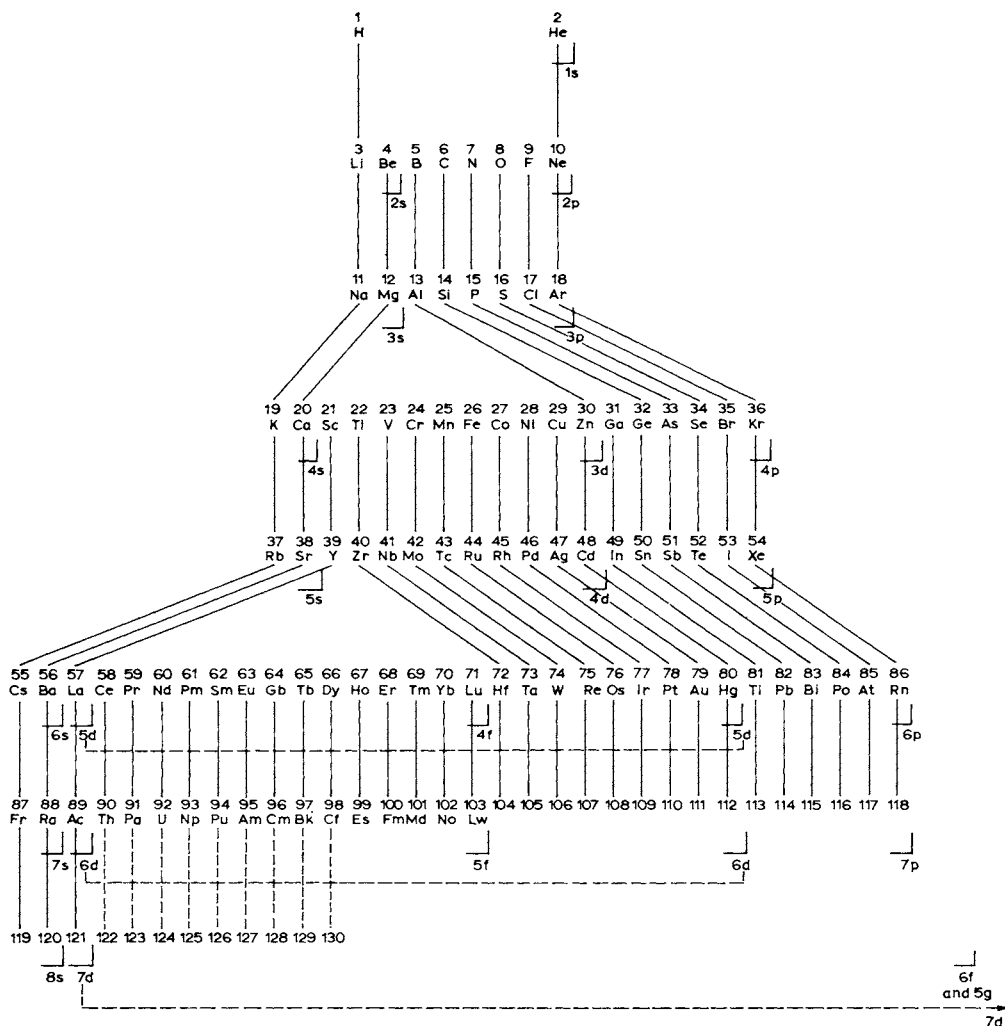


Fig. 4 Modified form of the Periodic Table and its continuation up to element E168 according to Seaborg⁵.

Seaborg.⁵ Because the elements beyond Lawrencium have not yet been assigned up to now no recognized chemical abbreviations we will use this simple nomenclature, eg E118, in the following sections when we speak of the element with 118 protons. In Table 3 all references are summarized in which

Tab. 3 Nearly complete list of references in which predictions of the chemistry of elements beyond E103 are given either by continuation of the trends of the elements or from calculations with chemical statements.

Authors	Year	Elements	Method	Remarks
A.V. Grosse ²⁴	1965	118	continuation	
B.B. Cunningham ²⁵	1968	104, 105, 117-120	continuation	
O.L. Keller et al. ²⁶	1968	104, 105	continuation	
A.C. Larson et al. ²⁷	1968	122-127	Hartree calculations plus exchange corrections	
C.K. Jørgensen ²⁸	1968	126		only one comment about a possible 126(IV) oxidation state
T.C. Tucker et al. ²⁹	1968	114, 126, 140	Dirac-Slater	electron binding energies only
G.T. Seaborg ⁵	1968	104-128		summary and discussion
C.K. Jørgensen et al. ³⁰	1969	114		discussion only
T.C. Tucker et al. ³⁰	1968	114, 126, 140	Dirac-Slater	electron binding energies only
D.C. Griffin et al. ³¹	1969	121-126	Hartree plus statistical exchange plus relativ. corrections	most stable configuration calculation only
J.T. Waber et al. ³²	1969	104-132	Dirac-Slater	
J.B. Mann ³³	1969	121-127	Dirac-Fock	most stable config. calc. only
B. Fricke et al. ³⁴	1969	160-170	Dirac-Slater	
J.T. Waber ³⁵	1969	104-131	Dirac-Slater Dirac-Fock	
J.B. Mann ³⁶	1969	121-172	Dirac-Fock	
B.B. Cunningham ³⁷	1969	104-112	continuation	
O.L. Keller et al. ³⁸	1970	113, 114	continuation and some calculations	
J.B. Mann et al. ³⁹	1970	118-131	Dirac-Fock	
R.A. Penneman et al. ⁴⁰	1971	164(184)	Dirac-Fock	184 no calculation
B. Fricke et al. ⁴¹	1971	104-172	Dirac-Slater	

predictions of the chemistry of elements with $Z > 103$ are given either by continuation of the trends of the elements or from calculations with chemical statements.²⁴⁻⁴¹

a. The 6d Transition Series E104 to E112

The experiments which proved that E104 is the chemical analogue to Hafnium and Zirconium⁴² and E105 behaves like Tantalum^{43,44} are the first indications that the elements E104 to E112 are the transition elements of the 7th period in which the 6d and 7s electrons determine the chemistry.

i. Electron Configuration

The electron configurations of the outer electrons of almost all the d-transition elements are given by the rule $(n-1)d^m ns^2$ where n is the number of the period (or the principal quantum number) and m goes from 1 to 10. This rule is not exactly valid in the 4th and 5th period where in some cases only one or even no electron occur in the outer s-shell. This behavior is well understood; both the increasing binding energy of the d-electrons and the greater shielding of the s-electrons with higher Z as well as the stabilizing effect of the half filled and filled d-shell, lead to configurations where the number of the ns electrons is less than two.³⁷

The main difference between the elements of the 4th and 5th period in comparison to the 6th period is caused by the fourteen 4f electrons which are filled in between the occupation of the $6s^2$ and the 5d electrons. The filling of the 4f shell shields the less penetrating 5d orbitals more effectively than the 6s because the d electrons are more fully barred from the inner parts of the atom by the centrifugal force $\frac{\ell(\ell+1)}{r^2}$. This means that the $d^m s^2$ configuration has increased stability in the 6th period except at the high end, where Pt and Au occur. This stabilizing effect of the $d^m s^2$ configuration becomes even larger in the next period, i.e., for the 6d elements, because the 7s electrons drop relatively deep into the atom. Thus they feel the very strong potential near the nucleus whereas the 6d electrons

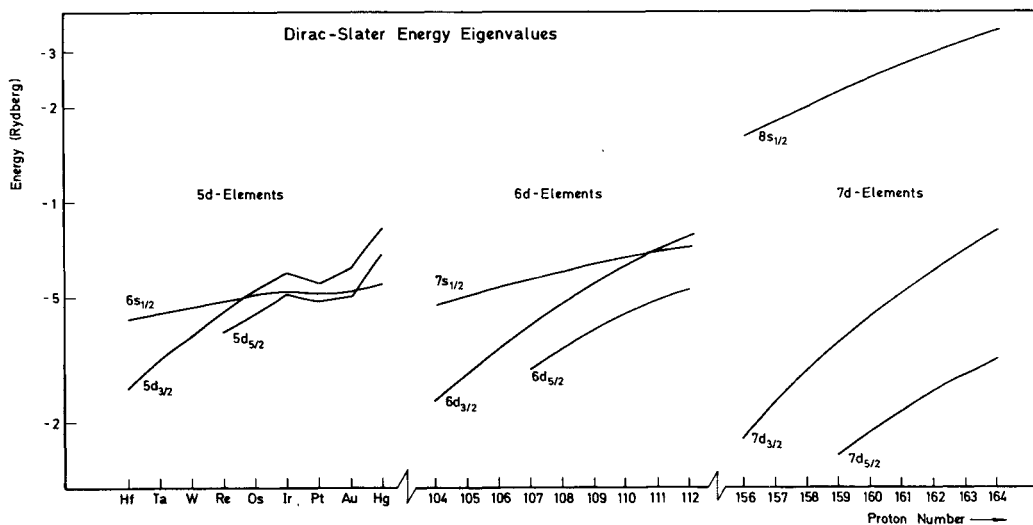


Fig. 5 The Dirac-Slater energy-eigenvalues of the outer electrons for the 5d-,6d- and 7d-elements. This shows the strong relativistic increase of the binding of the last s electronic shell and the increase in the splitting of the 7d subshells.

are shielded more strongly by the 5f electrons. Figure 5 shows the calculated energy eigenvalues of the outer d and s electrons for the d-transition elements of the 6th, 7th and 8th period. The trend is clear: the s electrons are bound stronger in the higher periods whereas the d electrons are lowered and, in addition, the d shell is split more and more into their two subshells and the energy separation increases. This altogether means that the electronic configuration of the neutral atoms of the transition elements in the 7th period will be given exactly by the rule $6d^m 7s^2$ as it is verified in the calculations shown in Table 4. The pairing energy which stabilizes filled and half filled shells is no longer sufficient to break this general rule. Furthermore, the pairing energy is as large as 1.1 eV per pair in the 3d elements and decreases for the higher periods. Cunningham³⁷ predicts this pairing energy to be less than 0.2 eV in the 7th period so that their influence will be very small.

