A new approach to describe elemental-property parameters

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The atomic number (AN) of the elements together with their 'periodic number' (PN) were found to form an efficient pair for the discussion of metallurgical and structural problems. The periodic number PN represents a different enumeration of the elements, emphasizing the role of the valence electrons. In contrast to the atomic number, PN depends in details on the underlying Periodic Table of the elements. As a first result we describe the elemental-property parameters 'atomic size SZ_a' and 'atomic reactivity RE_a', derived from fits to various experimental and theoretical data sets. These two parameters can be approximated as simple functions of AN and PN:

$$SZ_a = k_{SZ} [\log (AN + 1)] [k_{PN} - (\log PN)^3],$$
$$RE_a = k_{RE} \{[\log (AN + 1)] [k_{PN} - (\log PN)^3]\}^{-1} = k_{SZ} k_{RE} (SZ_a)^{-1},$$

where k_{PN} is a scaling factor, and k_{SZ}, k_{RE} are fit parameters for a fit to experimental data. We argue that all elemental-property parameter patterns are derived from AN and PN. AN and PN represent fundamental elemental-property parameters independent from each other. Any pattern, which shows well-defined functional behavior within each group number GN, as well as within each main quantum number QN, can be included. On the example of compound formers/non-formers in binary, ternary and quaternary chemical systems we demonstrate that a quantitative link exists between material properties and AN, PN (or simple functions of both) of the constituent elements.

Electronegativity / Atomic radii / Chemical elements

Introduction

Most of the chemical elements are well characterized by quite a number of experimental and theoretical elemental-property parameters, such as the atomic number AN, experimental and theoretical radii, ionization potential, bulk modulus, melting temperature, etc. On plotting these elemental-property parameters versus AN respectively PN, a vast majority of them can be grouped into distinctly different patterns. As examples, three of these patterns are shown in Figs. 1-3.

Elemental-property parameters belonging to the same pattern are qualitatively similar, although they are quantitatively different. The elemental-property parameters used in this work are listed in Tables 1a and 1b. In Table 1a the two fundamental pattern groups and in Table 1b four examples of derived pattern groups are given.

Our investigations revealed that the elemental-property parameters not only characterize the

chemical elements themselves but also reflect quantitatively their interaction behavior between each other, which thus opens the possibility to predict physical or chemical properties of multinary systems.

Fundamental elemental-property parameters: Atomic Number AN and Periodic Number PN

Looking closer at the data sets listed in Tables 1a and 1b we notice that integer parameter values are only found in the first two patterns (Table 1a). The only comprehensive ones are AN and PN. The atomic number AN and the periodic number PN (the possibly misleading name for PN was chosen to have a neutral designation for different variants of this parameter) define, in a different way, the position of each element in the Periodic System (or Periodic Table) of the elements. AN is a simple count of the protons of the nucleus, which is equal to the total number of electrons of this element. The periodic number PN is
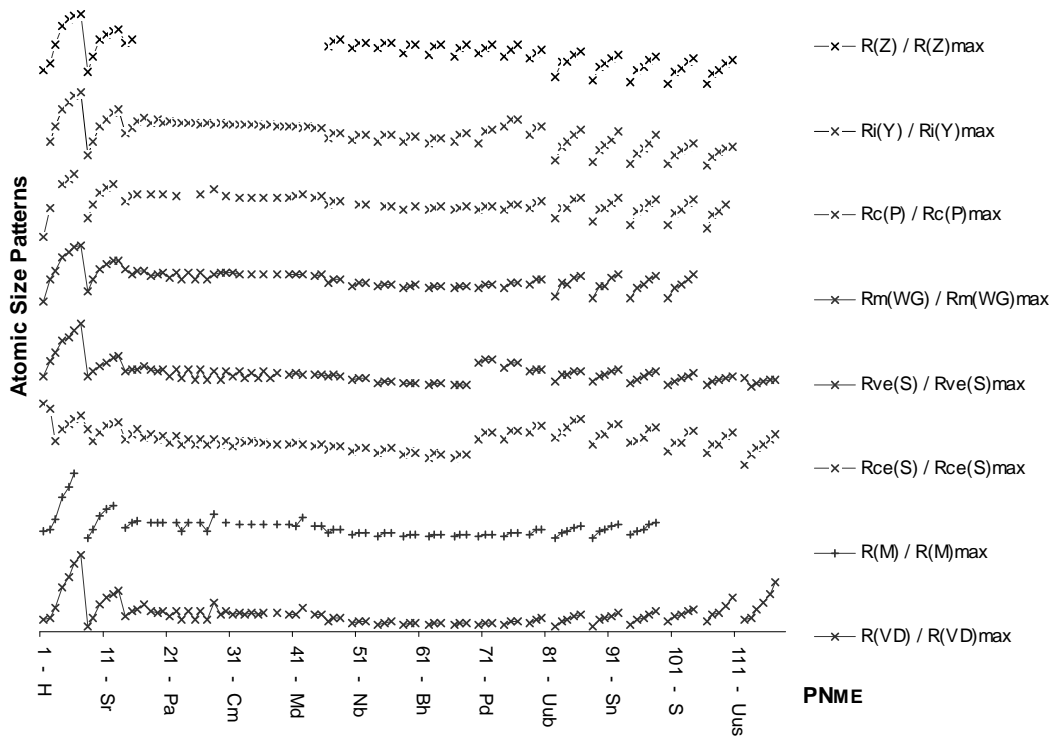
Fig. 1 Eight elemental-property parameters (normalized to their maximum values) belonging to the atomic-size group (Table 1b), plotted versus periodic number PN_{ME}.
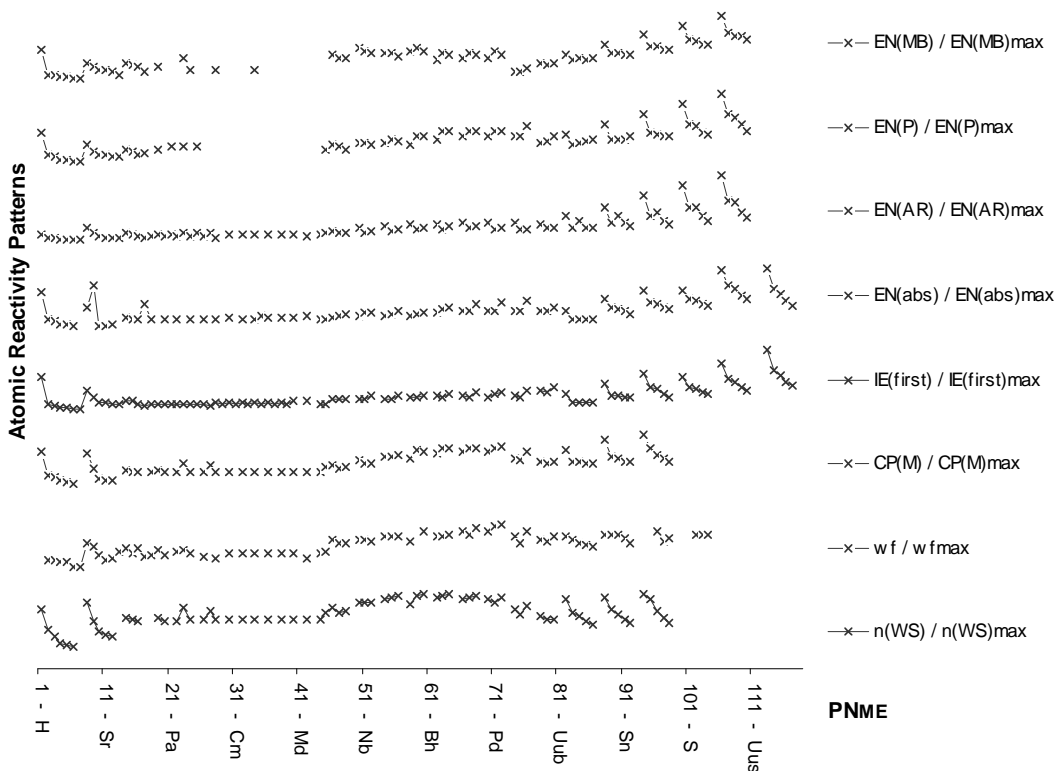

