ON THE CHEMISTRY OF SUPERHEAVY ELEMENTS AROUND $Z = 164$ *

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With a relativistic Hartree-Fock-Slater calculation we determined the most stable configurations of the elements of the possibly quasistable island around $Z = 164$. It is found that the expected noble gas at $Z = 168$ should not occur, but instead the element $Z = 164$ should have the properties of a noble gas.

Recent investigations of the stability of superheavy elements indicate the possibility of long lifetimes for elements around $Z = 164$ [1-4]. It is therefore of interest to predict the chemical properties of these elements. Some authors [5-11] presented already calculations for the elements up to $Z = 126$. They found, as expected, the continuation of the periodic system of elements up to $Z = 121$. From $Z = 122$ on the so called superactinide transition series [12] with a mixture of 5g 6f and 7d electron shells is filled [8,10,11].

We have calculated the total energies for various electron configurations of the elements between $Z = 160$ and $Z = 170$ with a relativistic Hartree-Fock-Slater program reported elsewhere [5,7,13]. The wave functions and eigenvalues are taken from the Dirac equation, which can be solved for an extend nucleus for $Z > 137$ [14]. The single particle description of the very low lying states might not be too good because of vacuum-polarization and fluctuation effects which might be large and can not be calculated in this region with standard perturbation techniques [15]. At present, however, we trust the differences of the total atomic energies for various outer electron configurations.

Slaters [16] exchange potential was taken with a factor of 2/3 together with Latters [17] correction, which has been shown to be a very good approximation for very heavy nuclei [18]. The nuclear radius is given by $r = 1.2 \cdot A^{\frac{1}{3}}$ where the nucleon number A is related to the proton number Z by $A = 0.0073\, Z^2 + 0.13\, Z + 63.6$.

For the superheavy nuclei in the region $Z = 160$-170 we calculated 83 different electron configurations for gaseous atoms. The accuracy in the computed energies was $\Delta E/E < 3 \times 10^{-6}$ and in the potential $\Delta V/V < 5 \times 10^{-5}$. This is good enough to trust the total energy up to ± 0.02 Ry, whereas, however, center off mass effects limit the accuracy of this method to about ± 0.1 Ry [8]. The most stable electron configurations for $Z = 160$-170 (from now on called region 3) are given in table 1 together with the total energies. Because these elements are expected to be the chemical analogs of the elements $Z = 78$-88 (from now on called region 1) and $Z = 110$-120 (from now on called region 2) the analogs of the most stable electron configurations of region 1 and 2 are also included.

The 3 most stable configurations of each element are energetically so close that on this basis one can not decide which configuration is the most stable one. However it is interesting to see from table 1 that these 3 most stable configurations are systematically analog to each other. Therefore, by this systematics the filling of these most stable configurations for elements from $Z = 160$-170 will be quite reasonable. This systematics also indicates by comparison with the lower regions that the most stable configurations are *not* analog to those of regions 1 and 2. The main differences are that the $8p_{\frac{1}{2}}$ shell is already filled before the 7d shell is closed and the 9s shell will be prefered over the $8p_{\frac{3}{2}}$ shell. This systematics will be underlined by the calculated eigenvalues of the outer electrons of the elements of the 3 regions as shown in fig. 1. The states of regions 1 and 2 are nearly analog to each other, so that one really can believe that the elements $Z = 110$-120 behave analog to Pt - Ra with not too great deviations from the extrapolation of the periodic system The states of region 3, however, show two important changes: Firstly the very strongly bound

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Table 1
Configuration (plus $Z = 118$ core + $8s^2\ 5g^{18}\ 6f^{14}$)