Tab. 4 The groundstate electronic configuration of the atoms for the elements E104 to E112 according to the calculations of Waber et al.³² For E104 the groundstate configuration $6d7p7s^2$ according to the calculation of Mann⁴⁶ is included.

Atomic number	Groundstate configuration	Remarks
104	$6d7p7s^2$	5f orbitals full
105	$6d^37s^2$	
106	$6d^47s^2$	$6d_{3/2}$ orbital full
107	$6d^46d^17s^2$	$6d_{5/2}$ orbital begun
108	$6d^46d^27s^2$	
109	$6d^46d^37s^2$	
110	$6d^46d^47s^2$	
111	$6d^46d^57s^2$	
112	$6d^46d^67s^2$	6d orbital full

ii. Ionization Energies

The groundstate electronic configuration of the elements are not accurate indications of their chemical properties as it already was pointed out in sections III and IV. But the ionization potentials can be calculated with quite high precision for the superheavy elements by the self consistent field methods. However, in a calculation of the thermodynamical stability of a special compound all the other quantities have to be known additionally, such as energy of sublimation or dissociation and the crystal as well as hydration energy. But if one compares known compounds with hypothetical analogous compounds of superheavy elements, the change in the ionization energy will be the most dominant factor, as long as the crystal structure or the radii do not change drastically. The crystal energy itself $\frac{M \cdot v \cdot v}{d} \frac{a}{c}$ can be estimated quite accurately for ionic compounds as long as Madelung's factor

M, the ionic charge v_c of the element, the ionicity v_a of the anion and the interionic separation d are known. Figure 6 shows the experimental first ionization energies of the transition elements for the 5d elements as well as the calculated values for the 6d and 7d elements. This figure is located below Fig. 5 so that the lapse of the ionization energy curves can be compared directly with the lapse of the energy eigenvalues of the outer electrons. At the beginning of the 5d elements the ionization of the 5d electrons and at the end the ionization of the 6s electrons leads to the most stable configurations of the ion. The decrease of the ionization energy between W and Re comes from the spin-orbit splitting. The $5d_{3/2}$ subshell which holds only four electrons is completed at W and the occupation of the less tightly bound $5d_{5/2}$ subshell begins with Re. The linear behaviour thereafter is not continued for Pt and Au where the 5d shell is closed sooner at the cost of the 6s electrons. This shows that the ionization

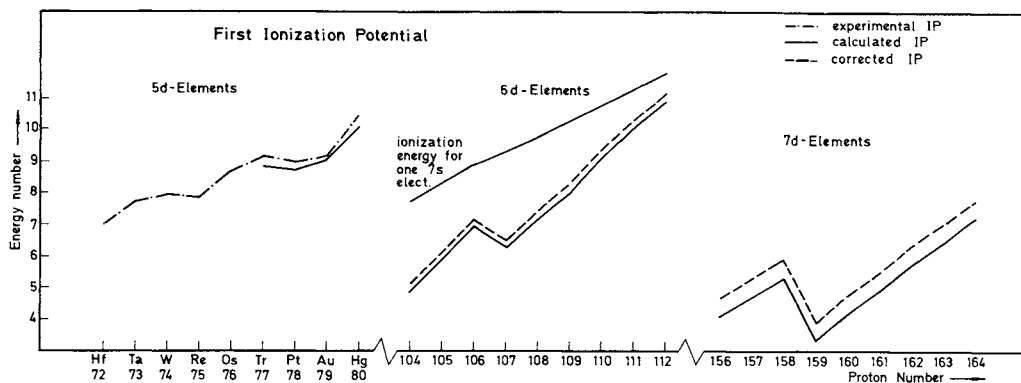


Fig. 6 The first ionization potential for the 5d-, 6d- and 7d-elements. Because of the averaged difference of 0.2 eV between the calculated and experimental values⁵⁵ in the 5d elements the values of the 6d elements are enlarged by this amount. Because the 7d electrons are the radially outermost electrons this correction factor was estimated to be 0.6 eV (in analogy to the correction factor of 0.8 eV for the 7p electrons in Fig. 9). For the 6d elements the calculated ionization energy for the removal of one 7s electron is also given.

energy of electrons in both s and d shells are very similar in binding energy so that the ionization energy is a measure for the removal energy of the outer electrons.

In the 6d elements instead always the ionization of one 6d electron leads to the most stable ion so that the ionization energy curve is quite parallel to the energy eigenvalues of the loosest bound 6d electron, which can be seen in Fig. 5 and Fig. 6. The calculated values are enlarged by 0.2 eV which equals the average difference between the experimental and calculated values for the 6th period. But in ionic compounds first the 7s electrons which are in the frontier orbital have to be removed and their ionization energy is nearly 3 eV higher at the beginning and about 1 eV higher at the end of the 6d elements which can be seen as well in Fig. 6. Nevertheless the relatively low ionization energy at the beginning of the 6d elements will be an indication that the maximal oxidation states for the 6d transition elements will be higher or at least equal to those in the 5d elements. Cunningham³⁷ gave a prediction of the sum of the ionization energies for the first four electrons for the 6d series elements which shows that this value will be about 6 eV smaller than for the 5d elements. This prediction will be not right at the end of the 6d elements where the relatively large ionization energy even for the 6d electrons will lead to larger values for the higher ionization energies. This indicates that their chemical behaviour is mainly inactive so that they are expected to be quite good noble metals.

iii. Radii

The metallic radii for the d elements for all periods are shown in Fig. 7. The large reduction of the radii at the beginning of the d elements stems mainly from the fact that at the beginning of the shell the outer electrons become itinerant conduction electrons and are distributed over the crystal and the d electrons from neighboring atoms overlap so that covalent bonding⁴⁵ becomes possible. Hence the d character of the binding is very large and the metallic radius decreases. The opposite is true at the end of the d transition series so that the metallic radius becomes larger again, whereas

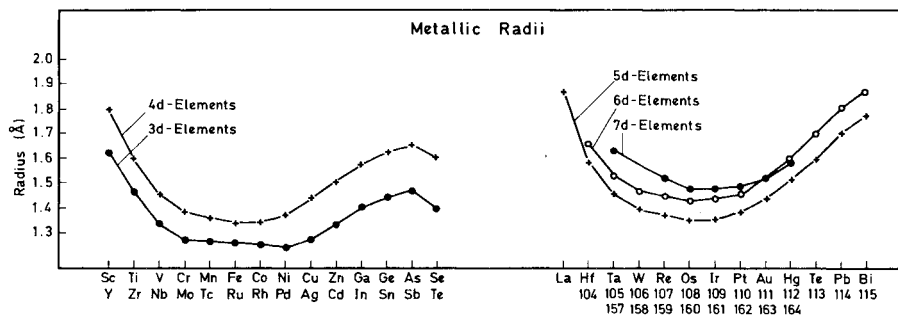


Fig. 7 The experimental 12-coordination metallic radii for the 3d-, 4d- and 5d-elements according to Pauling^{4,5} and the extrapolations for the 6d and 7d-elements. The metallic radii for the p elements are also drawn.