Fig. 2 Eight elemental-property parameters (normalized to their maximum values) belonging to the atomic-reactivity group (Table 1b), plotted versus periodic number PN_{ME}.

Table 1 (a) The 13 elemental-property parameters considered grouped into the two fundamental groups, the atomic number AN pattern group and the periodic number PN pattern group. (b) The 26 elemental-property parameters considered grouped into the four derived groups, the atomic size pattern group, the atomic reactivity pattern group, the atomic affinity pattern group, and the atomic density pattern group.

(a)

I) Atomic number pattern group

AN: Atomic number
QN: Main quantum number
AM: Atomic mass [1]
nc(C): Nuclear effective charge according to Clementi [2]
nc(S): Nuclear effective charge according to Slater [3]
cma(Mo): Coefficient of mass attenuation for Mo Kα [2]
cma(Cu): Coefficient of mass attenuation for Cu Kα [2]
aes: Atomic electron scattering factor [2]

II) Periodic number pattern group

PN_{ME}: Periodic number according to Meyer [4]
PN_{MD}: Periodic number according to Mendeleyev [5]
PN(P): Periodic number MN according to Pettifor's sequence [6]
VE: Valence electron number
GN: Group number

(b)

i) Atomic size pattern group

R(Z): Pseudo-potential radii according to Zunger [7]
Ri(Y): Ionic radii according to Yagoda [2]
Rc(P): Covalent radii according to Pauling [2]
Rm(WG): Metal radii according to Waber and Gschneidner [8]
Rve(S): Valence electron distance according to Schubert [9]
Rce(S): Core electron distance according to Schubert [9]
R(M): Radii according to Miedema (derived from his $V^{2/3}$ compilation) [10]
R(VD): Radii according to Villars and Daams (derived from their V compilation) [11]

ii) Atomic reactivity pattern group

EN(MB): Electronegativity according to Martynov and Batsanov [12]
EN(P): Electronegativity according to Pauling [1]
EN(AR): Electronegativity according to Allred and Rochow [2]
EN(abs): Absolute electronegativity [9]
IE(first): First ionization energy [1]
CP(M): Chemical potential according to Miedema [10]
wf: Work function [1]
n(WS): n(Wigner and Seitz) according to Miedema [10]

iii) Atomic affinity pattern group

T_{fus}: Temperature of melting [1]
T_{vap}: Temperature of boiling [1]
$\triangle H_{vap}$: Enthalpy of vaporization [1]
$\triangle H_{fus}$: Enthalpy of melting [1]
$\triangle H_{atom}$: Enthalpy of atomization [1]
$\triangle H_{surf}(M)$: Surface energy according to Miedema [10]
CE(B): Cohesion energy according to Brewer [13]
VC: Isothermal volume compressibility [13]

iv) Atomic density pattern group

D_X: X-ray density [1]
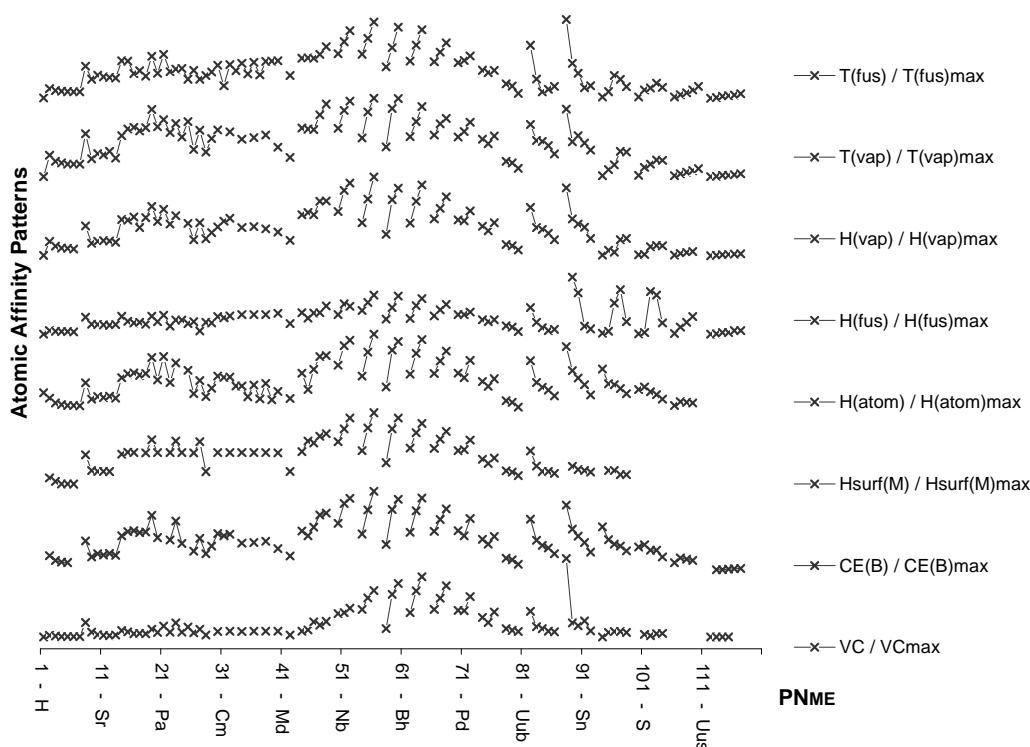D_m: measured density [1]