Z	$7d_{\frac{3}{2}}$	$7d_{\frac{5}{2}}$	$8p_{\frac{1}{2}}$	$8p_{\frac{3}{2}}$	$9s_{\frac{1}{2}}$	$-E_T$ (Ry)
160	4	4	-	-	-	317511.911
	4	3	1	-	-	13.899
	4	-	2	-	2	15.933
	4	1	2	-	1	16.004
	4	2	2	-	-	16.019
161	4	5	-	-	-	326554.610
	4	1	2	-	2	59.198
	4	2	2	-	1	59.337
	4	3	2	-	-	59.368
162	4	6	-	-	-	335869.191
	4	5	1	-	-	71.930
	4	3	2	1	-	74.435
	4	3	2	-	1	74.497
	4	4	2	-	-	74.568
163	4	6	1	-	-	345467.191
	4	4	2	1	-	69.477
	4	4	2	-	1	69.548
	4	5	2	-	-	69.640
164	4	5	2	1	-	355344.144
	4	5	2	-	1	44.198
	4	6	2	-	-	44.466
165	4	5	2	-	2	365499.725
	4	6	2	1	-	99.886
	4	6	2	-	1	99.922
166	4	6	2	2	-	375937.989
	4	6	2	1	1	38.093
	4	6	2	-	2	38.203
167	4	6	2	3	-	386659.628
	4	6	2	2	1	59.758
	4	6	2	1	2	60.070
168	4	6	2	4	-	397665.484
	4	6	2	3	1	65.605
	4	6	2	2	2	65.802
169	4	6	2	4	1	408955.733
	4	6	2	3	2	55.888
170	4	6	2	4	2	420530.281

This table presents the total energies for the most stable configurations of the elements from Z = = 160-170. For comparison the configurations which are expected from the normal continuation of the systematics of the periodic system are also listed.

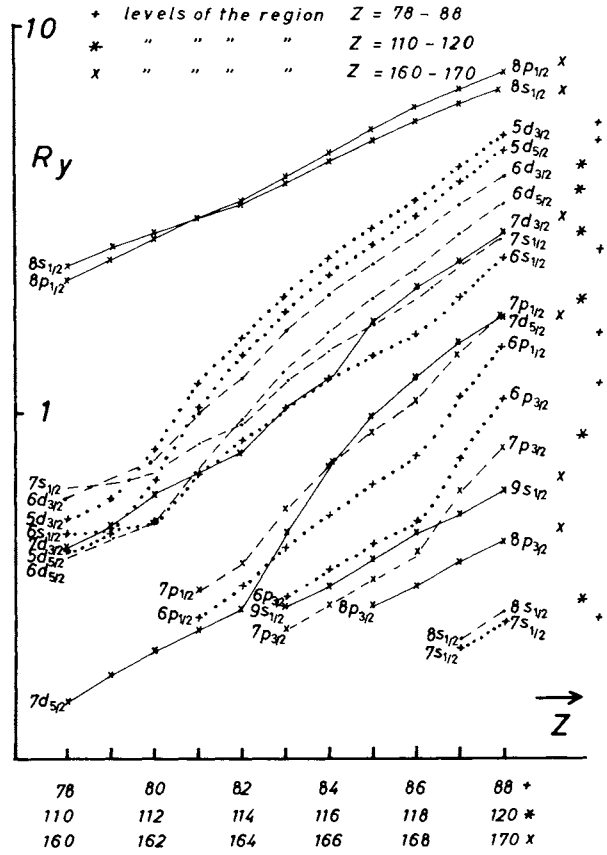

Fig. 1. The binding energies of the outer electron shells of the elements Z = 78-88, Z = 110-120 and Z = 160-170. The levels of the lower and middle region behave analog to each other whereas the upper region has two significant differences. Firstly the very strong bound $8p_{\frac{1}{2}}$ level and secondly the early filling of the 9s level.

$8p_{\frac{1}{2}}$ shell and, secondly, the $9s_{\frac{1}{2}}$ is stronger bound than the $8p_{\frac{3}{2}}$ shell. This last observation means that the expected "noble gas" at Z = 168 is, in fact, none. From the systematics of the periodic system and from the chemical behaviour of the noble gases this is not too astonishing. On the other hand, the very high lying $8p_{\frac{1}{2}}$ level will also be occupied in the region $Z < 160$.