it decreases again according to the normal contraction of the elements when the metallic character is nearly lost which is in the middle of the p elements (which are also shown in Fig. 7). The metallic radius becomes larger from the 4th to the 5th period but stays rather constant in the 6th period, where the normal expansion is nearly compensated by the lanthanide contraction since these elements are filled in between. The elements E104 to E112 are totally analogous to the 5d elements in their position in the Periodic Table and because the actinide contraction is not as large as the lanthanide contraction, the radii of these elements will become larger again. From comparisons of the principal maxima of the outer electron shells in the 5th, 6th and 7th periods, it is expected that the metallic radii of the d elements of the 7th period will be smaller at the beginning and somewhat larger at the end of the 7th period. In contrast, the atomic radii of the 5d and 6d elements will be nearly the same because of the approximately equal principal maxima of the outer s electron wavefunctions. Cunningham²⁵ as well as Keller²⁶ gave some predictions for the ionic radius of the +4 state of E104 in the oxide (E104)O₂. This ionic radius will be very near to the analogous value of Hf so that from this prediction the crystal and

aqueous chemistry in the same oxidation state will be very similar for Hf and E104. This is expected to be true as well for the next d transition elements. Differences may arise, however, because of possible larger ligand field effects to be expected in the 6d as contrasted to the 5d elements because of greater spatial extension of the 6d orbital charge clouds.³⁷

iv. Conclusion

The conclusion from these considerations for the transition elements E104 to E112 are the following: Because of the probably larger metallic and covalent radii and the relative small ionization energy at the beginning of this group, it is quite clear that these elements will readily reach their maximum oxidation states. In these states they will be quite hard Lewis acids with relatively low polarisability only a little bit softer than the analogous 5d elements in the same oxidation states because of the very similar ionic radii. The same argument is valid in the middle of this region. The characteristic stability of the filled d shell will be even reduced because of the two separated subshells. That is, the usual concept of a pairing energy will not be valid. Nevertheless, the high ionization energies towards the end of the series indicates that these elements will be good noble metals and their chemically dominant form will be metallic. This characteristic stems mainly from the fact that the 7s electrons being in the frontier orbital have a larger ionization energy than in the earlier periods of the Periodic Table because of the larger relativistic effects. Therefore, for element E112 the main oxidation state will be 1 rather than 2. In contradiction to the normal behavior that the elements in higher oxidation states are harder Lewis acids the elements Hg, Tl and even Pb are softer in their high oxidation states. In the case of E112 this behavior will be even similar because in the oxidation state +1, the other strongly bound 7s electron will shield the more polarizable 6d electrons whereas in (E112)²⁺ the 6d electrons are in frontier orbitals. Because of the relatively large binding of the 6d electrons

Tab. 5 Summary of the results for the physical and chemical properties of the elements E104 to E112 according to Keller et al.²⁶, Cunningham³⁷ and Fricke et al.⁴¹.

Element	104	105	106	107	108	109	110	111	112
Atomic weight ^{c)}	272	275	277	280	282	285	288	291	294
Chemical group	IV B	V B	VI B	VII B	VIII	VIII	VIII	I B	II B
Most stable oxidation states ^{b)}	+ 4	+ 5	+ 6	+ 7	+ 8	+ 6	+ 6	+ 1.3	+ 1.2
First ionisation potential, eV	5.1	6.2	7.1	6.5	7.4	8.2	9.4	10.3	11.2
Sum of first four Ionis. Pot., eV ^{b)}	66	70	74	78	82	86	90	94	98
Metallic radius, Å	1.6 ^{a)}	1.47	1.41	1.39	1.37	1.38	1.40	1.46	
		1.53	1.47	1.45	1.43	1.44	1.46	1.52	1.60
Density, g/cm ³	18 ^{a)}	21.6	23.2	27.2	35 ^{b)} 28.6	28.2	27.4	24.4	16.8
Oxidation potential, V ^{a)}	$M \rightarrow M^{4+} + 4e^-$ > 1.7								
Melting point, °C ^{a)}	2100								
Boiling point, °C ^{a)}	5500								
Ionic radius, Å ^{b)}	0.75								

a) ref. 26

b) ref. 37

c) atomic weight acc. to ref. 8

at the end of the 6d elements it is very unlikely that in E112 higher oxidation states than two might occur. This result shows that the 7s electrons are already affected strongly by the relativistic effects whereas the splitting of the 6d subshell is not so strong that a chemical observable effect will result.

A chemical separation of these superheavy elements in nature or from targets where they were produced should be done very carefully in the sense that one either should try to separate the elements of the expected chemical group from all other elements or to utilize an expected different ionization state for the separation. A too accurate chemical separation for a special known element, for example, Hg to separate as well E112 might lead to poor results because E112 may be extracted as well by this procedure. This statement will be especially true for the search of unknown elements in nature in very well defined chemical compounds where superheavy analogous elements will be more depleted than enriched.

In Table 5 the results of Cunningham³⁷, Fricke et al⁴¹ and Keller et al²⁶ are summarized for the elements of the 6d-transition series, namely, E104 to E112.

Very recently Mann⁴⁶ has shown that the ground state configuration of Lawrencium is $7p\ 7s^2$ rather than $6d\ 7s^2$ as anticipated by continuation of the structure of the early members of the actinide series and by comparison with the lanthanide series. For Kurchatovium (E104) Mann found that $6d\ 7p\ 7s^2$ is very slightly more stable than $6d^2\ 7s^2$. However, the electrostatic interaction between various electrons need to be taken into account before a final decision can be reached. A very brief discussion of energy levels and spectroscopic questions of the neutral atoms are taken up later in connection with elements E121 and E122.

b. The 7p and 8s elements E113 to E120

The predicted lifetimes of the elements near $Z = 114$ are expected to be years or even longer. Hence the knowledge of the chemistry of these

Tab. 6 The groundstate electronic configuration of the atoms for E113 to E120 according to the calculations of Waber et al³² and Fricke et al⁴¹.

Atomic number	Groundstate configuration	Remarks
113	$7p^1 7s^2$	
114	$7p^2 7s^2$	$7p_{1/2}$ orbital full
115	$7p^2 7p^1 7s^2$	$7p_{3/2}$ orbital begun
116	$7p^2 7p^2 7s^2$	
117	$7p^2 7p^3 7s^2$	
118	$7p^2 7p^4 7s^2$	7p orbital full
119	$8s^1$	
120	$8s^2$	8s orbital full

elements is most important. This is especially true because it might be possible to find small amounts of these elements on earth.

i. Electron Configuration

As it is expected from the simple continuation of the Periodic Table (see Fig. 4) the six 7p electronic states will be filled from E113 to E118 and the two 8s electrons will be added for the elements E119 and E120. The results of the relativistic Dirac-Slater calculations by Waber, Cromer and Liberman³² and by Fricke, Greiner and Waber⁴¹ for the groundstate configurations of the neutral atoms given in Table 6 reproduce exactly this simple prediction. The more complex Dirac-Fock calculations by Mann and Waber³⁹ also yield the same result.

The energy eigenvalues of the outer electrons for these elements are compared with the analogous values for the elements Tl to Ra in the form of a graph, namely Fig. 8. The great similarity in the occupation pattern of the valence electrons can be seen. However, two significant differences

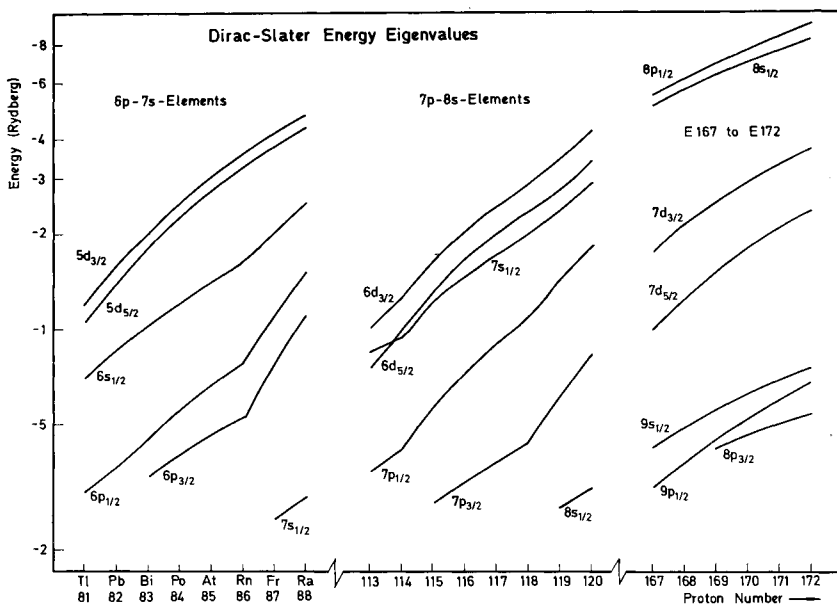


Fig. 8 The Dirac-Slater energy-eigenvalues of the outer electrons for Tl to Ra, E113 to E120 and E167 to E172. The main difference between these analog elements comes from the increased energetical splitting of the outer p shell. In the elements E167 to E172 the 8p_{3/2} and 9p_{1/2} electrons are very close to another so that they might form a p shell which is comparable to the 3p shell.