Fig. 3 Eight elemental-property parameters (normalized to their maximum values) belonging to the atomic-affinity group (Table 1b), plotted versus periodic number PN_{ME}.

the result of a slightly more differentiated enumeration of the elements, since it emphasizes the role of the valence electrons, i.e. it implicitly reflects the construction principle of the Periodic System, which is less obvious from AN. In other words, AN is the sequence number of the chemical elements within the Periodic Table obtained by counting in first priority within the same main quantum number QN with increasing group number GN, and in second priority with increasing main quantum number QN. AN sequence: H, He (QN 1), Li, Be, B, C, N, O, F, Ne (QN 2), Na, Mg, …. Ar (QN 3), K, Ca, … Kr (QN 4), etc. Smooth functions of AN are obtained, e.g. for the radii of maximum radial charge density or for the energies of innermost electrons, such as 1s, 2s, but not for outer electrons 4s, 5s, 5p. For most properties the problems begin with QN 4, where the occurrence of the 3d electrons leads to irregular valences and to Groups IA, IIA and IB, IIB in Meyer's Periodic Table, where IIA and IIB elements both possess two valence electrons.

For the periodic number PN the enumeration of the elements is performed in first priority within the same group number GN with increasing main quantum number QN, and in second priority with increasing group number GN. The familiar group names of the Periodic System of Meyer (Groups IA, IIA, IIIB, IVB, VB, VIB, VIIB, VIIIB (for the Fe, Co and Ni groups), IB, IIB, IIIA, IVA, VA, VIA, VIIA, VIIIA) are replaced by GN 1, 2, … 18 (18 = maximum number of s, p, and d electrons, 2 + 6 + 10). The details, however, depend on the arrangement of the underlying Periodic Table.

In the representation of Sanderson [14], e.g. the transition elements (including Cu, Ag and Au) are listed as insert below Be, Mg and between Ca, Sr, Ba, Ra and Zn, Cd, Hg, in the same way as the rare-earth elements Ce…Yb and the actinide elements Th…No are inserted below Sc, Y and between La, Ac and Lu, Lr. We mention this kind of Periodic Table because it visualizes the uncertainty in placing the elements Be, Mg, which are found above Ca, Sr, Ba, Ra in Meyer's Periodic Table [4], but above Zn, Cd, Hg in Mendeleyev's Periodic Table [5]. This rises the question whether the rare-earth elements and above all the actinide elements also should obtain proper group numbers (all the more as the early actinides really behave like transition elements).

A possibly more important question is that about the placement of the hydrogen atom, H. For a discussion of properties of non-metallic compounds a better placement of H is certainly above fluor, F (i.e. above the halogen group) as in Mendeleyev's Periodic Table, because the observed valence states of H are +1 and –1, in contrast to the elements of Group 1, Li, Na,…Fr, which can act only as cations.

The first time that a periodic-number-like scale was used as elemental-property parameter [15,16], it was employed as a kind of phenomenological scale. In a more recent work [17] several variants of PN were used with the aim to separate compound-forming systems from non-formers in plots of 2D- and 3D-elemental-property parameter expressions (also called features). Some variants were found to be of similar

efficiency. Still another variant was recently successfully applied in order to separate different atomic-environment types (AET) of the constituents in equiatomic binary compounds [18]. In most of the above cases the periodic-number-like scale or PN was then called 'Mendeleev number'.

In the following, we restrict our considerations to two basic variants, the "physicist's version" based on the Periodic Table of L. Meyer [4] and the "chemist's version" based on the Periodic Table of D.I. Mendeleyev [5] (the designation is not a judgment). The shift of H from $(AN, PN_{ME}) = (1, 1)$ to $(AN, PN_{MD}) = (1, 105)$ is the reason why only the elements with PN > 105 keep the same (AN, PN) pair numbers in both systems, i.e. PN(Meyer) ≠ PN(Mendeleyev) for H and all elements with GNs 1-16.

In order to distinguish between the two versions we name the particular periodic number PN derived from Meyer's Periodic Table the Meyer number PN_{ME}, and that derived from Mendeleyev's Periodic Table the Mendeleyev number PN_{MD}.

In Figs. 4, 5 we show the step-like functions $PN_{ME} / (PN_{ME})_{max}$ and GN / GN_{max} versus AN, and the reversed version AN / AN_{max} and QN / QN_{max} versus PN_{ME}, where $AN_{max} = PN_{max} = 118$, $GN_{max} = 18$,

$QN_{max} = 7$ for the elements known up to now together with some predicted ones.

Derived elemental-property parameters: Atomic Size SZ_a and its reciprocal, the Atomic Reactivity RE_a, as functions of AN and PN

In Figs. 1, 2 eight different data sets are plotted as functions of PN_{ME}. Within each pattern group the eight data sets show rather similar functional behavior (patterns) although they differ quantitatively from each other. Each pattern group shows well-defined functional behavior within each group number GN, as well as within each main quantum number QN.

