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THE MADELUNG ENERGY FOR THE STRUCTURE Y/2HFS5.(U)
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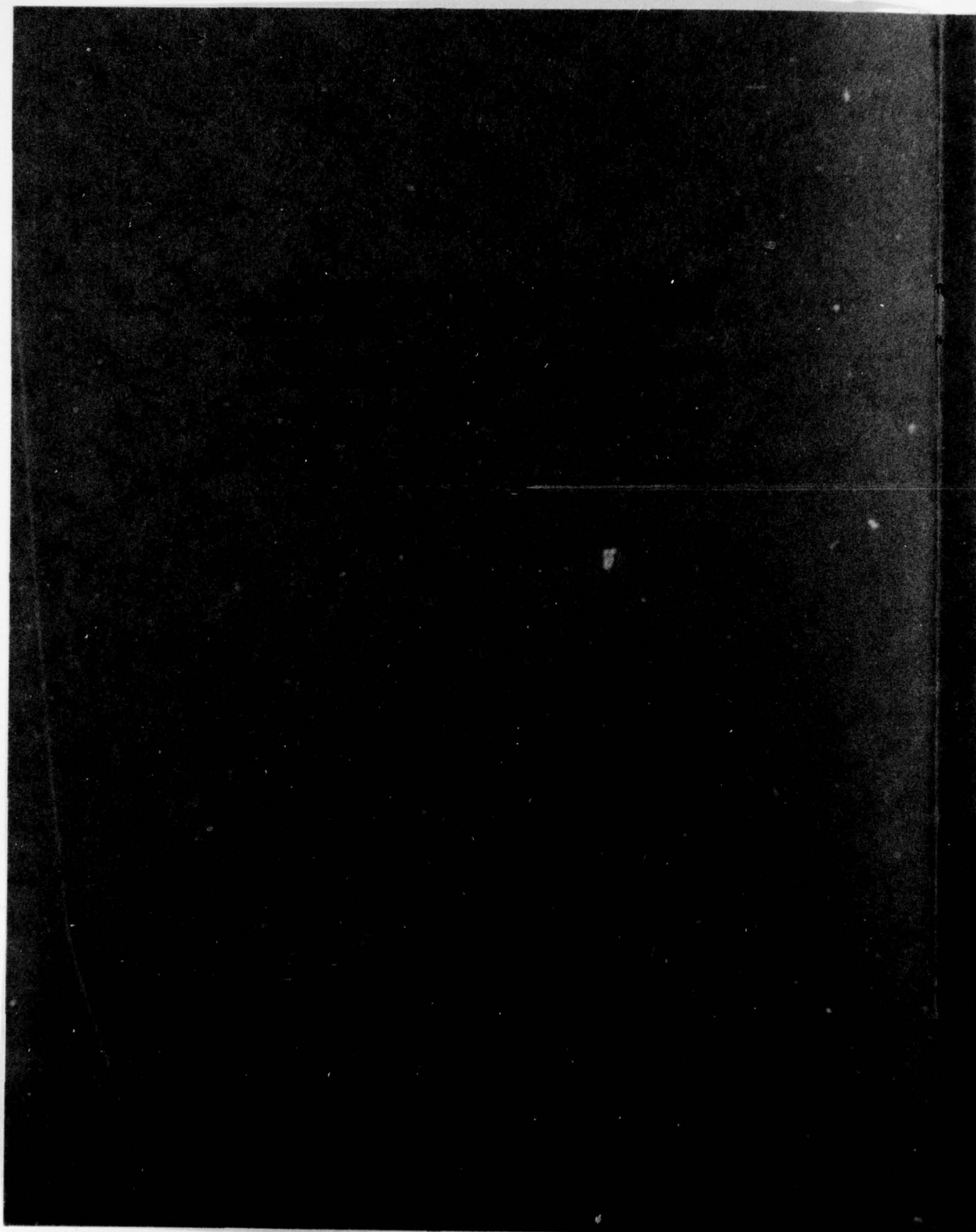| OF |

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The Madelung energy for the compound Y_2HfS_5 is calculated. The results are given as a function of the effective charge, w_E, on the sulfur ion, q_S, as $w_E = -56.495 - 59.420 q_S - 34.167 q_S^2$ with the normal valence of sulfur at $q_S = -2$. Assuming that the exponent in the Born potential is 10, the binding energy, W, of Y_2HfS_5 is found to be $W = -4222.8 - 4441.4 q_S - 2553.8 q_S^2$ kcal/mole.

CONTENTS

1. INTRODUCTION

In the theory of the cohesive energy of ionic or nearly ionic crystals, the electrostatic energy, or Madelung energy, is important. Generally, the electrostatic cohesive force persists, despite the presence of strong repulsive forces of ions of similar charge among the constituent ions. Because of the magnitude of the Madelung energy, it dominates the energy of formation of most ionic solids and therefore is of interest to chemists who are attempting to grow single crystals larger than microscopic powders.