occur: the analogous s electrons are bound more tightly for higher Z and this binding results from the fact that the outer s electrons have a large probability density near the nucleus, i.e. they feel the deep potential. This is especially true because the relativistic effects such as the mass-velocity correction are fully included by use of the Dirac equation. The spin-orbit splitting of the 7p shell has increased compared to the 6p shell. A complete subshell with quantum numbers $n\ell j$ is spherically symmetric just as the complete non-relativistic $n\ell$ shells are. There will be a change in the angular distribution of the electrons in the relativistic treatment which could lead to different chemical behaviors for incomplete $n\ell j$ subshells.

ii. Ionization Potential

The ionization potential of the p elements can be seen in Fig.9 where the experimental ionization energies for the 3rd, 5th and 6th period are drawn together with the calculated values for the 6th, 7th and 9th period. Again this figure is located below Fig. 8 for better comparison of the trends of the energy eigenvalues and ionization energy. The ionization energy in the early periods increase linearly with only one break between the configuration p^3 and p^4 . This behavior is caused by the half filled shell where 3

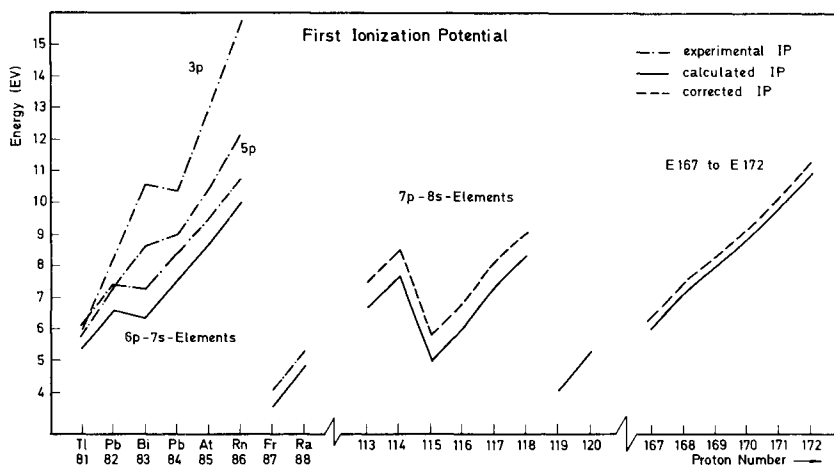


Fig. 9 The calculated first ionization potentials for the elements Tl to Ra, E113 to E120 and E167 to E172. For comparison the experimental values for the 3p, 5p and 6p-7s elements are also given⁵⁵. The increased spin-orbit splitting of the higher periods increases the difference between the ionization potential of the p^2 and p^3 configuration. The difference of 0.8 eV between the calculation and experiment for the 6p elements was taken to correct the calculated values for the 7p elements. The ionization potential is linear for the elements E167 to E172, because the $8p_{3/2}$ and $9p_{1/2}$ electrons have nearly the same eigenvalues. For these a correction of 0.4 eV was assumed because they are not the outer electrons as the normal outer p electrons are.

electrons occupy the three possible p orbitals with parallel spin. The pairing energy is negative so that the ionization energy is smaller for the 4th electron. This effect of the Russel-Saunders coupling is very significant in the first periods but becomes smaller for the higher periods because the LS coupling changes to intermediate coupling and from there to jj or spin-orbit coupling. In the 6th period, the break between p³ and p⁴ has vanished but instead a break occurs between p² and p³; this trend is expected since the spin-orbit coupling becomes more important than pairing energy in the p_{1/2} and p_{3/2} subshells. This break becomes even larger for the elements E113 to E118. Thus the ionization energy of the element E114 is nearly as large as the same value for the "noble gas" which occurs at Z = 118. The experimental and the calculated curves for the 6th period are very parallel with a difference of about 0.8 eV. Therefore, the first ionization energies of the elements E113 to E118 can be enlarged by 0.8 eV to get the more realistic values.

Keller et al.³⁸ gave very detailed predictions of the properties of elements E113 and E114. They extrapolated the difference between the experimental and calculated ionization energies in each group to the higher elements and found the correction factor for E113 to be 0.83 eV for E114 to be 0.72 eV. This procedure of extrapolation of the systematic difference between the experimental and calculated values is expected to give better results than were calculated by us.

iii. Oxidation States

The oxidation states of the E113 to E118 will follow the systematic trends for group III to VIII elements, namely, the elements with higher Z prefer the lower oxidation states. This is the expression of the fact that the s electrons with their greater binding energy lose their chemical activity as Z increases so that the p electrons will be the only valence electrons available. In the group III elements the oxidation state

is 3; only Tl has also a monovalent state which is associated with the ionization of the single $6p_{1/2}$ electron and the relative increased stability of the 6s electrons. From this behavior the high ionization energy of the $7p_{1/2}$ electron and the even more increased stability of the 7s electron the principle oxidation state of E113 is expected to be +1. It will be a soft Lewis acid but even stronger than in a possible +3 state.

From Si to Pb in the group IV elements the stability of the oxidation state +4 decreases and +2 increases instability because of these same reasons. The tetravalency especially in the chemical group IV is connected with a sp^3 hybridization and strong covalent bonding. The energetic difference between the ns and np electrons increases for the higher periods as it is seen in Fig. 3. Also, the spin of the p electrons couple strongly to their own angular momentum. Therefore, a sp^3 hybridization is no longer possible for E114 which means that the predominant oxidation state of $Z = 114$ will be divalent. However, since the outermost s and d electrons in E114 have approximately equal energies some form of an sd hybrid would be possible. Thus, one cannot exclude the possibility that a volatile hexafluoride might form. Another aspect is given by Drago⁴⁷ who uses thermodynamic arguments to explain the weakening of the covalent bonding with increasing Z. He points out that in higher Z elements the valence electrons are spread over a larger volume so that the overlap with the orbitals of the anion will be reduced and that the heavier elements have more inner electrons to repel the inner electrons of the ligand anion. He overlooked the relativistic effects; namely, that spin orbit splitting may offset hybridization energies and that the angular distribution of the charge density may also be altered.

Occupation of the $7p_{3/2}$ subshell begins at $Z = 115$ with a binding energy which is only half as large as that of the $7p_{1/2}$ electrons, so that the elements E115, E116 and E117 will have +1, +2 and +3, respectively, as their

normal oxidation state. The higher oxidation states will be possible only in the presence of strong oxidizers.

An interesting question is whether element E117 which is in the chemical group of the halogens would form the -1 anion. Cunningham²⁵ predicted a electron affinity of 2.6 eV whereas the calculations of Waber, Cromer and Liberman³² calculated a value of only 1.8 eV. Cunningham describes the solid element E117 to have a semi-metallic appearance. It should form stable oxyions of the (III), (V), and (VII) states and stable interhalogen compounds.²⁵ Because of the small electron affinity it might not exhibit the -1 oxidation state, which is even further suggested by the smaller value calculated by Waber et al.³² Certainly it will be a very soft base compared with flouride or chloride which have a electron affinity of 4 eV resp. 3 eV.

The "in-noble gas" at $Z = 118$ will be a very weakly noble gas in comparison to the chemical reactivity of He and Ar and will resemble Xe and Rn. Quite early, in 1965, Grosse³⁵ performed a very extensive study of this element, E118, based on pure extrapolation of a number of physical and chemical properties of the noble gases of lower atomic number. Specifically, he predicted that element E118 will be the most electropositive of all noble gases and will form a partially ionized diflouride compound, a tetraflouride and tetraoxide analogous to XeO_4 and perhaps stable compounds with chlorine.

Previous analysis by Waber⁴⁸, following an informal discussion with Fano, indicated that negative ions of the noble gases would have configurations such as $np^5 (n+1)s^2$. The spectra of such species have been found at the National Bureau of Standards following electron bombardment of the noble gases. It would be expected that E118 could readily form such anions. Calculations have not confirmed the likelihood of such species.

Independently Grosse²⁴ and Cunningham²⁵ found that the expected boiling point of liquid E118 is about $-15^\circ C$, so that it will be nearly a

"noble fluid". It might be predicted as well that the crystalline form would be much denser than the other noble gases. That is, the bonding in solid E118 would be stronger than given by van der Waals forces. These predictions from systematic continuation are supported by the calculation. Its first ionization energy is small, only 9 eV, and the strongly split p shell giving rise to frontier orbitals at the surface of the atom suggest that E118 will be more a normal element with many possible compounds than a noble element. Thus it will continue the trend towards chemical reactivity first observed in Xenon.

iv. Radii

In the predictions of atomic and ionic radii of these elements, one gets into great difficulties. The concept of atomic radii of Slater⁴⁹ who says that the atomic radius can be well defined as the radius of the principal maximum of the outermost electron shell, i.e. of the frontier orbital, works quite well in large parts of the Periodic System. However, at the end of the d-transition elements and for most of the p elements, this definition apparently yields inaccurate results. Tabulation of orbital radii have been made by Waber and Cromer⁵⁰ up to Lawrencium. The principle maxima of the outermost shells show that the deviation between experiment and their calculation increases for higher Z near the end of the d and at the beginning of the p elements. So a prediction of the atomic radii in the region between E110 and E118 from the calculation of the radius of the outermost shell seems to be not too accurate. If one continues, however, the trends in the behavior of metallic or ionic radii as it was done by Grosse²⁴, Keller et al³⁸ and Cunningham²⁵, one gets results which will be quite accurate. The question may be raised whether Slater's definition is good in the region of superheavy elements where the orbital shells split largely into their two subshells. Which subshell is representative for predictions of atomic radii and what does it mean in this context when a subshell is largely buried inside the

atom? Because of these reasons, the predictions of Fricke et al.⁴¹ were done on the basis of the comparison of the computed total electron density at large radii of known elements. The radius of the superheavy elements was taken where the density was equal to the density of the known analogous element at its experimental radius.

v. E113 and E114

Coming back to the most interesting elements E113 and E114, Keller et al.³⁸ predicted by pure extrapolation quantities such as metallic and ionic radii, densities, crystal structure and a lot of thermodynamical quantities which are also listed in Table 7. Most of these extrapolations are so suggestive that we give an example in Fig. 10 where the heat of sublimation of element E113 and E114 is deduced.