Encouraged by this experience and after observing by chance that in a first approximation Ri(Yagoda) ≈ (PN – GN), we started studying functions like $(PN / PN_{max})^x$, $(\log PN / \log PN_{max})^x$, $\{[k – PN] / [k – PN_{max}]\}^x$, $\{[k – (\log PN)] / [k – (\log PN_{max})]\}^x$,… versus AN and the analogous functions of AN versus PN, $x = \pm 1, \pm 2,…$ and $k = (PN_{max})^x$ or $(AN_{max})^x$, respectively $(\log PN_{max})^x$ or $(\log AN_{max})^x$, with the aim to reproduce elemental-property parameters by a combination of such simple functions of AN and PN.
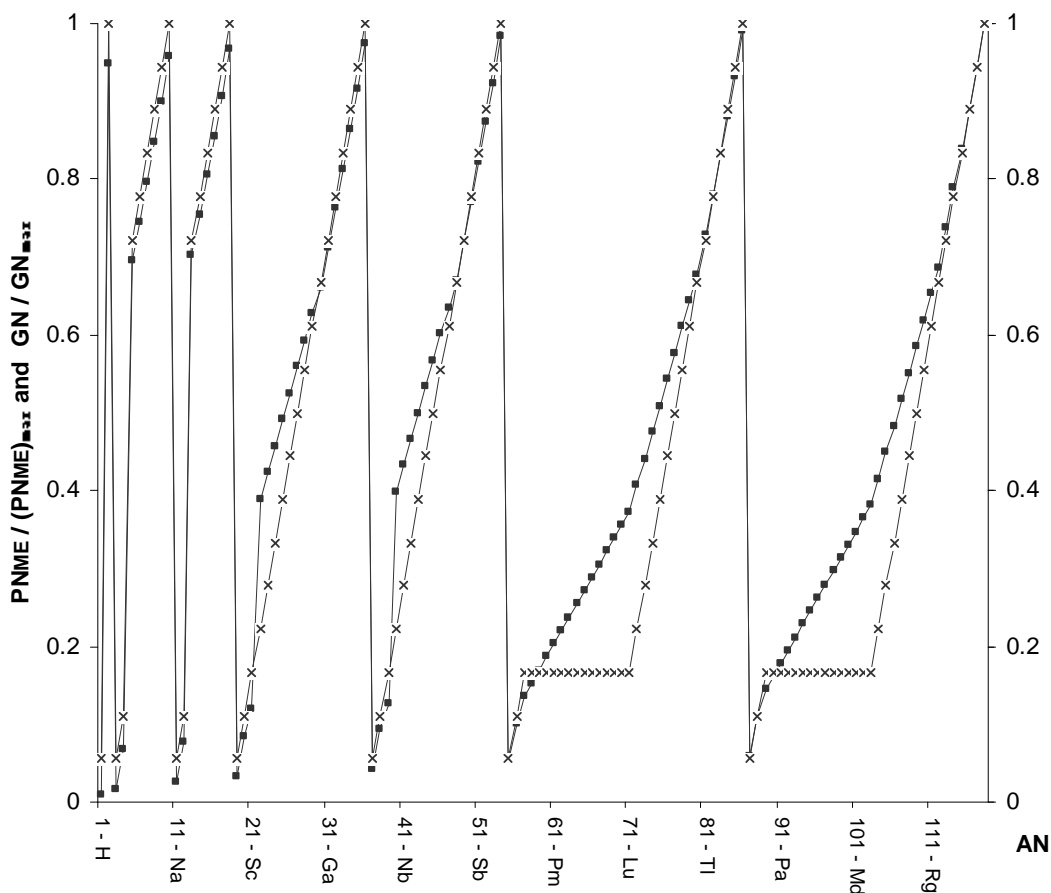

Fig. 4 Periodic number $PN_{ME} / (PN_{ME})_{max}$ and group number GN / GN_{max} versus atomic number AN; ■ $PN_{ME} / (PN_{ME})_{max}$, ✕ GN / GN_{max}.
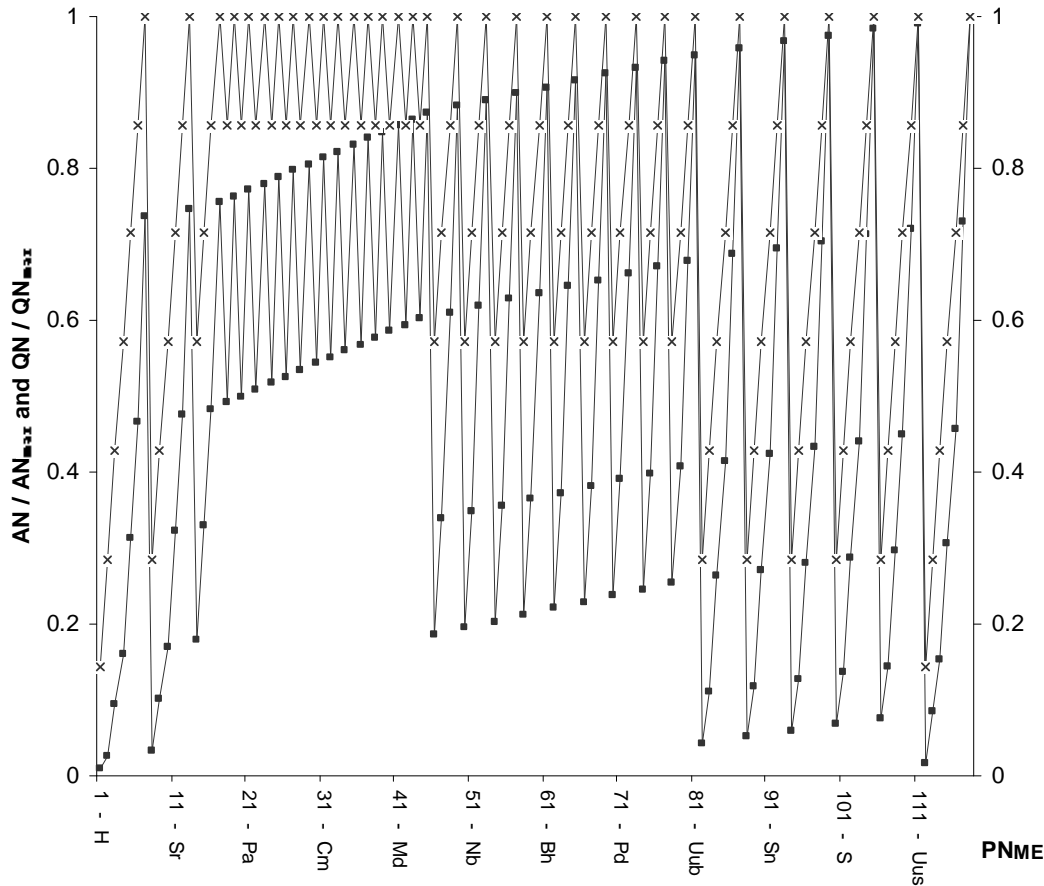
Fig. 5 Atomic number AN / AN$_{max}$ and main quantum number QN / QN$_{max}$ versus periodic number PN$_{ME}$; ■ AN / AN$_{max}$, × QN / QN$_{max}$.

A systematic trial and error approach led us finally to the following approximations for the atomic size SZ$_a$ and the atomic reactivity RE$_a$:

$$SZ_a = k_{SZ} [\log (AN + 1)] [k_{PN} - (\log PN)^3] \quad (1)$$

$$RE_a = k_{RE} \{[\log (AN + 1)] [k_{PN} - (\log PN)^3]\}^{-1} = k_{RE} k_{SZ} (SZ_a)^{-1}. \quad (2)$$

The scaling number k$_{PN}$ turned out to be defined by

$$k_{PN} = \log (AN + 1)_{max} + (\log PN_{max})^3 = 10.9695,$$

for PN$_{ME}$ and PN$_{MD}$.

The factors k$_{SZ}$ and k$_{RE}$ are fitting parameters for an adaptation to experimental or theoretical data sets. In Tables 2a and 2b we avoided this fitting by introducing normalized parameters:

$$SZ_a / (SZ_a)_{max} = SZ_a / SZ_a (Fr),$$

here

$$SZ_{aME} (Fr) = k_{SZME} (\log 88) [k_{PN} - (\log 7)^3] = 20.1564 \, k_{SZME}$$

for the Meyer Table, and

$$SZ_{aMD} (Fr) = k_{SZMD} (\log 88) [k_{PN} - (\log 6)^3] = 20.4138 \, k_{SZMD}$$

for the Mendeleyev Table.