The amount of literature on the various methods of calculating the electrostatic energy of an ionic solid is absolutely overwhelming, but a methodical compilation of the results of these calculations does not seem to exist. Practically every textbook of physical chemistry or solid-state physics lists a number of references to original papers on this subject. Consequently, we list here only references pertinent to the problem at hand: the calculation of the Madelung energy for Y_2HfS_5. The technique used here is elementary; however, it seems not to be in any of the textbooks.

2. CRYSTAL STRUCTURE

The crystal structure of Y_2HfS_5 is, according to Jeitschko and Donohue,[1] orthorhombic, Pnma, which is space group No. 62 in the International Tables of Crystallography.[2] As reported by Jeitschko and Donohue, the cell dimensions and the atomic position of each independent constituent ion are given in table I. The symmetry operations necessary to develop the position of the ions in a single cell are taken from the International Tables of Crystallography.[2] The number of molecular units, Z, in a cell is 4 so that there are 32 independent x, y, and z coordinates necessary to specify the position of all the ions in a single cell. The position of each of the atoms is shown in table II in a coordinate system so that the yttrium ion in position 1 is chosen as the origin and the ions are listed in the sequence chosen by Jeitschko and Donohue.[1] By translation, any lattice point is equivalent to any other point obtained from that point by adding an integer to any or all of its x, y, and z coordinates. Using this rule, one can choose a coordinate system so that $-1/2 \leq$ x or y or z $\leq 1/2$, and this system has been chosen in table II (p. 7).

---

[1]W. Jeitschko and P. C. Donohue, *Acta Crystallogr.*, *B31* (1975), 1890.
[2]*International Tables of Crystallography*, *I*, Kynach Press, Birmingham, England (1952), 151.

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TABLE I. ATOMIC POSITIONS OF CONSTITUENT IONS IN Y_2HfS_5*

Ion	Position	x	y	z
Y	8(d)	0.1778	0.9974	0.0251
Hf	4(m)	0.0060	1/4	0.5742
S_1	8(d)	0.4081	0.0367	0.1630
S_2	4(m)	0.1822	1/4	0.3331
S_3	4(m)	0.5032	1/4	0.5522
S_4	4(m)	0.2921	1/4	0.8152

*Reported in W. Jeitschko and P. C. Donohue, Acta Crystallogr., B31 (1975), 1890.
   Note: The x, y, and z coordinates are in units of a, b, and c, respectively, with a = 11.4585 Å, b = 7.7215 Å, and c = 7.2207 Å.
   The symmetries of the various positions are 8(d), ±(x, y, z), ±(1/2 + x, 1/2 - y, 1/2 - z), ±(x, 1/2 - y, z), ±(1/2 + x, y, 1/2 - z); 4(m), ±(x, 1/4, z), ±(1/2 - x, 3/4, 1/2 + z).

3. COMPUTATION

The electrostatic potential, ϕ, at a site, μ, due to ions of charge, q_ν, in the crystal is given by

$$\phi(\mu) = \sum_{\ell,m,n} \sum_{\nu=1}^{32} \frac{q_\nu}{R_{\ell mn,\nu\mu}} , \qquad (1)$$

where

$$R^2_{\ell mn,\nu\mu} = \left(x_\mu - x_\nu + \ell\right)^2 a^2 + \left(y_\mu - y_\nu + m\right)^2 b^2 + \left(z_\mu - z_\nu + n\right)^2 c^2 ,$$

with ℓ, m, and n integers ranging from positive to negative values large enough so that the sum converges to a unique limit (~10 for four significant digits). The charge on each ion, Z_ν, is in units of the electronic value so that $Z_\nu = eq_\nu$. For the normal valence,

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TABLE II. ATOMIC POSITIONS OF ALL IONS IN UNIT CELL OF SOLID Y_2HfS_5