In general, the chemical behavior of E114 as described by Keller et al.³⁸ is expected to be similar to that of divalent lead. The calculated

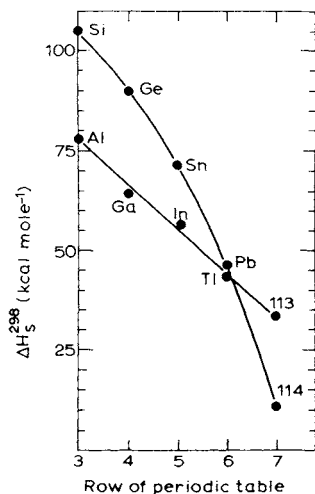


Fig.10 As an example for the extrapolation of chemical and physical quantities the evaluation of the heat of vaporization for the elements E113 and E114 is shown here as it was given by Keller et al.³⁸.

Tab. 7 Summary of the results for the physical and chemical properties of the elements E113 to E120 according to Grosse²⁴, Cunningham²⁵, Keller et al.³⁸ and Fricke et al.⁴¹.

Element	115 ^{a)}	114 ^{a)}	115	116	117 ^{e)}	118 ^{e)}	119 ^{e)}	120 ^{e)}
Atomic weight ^{c)}	297	302	303	308	313	314	315	320
Chemical group	III A	IV A	V A	VI A	VII A	0	I A	II A
Most probable oxidation state	+ 1	+ 2	+ 1,3	+ 2,4	+ 3,1	+ 4,0	+ 1	+ 2
First ionisation potential, eV	7.4	8.5	5.9	6.8	9.3	8.2 ^{b)}	9.8	9.0 ^{b)}
Second ionisation potential, eV		16.8			16	15	23	10
Oxidation potential, V	$M - M^+ + e^-$ - 0.6	$M - M^{2+} + 2e^-$ - 0.9			$2M^- - M_2 + 2e^-$ + 0.25-0.5		$M - M^+ + e^-$ + 2.9-3	$M - M^{2+} + 2e^-$ + 2.9
Metallic radius, Å	1.75	1.70 ^{b)}	1.87	1.83			2.9	2.6 ^{b)}
Ionic radius, Å	1.48	1.31			2.3		1.8	1.6
Density, g/cm ³	16	14	13.5	12.9				
Covalent radius, Å		1.70 ^{d)}			1.8	2.3	2.6 ^{g)}	2.0 ^{g)}
Melting point, °C	430	67			350-550	-15	0-30	680
Boiling point, °C	1130	147			610	-10	630	1700
Heat of vaporisation, kcal/g-atom	31	9	Critical temperature, °C ^{f)}		192			
Heat of sublimation, kcal/g-atom	34	10	Critical pressure, atm ^{f)}		65.5			
Debye temp., °K	70	46	Critical density, g/cm ³ ^{f)}		1.847			
Entropy, cal deg ⁻¹ (g-atom) ⁻¹ (25°)	17	20						

a) predicted properties acc. to ref. 38

e) predicted properties acc. to ref. 25

b) ref. 41

f) predicted properties acc. to ref. 24

c) Atomic weight acc. to ref. 8

g) atomic radius

d) extrapolation of Paulings covalent radius

oxidation potential of $-0.9V$ indicates, however, that E114 is considerably more noble. The large negative oxidation potential coupled with a large polarizability should enable $(E114)^{2+}$ to form strong complexes with anions; its large ionic radius would be favorable. In excess halogen acid, for example, complexes of the type $(E114)X_n^{(n-2)-}$ should be quite stable. A complex analogous to the plumbite ion is also expected. The sulfate and sulfide should be extremely insoluble. The acetate and nitrate would be soluble.

The chemistry of E113 is expected to be similar to that of the thallos ion although $E113^+$ being a soft Lewis acid should form complexes more easily. The predicted radius of $E113^+$ is 1.48\AA , the same as Rb^+ , and is not much larger than Tl^+ itself (1.40\AA). The large polarizability and moderately large negative oxidation potential of $-0.6V$ will increase the binding of anions to $E113^+$, but the large radius and the associated electronic repulsion may counteract these effects. Since H_2O is a hard solvent, salts of E113 should have low solubilities whereas the nitrate and fluoride should be quite soluble. The $E113^+$ ion may, however, form only a slightly soluble oxide whose solution will be alkaline; (considerable polymerization could be anticipated) and this solution will readily absorb carbon dioxide from the air. Like argentous and aurous oxides, the oxide of $E113^+$ may be soluble in ammonia.

vi. E119 and E120

In the two elements E119 and E120, the 8s electrons will be bound very tightly and therefore these two elements are expected to be chemically very similar to Cs and Ba or Fr and Ra. In Fig. 8 the energy eigenvalues of the outer electrons and in Fig. 9 the ionization potential for E119 and E120 are drawn in comparison to Fr and Ra. The main oxidation state of E119 and E120 will be 1 and 2 as it is normal for alkali and alkaline earth metals. Their ionization potential will be about 0.5 eV higher than in the

elements Fr and Ra which mainly results from the effect that the s electrons penetrate deep into the atom and feel the very strong potential near the nucleus. Therefore, their atomic radius according to the simple definition of Slater⁴⁹ is expected to be 2.6 and 2.0Å, very similar to the values of Cs and Ba. Cunningham²⁵ predicted the chemical behavior of both elements from extrapolations of the known alkali and alkaline earth metals. The main feature is the well known trend for elements to become more electropositive for higher Z. From the calculation of the ionization potentials given in Fig. 9 and atomic radii, it seems that this trend has been arrested and may possibly slightly have changed direction.* This would mean that the chemistry of E119 and E120 will be more similar to Cs and Ba respectively than to Fr and Ra. The ions will have larger radii than Cs⁺ and Ba²⁺ because of the larger extension of the filled 7p shell in comparison to the 5p shell so that hydration will be more important and crystal energies will be different. Another important point is that possibly higher oxidation states might be reached in the presence of strong oxidizers because the ionization energy of the outer 7p_{3/2} electrons are only of the order of 10 eV.

In Table 7 the chemical and physical properties for the elements E113 to E120 from different predictions are summarized.

c. The 5f and 6g Elements E121 to E154

In the region of the Periodic System starting with element E121, we come to the next very long transition series which is characterized not only by the filling of the 6f but also of the 5g electron shells. Seaborg^{5,10} called these elements Superactinides.

Unfortunately, it is expected that the chemistry of these elements cannot be studied because the theoretical investigations of the nuclear

* This new trend can even be seen much better for the 9s elements E165 and E166 in Fig. 14 and 15.

stability predict that these elements will be unstable with short lifetimes.^{8,9} A large number of theoretical calculations of the groundstate electronic configurations have been made for this region because the proton number $Z = 126$ was expected for a long time to be the center of the first island of stability. Now it is considered to be unlikely. Even if $Z = 124$ or 126 is a magic proton number besides $Z = 114$ the half-lives of these elements will be smaller than 10^{-3} sec because of rapid α -decay.⁹ Nevertheless, the chemistry of these elements would be very interesting.

i. Electron Configuration

In the lanthanides and actinides, the competition between the outer d and inner f shells determines the ground state electron configuration as well as the chemistry of these elements. Here in the superactinides not merely two but four electron shells, namely the $8p_{1/2}$, $7p_{3/2}$, $6f_{5/2}$ and $5g_{7/2}$ are expected to compete nearly simultaneously in the atom at the beginning of the series of these elements and these open shells determine together with the $8s$ electrons the chemistry. Many calculations have been performed to predict the groundstate electronic configurations of the neutral atoms in this region. The newest Dirac-Fock calculations of Mann and Waber³⁹ are expected to be the best and therefore their results are summarized in Table 8. Three most interesting things occur in these elements. First, the $7p_{1/2}$ electrons are filled in beginning with element El21 and remain in all following elements. This is clearly a direct relativistic effect which is mainly effective for all $j = \frac{1}{2}$ levels. Second, during the filling of the g electronic levels (and at the end of the superactinides of the f levels as well) some other electrons always remain in the groundstate configuration in contradiction to the analogous lanthanides and actinides where at the beginning some d electrons are filled in which are removed during the filling of the f shell. Third, the effective binding of an electron with large values of the angular momentum is accompanied by the radial collapse of the orbital, the centrifugal term $\ell(\ell+1)/r^2$ keeping

it extended. For example, the effective radius of 5g electrons changes from 25 Bohr units in E120 to 0.8 for E121 according to Griffin et al³¹ who used non-relativistic wavefunctions. The Dirac-Fock calculations by Mann and Waber³⁹ indicate that collapse occurs at higher atomic number as a direct consequence of the indirect relativistic effect so that the 5g shell starts first in element E125.