For the normalization of RE$_{aME}$ and RE$_{aMD}$ we did not choose RE$_{aME}$ (He) and RE$_{aMD}$ (He) respectively, which appeared to be too high, but we chose

$$RE_{aME} (Ne) = RE_{aMD} (Ne)$$

$$RE_a / RE_a (Ne) = SZ_a (Ne) / SZ_a = (\log 11) [k_{PN} - (\log 113)^3] / [\log (AN+1)] [k_{PN} - (\log PN)^3]$$

or

$$RE_{aME} / RE_{aME} (Ne) = 2.4114 \{[\log (AN + 1)] [k_{PN} - (\log PN_{ME})^3]\}^{-1}$$

for Meyer's Periodic Table. The analogous equation based on Mendeleyev's Periodic Table differs only in the log PN term:

$$RE_{aMD} / RE_{aMD} (Ne) = 2.4114 \{[\log (AN + 1)] [k_{PN} - (\log PN_{MD})^3]\}^{-1}.$$

Table 2a gives AN, PN$_{ME}$, SZ$_{aME}$ / (SZ$_{aME}$)$_{max}$ = SZ$_{aME}$ / SZ$_{aME}$ (Fr), and RE$_{aME}$ / RE$_{aME}$ (Ne) in Meyer's Periodic Table, while in Table 2b the corresponding values AN, PN$_{MD}$, SZ$_{aMD}$ / (SZ$_{aMD}$)$_{max}$ = SZ$_{aMD}$ / SZ$_{aMD}$ (Fr), and RE$_{aMD}$ / RE$_{aMD}$ (Ne) are listed according to Mendeleyev's Periodic Table. Figs. 6 and 7 display SZ$_{aME}$ / (SZ$_{aME}$)$_{max}$ and RE$_{aME}$ / RE$_{aME}$ (Ne) as function of PN$_{ME}$ and AN, respectively. As a consequence of our normalization with (Ne) instead of (He) the values for RE$_{aME}$ (Ne) / RE$_{aME}$ (He) (RE$_{aMD}$ (Ne) / RE$_{aMD}$ (He)) = 2.13768. In the Figs. 6, 7, 9, 11, where this value is used, we set it to 1.2 for representational reasons.

Table 2a Atomic number AN, periodic number PN_{ME}, atomic size $SZ_{aME} / (SZ_{aME})_{max}$ and atomic reactivity $RE_{aME} / (RE_{aME})_{max}$ using Meyer's Periodic System.

Legend of each cell:

AN	PN_{ME}
$SZ_{aME} / (SZ_{aME})_{max}$	
$RE_{aME} / (RE_{aME})_{max}$	

Main table (GN = group number, periods 1–7; each cell: AN · Symbol · PN_{ME} / $SZ_{aME}/(SZ_{aME})_{max}$ / $RE_{aME}/(RE_{aME})_{max}$):

GN →	IA 1	IIA 2	IIIB 3	IVB 4	VB 5	VIB 6	VIIB 7	VIIIB 8	VIIIB 9	VIIIB 10	IB 11	IIB 12	IIIA 13	IVA 14	VA 15	VIA 16	VIIA 17	VIIIA 18
1	1 H 1 0.16383 0.73024																	2 He 112 0.05596 2.13768
2	3 Li 2 0.32684 0.36603	4 Be 8 0.35485 0.33713											5 B 82 0.15287 0.78257	6 C 88 0.15167 0.78879	7 N 94 0.14730 0.81216	8 O 100 0.14058 0.85098	9 F 106 0.13207 0.90585	10 Ne 113 0.11963 1.0000
3	11 Na 3 0.58150 0.20573	12 Mg 9 0.55821 0.21431											13 Al 83 0.22186 0.53922	14 Si 89 0.20781 0.57567	15 P 95 0.19319 0.61926	16 S 101 0.17810 0.67171	17 Cl 107 0.16265 0.73553	18 Ar 114 0.14382 0.83180
4	19 K 4 0.69396 0.17239	20 Ca 10 0.65398 0.18293	21 Sc 14 0.63030 0.18980	22 Ti 46 0.43051 0.27789	23 V 50 0.41533 0.28804	24 Cr 54 0.40019 0.29894	25 Mn 58 0.38510 0.31065	26 Fe 62 0.37006 0.32328	27 Co 66 0.35507 0.33693	28 Ni 70 0.34013 0.35172	29 Cu 74 0.32526 0.36781	30 Zn 104 0.31044 0.38536	31 Ga 78 0.28706 0.41675	32 Ge 90 0.26414 0.45291	33 As 96 0.24164 0.49508	34 Se 102 0.21954 0.54493	35 Br 108 0.19779 0.60484	36 Kr 115 0.17263 0.69301
5	37 Rb 5 0.83298 0.14362	38 Sr 11 0.77674 0.15402	39 Y 15 0.74258 0.16110	40 Zr 47 0.50364 0.23753	41 Nb 51 0.48244 0.24797	42 Mo 55 0.46178 0.25907	43 Tc 59 0.44162 0.27090	44 Ru 63 0.42190 0.28356	45 Rh 67 0.40260 0.29715	46 Pd 71 0.38367 0.31181	47 Ag 75 0.36509 0.32768	48 Cd 79 0.34684 0.34493	49 In 85 0.31920 0.37479	50 Sn 91 0.29236 0.40920	51 Sb 97 0.26623 0.44935	52 Te 103 0.24076 0.49689	53 I 109 0.21589 0.55412	54 Xe 116 0.18743 0.63826
6	55 Cs 6 0.91053 0.13139	56 Ba 12 0.84609 0.14139	57 La 16 0.80695 0.14825	72 Hf 48 0.57475 0.20815	73 Ta 52 0.54867 0.21804	74 W 56 0.52343 0.22855	75 Re 60 0.49896 0.23976	76 Os 64 0.47519 0.25176	77 Ir 68 0.45206 0.26464	78 Pt 72 0.42952 0.27853	79 Au 76 0.40752 0.29356	80 Hg 80 0.38602 0.30991	81 Tl 86 0.35416 0.33779	82 Pb 92 0.32335 0.36997	83 Bi 98 0.29350 0.40761	84 Po 104 0.26452 0.45227	85 At 110 0.23633 0.50621	86 Rn 117 0.20429 0.58560
7	87 Fr 7 1.0000 0.11963	88 Ra 13 0.92721 0.12902	89 Ac 17 0.88292 0.13550	104 Rf 49 0.61579 0.19427	105 Db 53 0.58710 0.20377	106 Sg 57 0.55939 0.21386	107 Bh 61 0.53255 0.22464	108 Hs 65 0.50653 0.23618	109 Mt 69 0.48124 0.24859	110 Ds 73 0.45664 0.26199	111 Rg 77 0.43267 0.27650	112 Uub 81 0.40928 0.29230	113 Uut 87 0.37488 0.31913	114 Uuq 93 0.34165 0.35016	115 Uup 99 0.30950 0.38654	116 Uuh 105 0.27832 0.42983	117 Uus 111 0.24806 0.48228	118 Uuo 118 0.21372 0.55975