Ion No. ν	Ion	Coordinate			Charge q_ν
		x_ν	y_ν	z_ν	
1	Y	0.0000	0.0000	0.0000	3
2	Y	0.5000	-0.4948	0.4498	3
3	Y	-0.3556	-0.5000	-0.0502	3
4	Y	0.1444	0.0052	0.5000	3
5	Y	-0.3556	0.0052	-0.0502	3
6	Y	0.1444	-0.5000	0.5000	3
7	Y	0.0000	-0.4948	0.0000	3
8	Y	0.5000	0.0000	0.4498	3
9	Hf	-0.1718	0.2526	-0.4509	4
10	Hf	-0.1838	-0.2474	0.4007	4
11	Hf	0.3162	-0.2474	0.0491	4
12	Hf	0.3282	0.2526	-0.0993	4
13	S_1	0.2303	0.0393	0.1379	-2
14	S_1	-0.2697	0.4659	0.3119	-2
15	S_1	0.4141	-0.4607	-0.1881	-2
16	S_1	-0.0859	-0.0341	-0.3621	-2
17	S_1	0.4141	-0.0341	-0.1881	-2
18	S_1	-0.0859	-0.4607	-0.3621	-2
19	S_1	0.2303	0.4659	0.1379	-2
20	S_1	-0.2697	0.0393	0.3119	-2
21	S_2	0.0044	0.2526	0.3080	-2
22	S_2	-0.3600	-0.2474	-0.3582	-2
23	S_2	0.1400	-0.2474	-0.1920	-2
24	S_2	-0.4956	0.2526	0.1418	-2
25	S_3	0.3254	0.2526	-0.4729	-2
26	S_3	0.3190	-0.2474	0.4227	-2
27	S_3	-0.1810	-0.2474	0.0271	-2
28	S_3	-0.1746	0.2526	-0.0773	-2
29	S_4	0.1143	0.2526	-0.2126	-2
30	S_4	-0.4699	-0.2474	0.1624	-2
31	S_4	0.0301	-0.2474	0.2874	-2
32	S_4	-0.3587	0.2526	-0.3376	-2

Note: The x, y, and z coordinates are in units of a, b, and c so that the actual distance from ion 1 to any ion, ν, is $\left(a^2 x_\nu^2 + b^2 y_\nu^2 + c^2 z_\nu^2\right)^{\frac{1}{2}}$.

$$q_\nu = 3, \quad 1 \le \nu \le 8 \quad (Y^{3+}),$$

$$q_\nu = 4, \quad 9 \le \nu \le 12 \quad (Hf^{4+}),$$

$$q_\nu = -2 \quad 13 \le \nu \le 32 \quad (S^{2-}),$$

and $\sum_{\nu=1}^{32} q\nu = 0$ for neutrality of the entire solid. The sum given by equation (1) was calculated for each of the different ions (Y, Hf, S_1, S_2, S_3, and S_4) given in table I. The sum was done so that, for each value of ℓ, m, and n, the sum over all the constituent ions in a cell was done first. The results of this calculation are shown in table III. As a check on the convergence of the sum for a given site, μ was computed and compared to a site that is identical, that is, $\phi(1)$

TABLE III. ELECTRIC POTENTIAL AT VARIOUS SITES IN
Y_2HfS_5; UNITS ARE $(\text{Å})^{-1}$

Ion	Type μ	$\phi(\mu)$, $q_S = -2$	$\phi(\mu)$, $q_S = 0$	ϕ_1
Y	1	-2.2256	-1.7304	0.24760
Y	5	-2.2256	-1.7302	0.24770
Hf	9	-2.4535	2.9775	2.71555
S_1	13	1.4336	-0.72753	-1.08057
S_1	17	1.4337	-0.72777	-1.08074
S_2	21	1.4088	0.13992	-0.63444
S_3	25	1.1192	1.1824	0.03160
S_4	29	1.6004	0.11696	-0.74172

Note: The charge on yttrium ion q_Y is +3, while the charge on the hafnium ion is q_{Hf} chosen so that $q_{Hf} + 5q_S = -6$, where q_S is the charge on the sulfur ion. The last column is the constant in the expression $\phi(\mu) = \phi_0(\mu) + q_S\phi_1$.

and $\phi(2)$ or $\phi(9)$ and $\phi(10)$, and the sum was extended until the agreement was satisfactory. A limit of ± 10 on the ℓ, m, and n sums gave agreement to four digits. The total electrostatic energy, U_E, for a cell is given by

$$U_E = \frac{e^2}{2} \sum_{i=1}^{6} q_i N_i \phi(\mu_i) , \qquad (2)$$

where N_i is the number of each type of ion (such as $q_i = 3$, $N_i = 8$ for Y and $q_i = 4$, $N_i = 4$ for Hf) and μ covers the distinct types of ions.