Tab. 8 The groundstate electronic configuration for the elements E121 to E152 according to the calculations of Mann and Waber³⁹ and Mann³⁶.

Atomic number	Groundstate configuration					Remarks
	5g	6f	7d	8p	8s	
121	--	--	--	1	2	
122	--	--	1	1	2	
123	--	1	1	1	2	
124	--	3	--	1	2	
125	1	3	--	1	2	
126	2	2	--	2	2	p _{1/2} orbital full
127	3	2	--	2	2	
128	4	2	--	2	2	
129	5	2	--	2	2	
130	6	2	--	2	2	
131	7	2	--	2	2	
132	8	2	--	2	2	g _{7/2} orbital full
134	8	4	--	2	2	
136	10	4	--	2	2	g _{9/2} orbital begun
138	12	3	1	2	2	
140	14	3	1	2	2	
141	15	2	2	2	2	
142	17	2	2	2	2	
144	18	1	3	2	2	g orbital full
145	18	3	2	2	2	
146	18	4	2	2	2	
149	18	6	3	2	2	f _{5/2} orbital full
150	18	6	4	2	2	
152	18	9	3	2	2	

ii. Level Structure of E121 and E122

In view of the large amount of information which has been accumulated about these elements, it is appropriate to discuss briefly the energy level structures of two of the lightest superactinides E121 and E122, which is mainly interesting for optical spectroscopy. Mann and Waber³⁹ and Cowan and Mann⁵¹ investigated the multiplet structures of these two elements and compared these with their congeners Ac and Th. The purpose was to confirm whether after including electrostatic interaction between electrons the increased splitting of the 8p electrons would be sufficient to alter the ground-state configuration.

The solid lines in Fig. 11 taken from the paper by Mann and

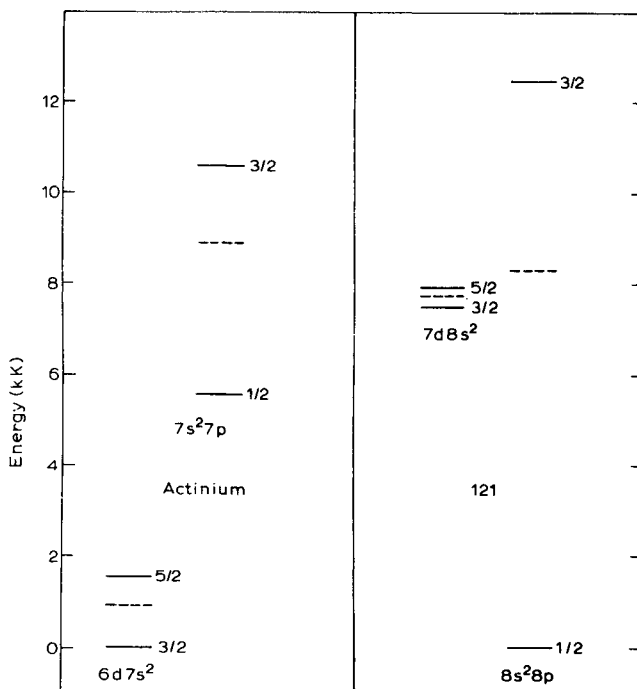


Fig.11 Calculated term levels of ds^2 and s^2p configurations of Ac and element E121. The dashed lines indicate average energies. This figure was taken from the paper of Mann and Waber³⁹.

Waber³⁹ represent subconfigurations for the individual sets of quantum numbers $|nlj\rangle$ and the barycentre, which is the weighted average taken over the j values, is indicated by a dashed line. The $8p$ state will occur in the groundstate of E121 because (a) the $8p$ barycentre is lower than that of the $7d$ electrons and (b) the spin-orbit splitting ΔE of the $8p$ electrons is much greater than ΔE for the $7d$ electrons. The one-electron treatment of Ac and E121 gives the complete energy-level diagram.

When there are two electrons occupying incomplete shells, the level structure can only be obtained by including the inter-electron Coulomb and exchange interactions (which slightly lift the degeneracy of the various $|nlj\rangle$ orbitals). The necessary Slater F^k and G^k integrals are evaluated in the course of the Dirac-Fock program. By comparing the ratio of the spin-orbital energy parameter ζ_{8p} to $F^2(8p,8p)$ for the group IV elements it was clear that the Dirac-Fock (DF) calculations indicate more jj coupling than occurs in ^{82}Pb .

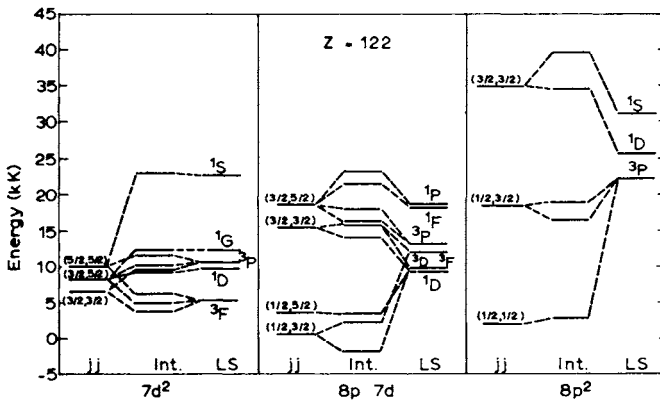


Fig.12 Computed energy levels of the three low-lying configurations of element E122 in the proper intermediate coupling (Int.) and also in the jj - and LS-coupling limit, with state correspondences among the three calculations indicated by dashed lines. This figure was taken from the paper of Cowan and Mann⁵¹.

Cowan and Mann⁵¹ made detailed calculations of level structure for LS, jj and intermediate coupling conditions and their work for E122 is summarized in Fig. 12. In this example, the ground state configuration is unchanged in the 3 distinct calculations. However, the effect on optical spectra would not be small.

They suggest that at first glance, the level structure of E124 with the configuration f^3ps^2 will give rise to fewer lines than its homologue $92U$ which has the ground state configuration (f^3ds^2 or f^4s^2). However, since the barycentre of the f^3ds^2 configuration is only slightly higher than that of f^3ps^2 , configuration interactions with this as well as with other low-lying configurations will cloud the issue and virtually uninterpretable optical spectra will probably occur.

iii. Oxidation States

The very small binding energies of all electrons in the $8s_{1/2}$, $8p_{1/2}$, $7d_{3/2}$, $6f_{5/2}$ and $5g_{7/2}$ shells makes it plausible that most of these electrons might be oxidized in chemical compounds so that very high oxidation states might be reached in complex compounds. Hybridizations of unknown complex character will be possible because four (possibly also the 5g) shells are available with different angular momenta but nearly the same energy.

From the point of view of forming ionic compounds, it is not too relevant whether the 8p or 7d shell is occupied in the neutral atom, as has been studied in extenso by Mann and Waber.³⁹ The significant question for more ionic compounds instead is whether in the ions after all outer s, p and d electrons are removed, some g or f electrons will be in frontier orbitals or whether they might be easily excited to an outer electron shell so that they can be removed as well. Prince and Waber⁵² made both Dirac-Slater and Dirac-Fock calculations for various configurations of the monovalent and divalent states of E125 and E126. Results were presented in the monograph by Waber.³⁵ It was shown that already in the divalent state of E126 one g electron

has changed to an f electronic state. However, the $8s$ electrons are not the first to be removed. Thus the divalent ions will be expected to act as soft Lewis acids and possibly form covalent complex ions readily. Crystal or ligand fields influence the nature of the hybridization. Details such as directionality of bonds will be determined by the occupation of the frontier orbitals and, of course, of the approaching anion.