Lanthanides (QN **6**):

57 La 16 0.80695 0.14825	58 Ce 18 0.78996 0.15144	59 Pr 20 0.77343 0.15468	60 Nd 22 0.75734 0.15796	61 Pm 24 0.74165 0.16131	62 Sm 26 0.72634 0.16471	63 Eu 28 0.71138 0.16817	64 Gd 30 0.69675 0.17170	65 Tb 32 0.68242 0.17531	66 Dy 34 0.66837 0.17899	67 Ho 36 0.65459 0.18276	68 Er 38 0.64105 0.18662	69 Tm 40 0.62774 0.19058	70 Yb 42 0.61466 0.19463	71 Lu 44 0.60177 0.19880

Actinides (QN **7**):

89 Ac 17 0.88292 0.13550	90 Th 19 0.86292 0.13864	91 Pa 21 0.84352 0.14183	92 U 23 0.82469 0.14506	93 Np 25 0.80639 0.14836	94 Pu 27 0.78857 0.15171	95 Am 29 0.77122 0.15512	96 Cm 31 0.75429 0.15860	97 Bk 33 0.73775 0.16216	98 Cf 35 0.72159 0.16579	99 Es 37 0.70577 0.16951	100 Fm 39 0.69028 0.17331	101 Md 41 0.67509 0.17721	102 No 43 0.66019 0.18121	103 Lr 45 0.64556 0.18532

Table 2b Atomic number AN, periodic number PN_{MD}, atomic size $SZ_{aMD} / (SZ_{aMD})_{max}$ and atomic reactivity $RE_{aMD} / (RE_{aMD})_{max}$ using Mendeleyev's Periodic System.

Legend (per cell):
$$\text{AN} \quad \text{Symbol} \quad PN_{MD}$$
$$SZ_{aMD} / (SZ_{aMD})_{max}$$
$$RE_{aMD} / (RE_{aMD})_{max}$$

GN	Group	Period / QN	AN	Element	PN_{MD}	$SZ_{aMD}/(SZ_{aMD})_{max}$	$RE_{aMD}/(RE_{aMD})_{max}$
1	IA	2	3	Li	1	0.32352	0.36512
2	IIA	2	4	Be	75	0.14987	0.78817
13	IIIA	2	5	B	81	0.15317	0.77120
14	IVA	2	6	C	87	0.15208	0.77673
15	VA	2	7	N	93	0.14784	0.79901
16	VIA	2	8	O	99	0.14125	0.83626
17	VIIA	2	9	F	106	0.13040	0.90585
18	VIIIA	2	10	Ne	113	0.11812	1.0000
17	VIIA	1	1	H	105	0.04000	2.95309
18	VIIIA	1	2	He	112	0.05526	2.13768
1	IA	3	11	Na	2	0.57846	0.20420
2	IIA	3	12	Mg	76	0.23553	0.50153
13	IIIA	3	13	Al	82	0.22232	0.53132
14	IVA	3	14	Si	88	0.20841	0.56679
15	VA	3	15	P	94	0.19392	0.60912
16	VIA	3	16	S	100	0.17899	0.65995
17	VIIA	3	17	Cl	107	0.16060	0.73553
18	VIIIA	3	18	Ar	114	0.14201	0.83180
1	IA	4	19	K	3	0.69920	0.17065
2	IIA	4	20	Ca	7	0.67141	0.17593
3	IIIB	4	21	Sc	11	0.64709	0.18255
4	IVB	4	22	Ti	43	0.44100	0.26786
5	VB	4	23	V	47	0.42558	0.27756
6	VIB	4	24	Cr	51	0.41024	0.28794
7	VIIB	4	25	Mn	55	0.39497	0.29907
8	VIIIB	4	26	Fe	59	0.37977	0.31104
9	VIIIB	4	27	Co	63	0.36466	0.32393
10	VIIIB	4	28	Ni	67	0.34962	0.33787
11	IB	4	29	Cu	71	0.33466	0.35297
12	IIB	4	30	Zn	77	0.31091	0.37993
13	IIIA	4	31	Ga	83	0.28769	0.41060
14	IVA	4	32	Ge	89	0.26493	0.44586
15	VA	4	33	As	95	0.24261	0.48689
16	VIA	4	34	Se	101	0.22068	0.53528
17	VIIA	4	35	Br	108	0.19530	0.60484
18	VIIIA	4	36	Kr	115	0.17045	0.69301
1	IA	5	37	Rb	4	0.83202	0.14197
2	IIA	5	38	Sr	8	0.79756	0.14811
3	IIIB	5	39	Y	12	0.76224	0.15497
4	IVB	5	40	Zr	44	0.51595	0.22894
5	VB	5	41	Nb	48	0.49439	0.23893
6	VIB	5	42	Mo	52	0.47342	0.24951
7	VIIB	5	43	Tc	56	0.45299	0.26076
8	VIIIB	5	44	Ru	60	0.43305	0.27277
9	VIIIB	5	45	Rh	64	0.41355	0.28663
10	VIIIB	5	46	Pd	68	0.39446	0.29946
11	IB	5	47	Ag	72	0.37574	0.31438
12	IIB	5	48	Cd	78	0.34740	0.34003
13	IIIA	5	49	In	84	0.31994	0.36921
14	IVA	5	50	Sn	90	0.29328	0.40277
15	VA	5	51	Sb	96	0.26735	0.44184
16	VIA	5	52	Te	102	0.24207	0.48798
17	VIIA	5	53	I	109	0.21317	0.55412
18	VIIIA	5	54	Xe	116	0.18507	0.63826
1	IA	6	55	Cs	5	0.91016	0.12978
2	IIA	6	56	Ba	9	0.86879	0.13596
4	IVB	6	72	Hf	45	0.58884	0.20060
5	VB	6	73	Ta	49	0.56231	0.21007
6	VIB	6	74	W	53	0.53670	0.22009
7	VIIB	6	75	Re	57	0.51190	0.23076
8	VIIIB	6	76	Os	61	0.48784	0.24214
9	VIIIB	6	77	Ir	65	0.46446	0.25432
10	VIIIB	6	78	Pt	69	0.44171	0.26743
11	IB	6	79	Au	73	0.41953	0.28157
12	IIB	6	80	Hg	79	0.38669	0.30547
13	IIIA	6	81	Tl	85	0.35503	0.33272
14	IVA	6	82	Pb	91	0.32443	0.36410
15	VA	6	83	Bi	97	0.29478	0.40072
16	VIA	6	84	Po	103	0.26601	0.44406
17	VIIA	6	85	At	110	0.23335	0.50621
18	VIIIA	6	86	Rn	117	0.20171	0.58560
1	IA	7	87	Fr	6	1.00000	0.11812
2	IIA	7	88	Ra	10	0.95203	0.12408
4	IVB	7	104	Rf	46	0.63093	0.18722
5	VB	7	105	Db	50	0.60177	0.19630
6	VIB	7	106	Sg	54	0.57364	0.20592
7	VIIB	7	107	Bh	58	0.54644	0.21617
8	VIIIB	7	108	Hs	62	0.52011	0.22712
9	VIIIB	7	109	Mt	66	0.49455	0.23885
10	VIIIB	7	110	Ds	70	0.46971	0.25148
11	IB	7	111	Rg	74	0.44554	0.26513
12	IIB	7	112	Uub	80	0.41004	0.28808
13	IIIA	7	113	Uut	86	0.37584	0.31429
14	IVA	7	114	Uuq	92	0.34284	0.34455
15	VA	7	115	Uup	98	0.31091	0.37993
16	VIA	7	116	Uuh	104	0.27996	0.42192
17	VIIA	7	117	Uus	111	0.24493	0.48228
18	VIIIA	7	118	Uuo	118	0.21103	0.53975
		QN 6	57	La	13	0.82819	0.14263
		QN 6	58	Ce	15	0.81046	0.14575
		QN 6	59	Pr	17	0.79323	0.14891
		QN 6	60	Nd	19	0.77648	0.15213
		QN 6	61	Pm	21	0.76019	0.15539
		QN 6	62	Sm	23	0.74432	0.15870
		QN 6	63	Eu	25	0.72885	0.16207
		QN 6	64	Gd	27	0.71375	0.16550
		QN 6	65	Tb	29	0.69898	0.16899
		QN 6	66	Dy	31	0.68454	0.17256
		QN 6	67	Ho	33	0.67039	0.17620
		QN 6	68	Er	35	0.65651	0.17993
		QN 6	69	Tm	37	0.64290	0.18374
		QN 6	70	Yb	39	0.62952	0.18764
		QN 6	71	Lu	41	0.61638	0.19164
		QN 7	89	Ac	14	0.90600	0.13038
		QN 7	90	Th	16	0.88516	0.13345
		QN 7	91	Pa	18	0.86498	0.13656
		QN 7	92	U	20	0.84542	0.13972
		QN 7	93	Np	22	0.82645	0.14293
		QN 7	94	Pu	24	0.80802	0.14619
		QN 7	95	Am	26	0.79009	0.14951
		QN 7	96	Cm	28	0.77264	0.15288
		QN 7	97	Bk	30	0.75563	0.15633
		QN 7	98	Cf	32	0.73902	0.15984
		QN 7	99	Es	34	0.72280	0.16343
		QN 7	100	Fm	36	0.70693	0.16709
		QN 7	101	Md	38	0.69140	0.17085
		QN 7	102	No	40	0.67618	0.17469
		QN 7	103	Lr	42	0.66125	0.17864