In the discussion of the structure, Jeitschko and Donohue[1] state that the compound Y_2HfS_5 is semi-ionic. That is, the yttrium site is ionic along with the complex $(HfS_5)^{6-}$, but within this latter complex the hafnium and sulfur may be covalent. To account partially for this covalency, one can choose an effective charge on the sulfur ion, q_S, so that

$$q_{Hf} + 5q_S = -6 , \qquad (3)$$

which retains the ionic character of the HfS_5 complex. Also, the lattice sum was performed by using $q_S = -0$ ($q_{Hf} = -6$, not realistic), and the results are given in table III. Since the electrostatic potential is linear in the charge, each potential can be written in the form

$$\phi(\mu) = \phi_0(\mu) + q_S \phi_1(\mu) \qquad (4)$$

for each distinct site. In the simple compounds where the x-ray data depend on only one parameter such as "a," the cell size in a simple cubic material, the electrostatic potential is generally expressed as

$$\phi = - \frac{eM}{a} , \qquad (5)$$

where M is the Madelung constant, which has been tabulated for most of the simpler cubic structures.[3] In the more complicated structures, an effective Madelung constant can be defined by

[1]W. Jeitschko and P. C. Donohue, Acta Crystallogr., B31 (1975), 1890.
[3]Mendel Sachs, Solid State Theory, McGraw-Hill Book Co., New York (1963), 55.

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$$M_\mu = \phi(\mu) v^{1/3} \; , \tag{6}$$

where $\phi(\mu)$ is given by Jeitschko and Donohue,[1] and v is the volume of the unit cell. We do not use this definition here.

If one chooses a solid of dimension A, B, and C lengths in the x, y, and z dimensions, respectively, then the number of cells in its volume is given by

$$N = \left(\frac{A}{a}\right)\left(\frac{B}{b}\right)\left(\frac{C}{c}\right) \; , \tag{7}$$

and the total energy per unit volume, w_E, is

$$w_E = \frac{U_E}{v} \; , \tag{8}$$

where we have used equation (2) and $v = abc$.

In many calculations, where the complete binding energy is required, one must assume some form of a repulsive force. Many different types are assumed in the literature with various degrees of sophistication. We assume the simple Born potential, B/r^n, where B is a constant and n is determined from experimental data.[4] If the material is assumed isotropic, then both the electrostatic energy and the repulsive energy can be written in terms of a single variable such as $v^{1/3}$, where v is the volume of the unit cell. The condition that the energy is a minimum then requires that the derivative of the energy vanish at the equilibrium value of v. If it does, then

$$w = w_E\left(1 - \frac{1}{n}\right) \; , \tag{9}$$

with w_E given by equation (8).

It is customary to express the energy in kilocalories per mole, W_E; for Y_2HfS_5, this conversion of the various constants gives

[1] W. Jeitschko and P. C. Donohue, Acta Crystallogr., B31 (1975), 1890.
[4] A. J. Dekker, Solid State Physics, Prentice-Hall, Inc., Englewood Cliffs, NJ (1965), Ch 5.

$$W_E = 83.05w_E \tag{10}$$

if w_E is in reciprocal angstroms. Using the results given in table III, one can write electrostatic energy as

$$w_E(q_s) = -56.495 - 59.420q_s - 34.167q_s^2 . \tag{11}$$

The result given in equation (11) for several values of q_s is $w_E(-2) = -74.325$, $w_E(-1) = -32.240$, and $w_E = -40.275$, all in reciprocal angstroms. With the result of equation (11) substituted in equations (10) and (9), one finally has for W, the total energy,

$$W = 83.05\left(1 - \frac{1}{n}\right)\left(-56.495 - 59.420q_s - 34.167q_s^2\right) \text{ kcal/mole.} \tag{12}$$

Generally, the best fit value of n is obtained by matching the theoretical and experimental values, and in simple compounds the resulting value of n varies from 6 to 12. Choosing a value of n = 10 in equation (12), one obtains

$$W = -4222.8 - 4441.4q_s - 2553.8q_s^2 \text{ kcal/mole .} \tag{13}$$

This gives the lattice energy frequently used in the Born-Haber cycle calculations.[5]

Occasionally, the lattice energy is calculated by assuming that the solid is formed from preexisting ionic complexes.[6] A reasonable approach from this assumption is $2Y^{3+} + (HfS_5)^{6-}$, which forms the solid. In this approach, one needs the energy of formation, U, of a single $(HfS_5)^{6-}$. This energy as a function of q_s is

$$U\left[(HfS_5)^{6-}\right] = -46.709q_s - 28.332q_s^2 (\overset{\circ}{A})^{-1} . \tag{14}$$

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[5]F. A. Cotton, *Advanced Inorganic Chemistry*, *Interscience Publishers*, *New York* (1972), 61.

[6]A. B. Blake and F. A. Cotton, *Inorganic Chemistry*, <u>2</u> (1963), 906.

If this result is subtracted from equation (11) and the result is multiplied by $83.05[1 - (1/n)]$, then the energy of formation of the solid Y_2HfS_5 from the constituent ionic complexes $2Y^{3+}$ and $(HfS_5)^{6-}$ is

$$U = -4222.8 - 948.74q_s - 436.14q_s^2 \text{ kcal/mole} . \tag{15}$$

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ACKNOWLEDGEMENT

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