No calculations for highly ionized states have been done to date. However, from a comparison of the energy eigenvalues of the different shells, it seems that oxidation states may well reach very high values at about or near E128 in complex compounds but normally these elements will have 4 as their main oxidation state in ionic compounds. The maximal valency will be reduced to 6 at E132 and three or four it will be in the region of E144. At the end of the superactinide series, the normal oxidation states are expected to be only 2 (or possibly 0) because the $6f$ shell is buried deep inside the atom and the $8s$ and $8p_{1/2}$ electrons which are in frontier orbitals are bound so strongly that they will be chemically inactive. Only the $7d$ electrons may be available for bonding. The calculations show that in element E154 the f shell is filled and no electrons extend radially beyond the $8s$ and $8p_{1/2}$ shell. This would mean that this element would be chemically very inactive with a behavior of a noble element. This is especially interesting because the newest calculations of the nuclear stability⁸ show that the center of the second island of stability lies lower than $Z = 164$.

The lanthanide contraction of about 0.044\AA per element is larger than the actinide contraction of about 0.03\AA per element, because the $4f$ wavefunction is less localized than the $5f$ wavefunction and shrinks more rapidly with increasing nuclear charge. The analogous contraction is expected in the superactinide series. The total effect will be very large because of the 32 electrons which will be filled in the deep $5g$ and $6f$ shell. From a comparison of the outer electron wavefunctions of the lanthanides, the

actinides and superactinides, a contraction of about 0.02\AA per element starting with elements E121 and continuing to E154 can be expected.

d. The Elements E156 to E172

The latest calculations of Grumann et al⁸ make it plausible that a large island of stability might be possible around the magic proton

Tab. 9 The groundstate electronic configuration for the elements E156 to E172 according to the calculations of Fricke³⁴, Mann³⁶ and Fricke et al⁴¹. The preliminary results for E184 are also included.

Atomic number	Groundstate configuration	Remarks
156	$7d^2$	
157	$7d^3$	
158	$7d^4$	$7d_{3/2}$ orbital full
159	$7d^4 7d^1$	$7d_{5/2}$ orbital begun
160	$7d^4 7d^2$	
161	$7d^4 7d^3$	
162	$7d^4 7d^4$	
163	$7d^4 7d^5$	
164	$7d^4 7d^6$	7d orbital full
165	$9s^1$	
166	$9s^2$	9s orbital full
167	$9s^2 9p_{1/2}^1$	
168	$9s^2 9p_{1/2}^2$	$9p_{1/2}$ orbital full
169	$9s^2 9p_{1/2}^2 8p_{3/2}^1$	$8p_{3/2}$ orbital begun
170	$9s^2 9p_{1/2}^2 8p_{3/2}^2$	
171	$9s^2 9p_{1/2}^2 8p_{3/2}^3$	
172	$9s^2 9p_{1/2}^2 8p_{3/2}^4$	8p orbital full

Periodic table of the elements

H 1																	He 2																												
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10																												
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	Ar 18																												
K 19	Ca 20	Sc 21											Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	Kr 36																		
Rb 37	Sr 38	Y 39											Zr 40	Nb 41	Mo 42	Tc 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54																		
Cs 55	Ba 56	La 57											Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	At 85	Rn 86																		
Fr 87	Ra 88	Ac 89											Th 90	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101	No 102	Lw 103																			
119	120	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150	151	152	153	154	155	156	157	158	159	160	161	162	163	164
165	166																													167	168	169	170	171	172										

Fig. 13 The Periodic Table of the elements in its continuation up to element E172, taken from Fricke et al.⁴¹.

number of 164 with lifetimes of the order of minutes and possibly years. This is seen in Fig. 2. Therefore, the knowledge of the chemistry of these elements might become very important.

i. Electron Configuration

In Table 9, the electron configurations of the expected ground-state configurations for the neutral atoms according to the calculations of Mann³⁶ and Fricke et al^{34,41} are given. This shows that formally the elements E155 to E164 are the d-transition elements of the 8th period.

The relativistic enhancement of the subshells with $j = 1/2$ is so large that in the elements E165 to E168 instead of the $8p_{3/2}$ shell, the $9s$ and $9p_{1/2}$ states will be occupied. Hence the filling of the $8p_{3/2}$ electrons can follow only in the elements E169 to E172. This surprising result makes it possible to give a formal continuation of the Periodic Table as it is shown in Fig. 13 because there are six p electrons from two different shells which are energetically very close so that they will nicely form a normal p shell. Therefore, the 9th period will be quite analogous to the 2nd and 3rd period.

ii. E156-E164

In the periods before the 8th period normally all d and p elements are influenced in their chemical behavior more or less by the outer s electrons. This is no longer true for the d-transition elements E155 to E164 where the 8s and $8p_{1/2}$ electrons are bound so strongly that they will not participate in the chemical binding. Their principal maxima are well inside the d electrons so that for the elements up to E164 the 7d electrons with their relatively low ionization energy shown in Fig. 6 are the only valence electrons. (A difference of 0.6 eV between calculated and probable realistic values was assigned because the 7d shell is here the outer shell as it is the 7p shell in the elements E113 to E118, where the difference was 0.8 eV.) This electron structure is quite similar to the d elements of the lower periods where the outer s electrons are removed, so that as an

initial overall prediction, one can say that the aqueous and ionic behavior of an E^{m+2} ion of the lower d-elements is comparable to an E^m ion of the elements E155 to E164 in the same group after making allowance for the different ionic sizes and charge. Even for the neutral atom, a similarity can be seen between Pd where ten d electrons and no s electrons form the outer shell and element $Z = 164$. Penneman et al⁴⁰ recently gave a very extensive and sophisticated chemical discussion about element E164. They conclude that it would be chemically quite active. In aqueous solutions, it will be predominantly bivalent but stronger ligands will form tetra- and hexavalent bonds. In other words, it will be a soft Lewis acid. They arrived at their conclusions from calculations of very high ionization energies and semi-empirical formulas for the hydration energy. They agree with Fricke et al⁴¹ that the metallic form might be quite stable but they compare it more with Hg whereas Fricke et al predict E164 to be a noble metal which should be in the same chemical group as Pd and Pt. The metallic form should lead to a larger cohesive energy than nearly any other element would do because of the covalent bonding, so that its melting point, therefore, should be quite high. Their prediction of the metallic radius are given in Fig. 7.

Fricke et al⁴¹ compare this element $Z = 164$ for formal reasons also with the "innoble gas" E118. The structure of the valence electrons: a filled outer shell, the ionization energy, the radii, the energy, eigenvalues and the energetic splitting of the filled d-states in E164 and the p shell in E118 are very similar.

Therefore, a detailed study of element E118 could lead to some better chemical predictions of element E164; of course, the angular distribution of the p and d shell is different. The predictions of the chemistry of 164 by Penneman et al⁴⁰ go much further. They discuss the aqueous chemistry, ionic state and the tendency towards complex-formation of this element in great detail. They predict a large number of compounds which will be possible and as well as impossible ones.

iii. E165 and E166

After element E164 with the complete 7d shell the 9s electrons will be filled in E165 and E166. This means that a new period with elements of the chemical group I and II has begun. In Fig. 14 the energy eigenvalues of the outer electrons and in Fig. 15 the ionization energies for all Ia, IIa and Ib, IIb chemical group elements beginning with Cs are drawn, because the elements E165 and E166 have a as well b characteristic. The ionization energies of the 9s electrons are greater than the values of Na in the 3rd and Ca in the 4th period which results from the strong relativistic

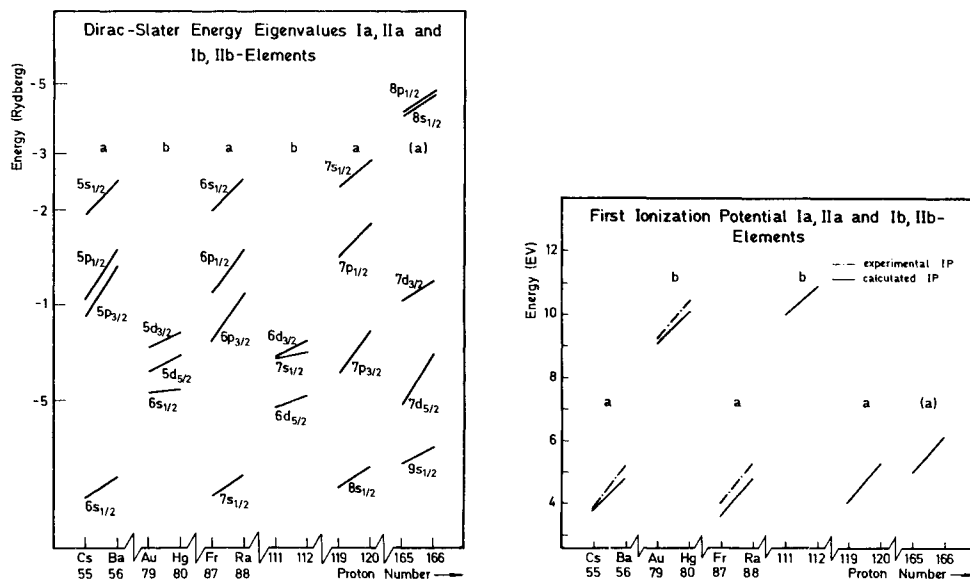


Fig.14 The Dirac-Slater energy-eigenvalues for all Ia,IIa and Ib,IIb elements beyond Cs. This comparison is done to show the intermediate character of the elements E165 and E166: a low lying s shell as in the Ia and IIa elements but an underlying d shell as in the Ib and IIb elements.