The large separation of the $8p_{\frac{1}{2}}$ and $8p_{\frac{3}{2}}$ shell is obtained from the many body-Dirac equation containing, of course, the spinorbit interaction and the mutual static Coulomb interaction of all the electrons as well as their exchange. A *jj*-coupling calculation could possibly improve these calculations. Nevertheless, an early occupation of the $8p_{\frac{1}{2}}$ shell seems quite reasonable.

The discussion of the oxydation states of the elements from Z = 160-170 is based on the following considerations: The $8s_{\frac{1}{2}}$ level is very strongly bound. In fact, the maximum of its wave function lies well within the radius of the 7d shell. Thus these two electrons will be chemically inactive (in fact, the ionisation potential is $\approx$40 eV) in contrast to the lower region where the 7s and

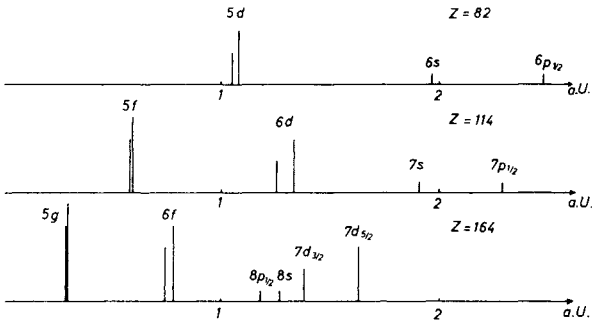

Fig. 2. The principal maxima of the wave functions of the outer electron shells for the elements $Z = 82$ (lead), $Z = 114$ and $Z = 164$. For the first two elements the 6s and $6p_{\frac{1}{2}}$ shells and the 7s and $7p_{\frac{1}{2}}$ shells respectively will be responsible for the chemical activity whereas for $Z = 164$ the 8s and $8p_{\frac{1}{2}}$ shell are shielded by the spherically symmetric filled 7d shell.

6s shell respectively center far outside of the atom with a quite small binding energy (ionisation energy ≈ 7 eV) (see fig. 2). The same arguments are valid for the $8p_{\frac{1}{2}}$ shell. So it is clear that the chemical behaviour for the elements up to $Z = 163$ is solely determined by the open 7d shell. The main oxydation states vary between 2 and 5 for the elements $Z = 160$ up to $Z = 163$ respectively.

From the standpoint of nuclear physics the element $Z = 164$ is the most interesting one. It has a filled 7d shell which will be chemical inactive and because of the tightly bound $8p_{\frac{1}{2}}$ and 8s shells (the radii of these shells are well within the radius of the 7d shell) (see fig. 2) this element seems to have a chemical behaviour as it is expected for a noble gas in this region. This is underlined by the "great" difference of the two most stable configurations of $Z = 164$, e.g. $(7d_{\frac{3}{2}})^4\ (7d_{\frac{5}{2}})^6\ (8p_{\frac{1}{2}})^2$ and $(7d_{\frac{3}{2}})^4\ (7d_{\frac{5}{2}})^5\ (8p_{\frac{1}{2}})^2\ (9s_{\frac{1}{2}})^1$, as shown in table 1. Also the relatively "larger" ionisation potential of ≈ 8 eV (whereas the ionisation potential for $Z = 163$ is ≈ 6 eV and for $Z = 165$ is ≈ 5 eV) supports this interpretation. (Thereby our model gives us for the closed 7d shell certainly a value which is too small).

Moreover the $8p_{\frac{3}{2}}$ and the $9s_{\frac{1}{2}}$ shells are close in energy. The principal maxima of their wave functions are quite outside of the atom (see fig. 2), so that the chemical properties will be determined by these two shells. This suggests that the elements between $Z = 165$ and $Z = 170$ will have normally 1, 2, 3, 4, 5 and 2 as their main oxydation states.

The reason for this different chemical behaviour of these elements from $Z = 160$-170 compared to the elements of the lower regions stems mainly from the fact that 18 electrons of the 5g shell and 14 electrons of the 6f shell are filled in between. The nuclear number has increased so much that the filling of the 7d, 8p and 9s shells is not "normal" as compared to the lower regions because of the very different behaviour of the wave functions and their different shieldings from the electrons of the superactinide series.

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