Fig.15 The first ionization potentials for all Ia, IIa and Ib, IIb elements beyond Cs. The increase for the high Ia and IIa elements is remarkable because of the strong relativistic binding of the analog s electrons.

Tab. 10 The results for some physical and chemical properties of the elements E156 to E172, partially taken from the results of Fricke et al.⁴¹

Element	156	157	158	159	160	161	162	163	164	165	166
Atomic weight	445	448	452	456	459	463	466	470	474	477	481
Chemical most analog group		III B	IV B	V B	VI B	VII B	VIII	VIII	VIII	I A	II A
Outer electrons	$7d^2$	$7d^3$	$7d^4$	$7d^5$	$7d^6$	$7d^7$	$7d^8$	$7d^9$	$7d^{10}$	$9s^1$	$9s^2$
Most probable oxydation state	+ 2	+ 3	+ 4	+ 1	+ 2	+ 3	+ 4	+ 5	+ 2,4,6	+ 1,3	+ 2
Ionisation potential (eV)	4.7	5.3	5.9	3.9	4.8	5.5	6.4	7.0	7.8	5.0	6.1
Metallic radius (Å)	1.7	1.63	1.57	1.52	1.48	1.48	1.49	1.52	1.58	2.1	1.8
Density (g/cm^3)	26	28	30	33	36	40	45	47	46	7	11
Element	167	168	169	170	171	172					
Atomic weight	485	489	493	496	500	504					
Chemical most analog group	III A	IV A	V A	VI A	VII A	0					
Outer electrons	$9s^2 9p_{1/2}^1$	$9s^2 9p_{1/2}^2$	$9s^2 9p_{1/2}^1 8p_{3/2}^1$	$9s^2 9p_{1/2}^2 8p_{3/2}^2$	$9s^2 9p_{1/2}^2 8p_{3/2}^3$	$9s^2 9p_{1/2}^2 8p_{3/2}^4$					
Most probable oxydation state	+ 3	+ 4	+ 5	+ 6	+ 7,3, - 1	0,4,6,8					
Ionisation potential (eV)	6.4	7.5	8.3	9.2	10.2	11.3					
Atomic radius (Å)	1.58	1.48	1.39	1.33	1.27	1.22					
Density (g/cm^3)	17	19	18	17	16	9					

effects in the binding of the 9s electrons. Here the trend becomes very obvious that the radii and ionization energies of alkaline and alkaline earth elements increase with Z whereas in the first part of the Periodic System they decrease. From this side E165 and E166 will be members of the groups Ia and IIa. From a more chemical point of view, they will be likely more members of the Ib and IIb groups because of the 7d shell which is more comparable to the elements Au and Hg (but also to the elements E119 and E120) as can be seen from Fig. 14. Therefore, higher oxidation states than 1 and 2 might readily occur.

iv. E167-E172

Between E167 and E172 the $9p_{1/2}$ and $8p_{3/2}$ electrons will be filled and it is quite an accident that the energy eigenvalues are so close together (see Fig. 8) that a p shell will occur containing 6 electrons with nearly no splitting of the subshells but different principal quantum numbers which is very analogous to the non-relativistic p shell in the 3rd period. Therefore, the normal oxidation states of the elements E167 to E170 will be 3 to 6. Element E171 is expected to have many possible oxidation states between -1 and +7 as the halogens do. Here again, the electron affinity will be high enough to form a hydrogenhalide like H(E171). We calculated a value of the electron affinity of 3.0 eV which is as high as the value of Cl^- so that $(\text{E171})^-$ will be quite a hard base. Element E172 will be a noble gas with a closed p shell outside. The ionization energy of this element as shown in Fig. 9 is very near to the value of Xe, so that it might be very similar to this element. The great difference between Xe and E172 will only be that element E172 is expected to be a fluid or even a solid because of its large atomic weight.

As indicated in connection with E118, it will tend to be a strong Lewis acid and hence compounds with F and O are expected as have

been demonstrated for Xenon. In Table 10, the chemical and physical properties of the elements $Z = 156$ to 172 are tabulated.

v. E184

Penneman, Mann and Jorgenson⁴⁰ speculated about the chemistry of element E184, because some authors believe in a double magic configuration at $Z = 184$ and $N = 318$. This can be regarded mere speculation because (a) no calculations about nuclear stability have been made up to that time, (b) it seems to be nearly impossible to create this nucleus with any known combination of nuclei and (c) no calculations of the atomic behavior were available to them. Mann reported that his Dirac-Fock program was unable to go beyond $Z = 176$. Nevertheless, their speculation might be quite plausible because after element E172 another extremely long transition series would start with the filling of the 6g, 7f and 8d shells. The complication of new 5h electronic states with ($\ell=5$) would also arise. These loosely bound electrons make it plausible that it would be very easy to reach very high oxidation states as stated by Penneman et al.⁴⁰ A model calculation of Fricke and Waber⁵³ taking into account a phenomenological formulation of quantum electrodynamical effects shows that the groundstate of E184 is given by $(Z=164)$ core + $9s^2 9p_{1/2}^2 8p_{3/2}^4 6g^5 7f^4 8d^3$. But only the $8d^3$ and $7f^4$ electrons might be available for chemical bonding. The $10s$ and $10p_{1/2}$ electrons do not appear in the groundstate configuration as well as the $6h \frac{11}{2}$ electrons. So it seems that the chemical behavior of element E184 is even simpler than the early superactinides and a comparison with U or Np might be reasonable.

VI. Conclusion and Critical Analysis of the Predictions

The predictions of the chemical behavior of superheavy elements as they are summarized here will be only a starting point for chemical studies in this region. Nevertheless, it seems very plausible that the predictions will not be too far away from reality for the elements up to $Z = 120$. In

this region one is close enough to the part of the Periodic Table with known elements that a combination of the simple continuation of the trends in the chemical groups together with the very credible calculations, tested in the known part of the elements will lead to quite good predictions about the physical and chemical behavior of superheavy elements. The relativistic effects will be not so strong (as well as other uncertainties which may be not so large) that really unknown new occurrences should arise.

This statement is not true for the elements beyond element E120. In the superactinides the unknown chemical behavior of five quite loosely bound shells together with the unknown behavior of g-electrons enters and in the region beyond, the structure of the outer electron shells has changed so drastically that only conclusions by analogy can give an idea about the chemical behavior. Even the classifying of these elements to chemically analogous groups is not unambiguous so that the continuation of the Periodic Table can be done by more formal or by more chemical arguments.

The prediction of the chemistry of the elements in the second quasi-stable island is supported only by the calculations which are true within the Dirac-Fock or Dirac-Slater model only. The main question arises if the single particle Dirac equation is still a good equation for $Z > 137$. By taking into account the extended nucleus a formal solution of the Dirac equation is possible up to about $Z = 175$. At this point the 1s level drops into the continuum of electrons with negative energy. If this and how this is possible is up to now unknown. Greiner⁵⁴ suggested that the inner levels will not reach the lower continuum but will go to an Z independent limit which would mean that in nature exists a maximal field strength.

Anyhow, the interaction between the bound levels and the vacuum becomes large so that the bound electrons and the whole vacuum has to be treated together as it was done theoretically by Reinhardt et al.¹⁸ This calculation includes all quantum electrodynamical effects vacuum polarization and

fluctuation, the expressions for which are known in the region $Z\alpha < 1$ only in a perturbation expansion. Also, the effects of retardation, Breit interaction and correlation have to be calculated in addition for large Z elements.

In the region of the first quasistable island those effects will not change the chemistry of the elements. The only differences one would expect are some small changes of the binding energy of the $j = \frac{1}{2}$ electrons. The changes in the region of the second quasistable island of nuclear stability might be larger, however. A first heuristic study was done by Fricke⁵³ by changing the potential near the nucleus drastically where most of the effects are expected to be maximal so that the energy eigenvalue of the $1s$ state was raised by about 30 percent. Although this change was very large, the filling of the shells was only affected between $Z = 167$ and 172 where the $8p_{3/2}$ and $9p_{1/2}$ shells are filled in the opposite order in the new calculation. But this would not change the chemistry significantly. This may be a first indication that the calculations are quite valuable also for the very heavy elements and the coupling between the behavior of the inner electrons and the valence electrons is quite small. With this method also first very preliminary calculations of element E184 were done.

Nevertheless, we hope that when the synthesis of these elements will be complete and that should be in the not too distant future, the results of this type of "computer chemistry" can be compared with the real chemical behavior of superheavy elements.

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