Table 2c Atomic number AN, periodic number PN_{ME} and diagonal number DN_{ME} in Meyer's Periodic System.

Cell legend: each cell shows element symbol with AN (atomic number) and PN_{ME} in the upper part, and DN_{ME} in the lower part — format below: **Sym (AN, PN_{ME}, DN_{ME})**.

GN →	1 IA	2 IIA	3 IIIB	4 IVB	5 VB	6 VIB	7 VIIB	8 VIIIB	9 VIIIB	10 VIIIB	11 IB	12 IIB	13 IIIA	14 IVA	15 VA	16 VIA	17 VIIA	18 VIIIA
QN 1	H (1, 1, 101)																	He (2, 112, 1)
QN 2	Li (3, 2, 103)	Be (4, 8, 102)											B (5, 82, 17)	C (6, 88, 12)	N (7, 94, 8)	O (8, 100, 5)	F (9, 106, 3)	Ne (10, 113, 2)
QN 3	Na (11, 3, 106)	Mg (12, 9, 104)											Al (13, 83, 23)	Si (14, 89, 18)	P (15, 95, 13)	S (16, 101, 9)	Cl (17, 107, 6)	Ar (18, 114, 4)
QN 4	K (19, 4, 109)	Ca (20, 10, 107)	Sc (21, 14, 105)	Ti (22, 46, 64)	V (23, 50, 60)	Cr (24, 54, 56)	Mn (25, 58, 52)	Fe (26, 62, 48)	Co (27, 66, 44)	Ni (28, 70, 40)	Cu (29, 74, 36)	Zn (30, 78, 32)	Ga (31, 84, 28)	Ge (32, 90, 24)	As (33, 96, 19)	Se (34, 102, 14)	Br (35, 108, 10)	Kr (36, 115, 7)
QN 5	Rb (37, 5, 113)	Sr (38, 11, 110)	Y (39, 15, 108)	Zr (40, 47, 68)	Nb (41, 51, 65)	Mo (42, 55, 61)	Tc (43, 59, 57)	Ru (44, 63, 53)	Rh (45, 67, 49)	Pd (46, 71, 45)	Ag (47, 75, 41)	Cd (48, 79, 37)	In (49, 85, 33)	Sn (50, 91, 29)	Sb (51, 97, 25)	Te (52, 103, 20)	I (53, 109, 15)	Xe (54, 116, 11)
QN 6	Cs (55, 6, 116)	Ba (56, 12, 114)	La (57, 16, 111)	Hf (72, 48, 71)	Ta (73, 52, 69)	W (74, 56, 66)	Re (75, 60, 62)	Os (76, 64, 58)	Ir (77, 68, 54)	Pt (78, 72, 50)	Au (79, 76, 46)	Hg (80, 80, 42)	Tl (81, 86, 38)	Pb (82, 92, 34)	Bi (83, 98, 30)	Po (84, 104, 26)	At (85, 110, 21)	Rn (86, 117, 16)
QN 7	Fr (87, 7, 118)	Ra (88, 13, 117)	Ac (89, 17, 115)	Rf (104, 49, 74)	Db (105, 53, 72)	Sg (106, 57, 70)	Bh (107, 61, 67)	Hs (108, 65, 63)	Mt (109, 69, 59)	Ds (110, 73, 55)	Rg (111, 77, 51)	Uub (112, 81, 47)	Uut (113, 87, 43)	Uuq (114, 93, 39)	Uup (115, 99, 35)	Uuh (116, 105, 31)	Uus (117, 111, 27)	Uuo (118, 118, 22)

f-block:

QN 6	Ce (58, 18, 99)	Pr (59, 20, 97)	Nd (60, 22, 95)	Pm (61, 24, 93)	Sm (62, 26, 91)	Eu (63, 28, 89)	Gd (64, 30, 87)	Tb (65, 32, 85)	Dy (66, 34, 83)	Ho (67, 36, 81)	Er (68, 38, 79)	Tm (69, 40, 77)	Yb (70, 42, 75)	Lu (71, 44, 73)
QN 7	Th (90, 19, 112)	Pa (91, 21, 100)	U (92, 23, 98)	Np (93, 25, 96)	Pu (94, 27, 94)	Am (95, 29, 92)	Cm (96, 31, 90)	Bk (97, 33, 88)	Cf (98, 35, 86)	Es (99, 37, 84)	Fm (100, 39, 82)	Md (101, 41, 80)	No (102, 43, 78)	Lr (103, 45, 76)

Fig. 6 Atomic size SZ_{aME} and atomic reactivity RE_{aME} (normalized to $(SZ_{aME})_{max}$ and RE_{aME} (Ne)) versus periodic number PN_{ME}; ■ $SZ_{aME} / (SZ_{aME})_{max}$, ✕ RE_{aME} / RE_{aME} (Ne).
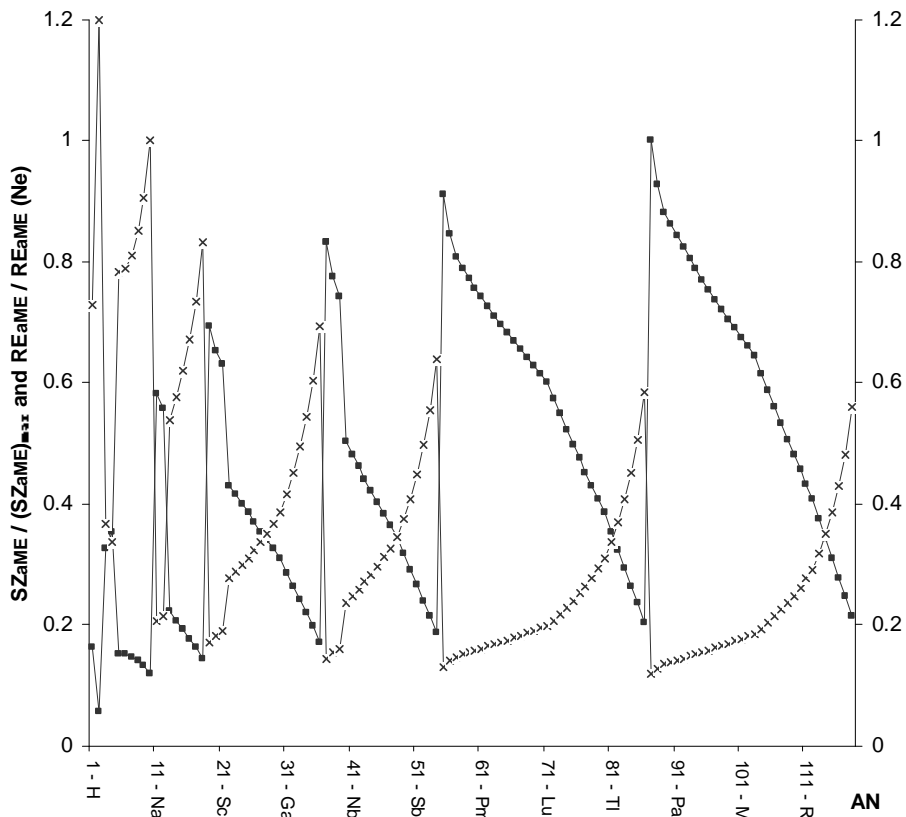
Fig. 7 Atomic size SZ_{aME} and atomic reactivity RE_{aME} (normalized to $(SZ_{aME})_{max}$ and RE_{aME} (Ne)) versus atomic number AN; ■ $SZ_{aME} / (SZ_{aME})_{max}$, ✕ RE_{aME} / RE_{aME} (Ne).

In Fig. 8 we compare the atomic size SZ_{aME} and SZ_{aMD} (normalized to $(SZ_{aME})_{max} = (SZ_{aMD})_{max}$) versus atomic number AN. For the calculation of SZ_{aME} we used the periodic number PN_{ME} of Meyer's Periodic Table, and for the calculation of SZ_{aMD} we used the corresponding periodic number PN_{MD} of Mendeleyev's Periodic Table. Fig. 9 gives the analog for the atomic reactivity RE_{aME} and RE_{aMD}. It visualizes clearly that, with the exception of those of H, Be and Mg, the values are just slightly different (see also Tables 2a and 2b).

Figs. 10 and 11 compare $R(Z)$ / $R(Z)_{max}$, $Ri(Y)$ / $Ri(Y)_{max}$ and SZ_{aME} / $(SZ_{aME})_{max}$, respectively $EN(AR)$ / $EN(AR)_{max}$, $EN(MB)$ / $EN(MB)_{max}$ and RE_{aME} / RE_{aME} (Ne) as function of PN_{ME}. $R(Z)$, $Ri(Y)$, $EN(AR)$, and $EN(MB)$ showed so far to be the most effective ones among the derived elemental-property parameters for separating compound formers from non-formers in multinary systems [17], as well as in structure maps [18]. SZ_{aME} (SZ_{aMD}) and RE_{aME} (RE_{aMD}) fit very well in the general trend of the pattern behaviors shown in Figs. 1 and 2.

As an example and in order to demonstrate the 'linear correlation' between SZ_{aME} and $R(Z)$ in Fig. 12 we plot SZ_{aME} / $(SZ_{aME})_{max}$ versus $R(Z)$ / $R(Z)_{max}$. The slope of its linear regression line represents the scaling factor k_{SZ} in our equation (1) for the case SZ_{aME} and $R(Z)$.

Equations (1) and (2) correlate quantitatively both the size of an atom and its ability to attract different atoms with two different ways of counting the chemical elements in the Periodic Table.

In Table 2c we introduce an additional enumeration of the elements in Meier's Periodic Table, called 'diagonal number' DN_{ME}, whereby the enumeration of the elements follows diagonal lines. Its starts with He and ends with Fr following the general trend from top → down in first priority and left → right in second priority.

In Fig. 13 DN_{ME} / $(DN_{ME})_{max}$ and our calculated SZ_{aME} / $(SZ_{aME})_{max}$ versus PN_{ME} are shown. The only significant differences are observed for the first three group numbers GN 1-3, but the two functions are remarkably similar. This observation shows that the atomic size can be directly obtained with this alternative kind of enumeration within Meier's Periodic Table. It might open a more direct approximation of the atomic size of the elements and possibly other derived elemental-property parameters using AN and PN.
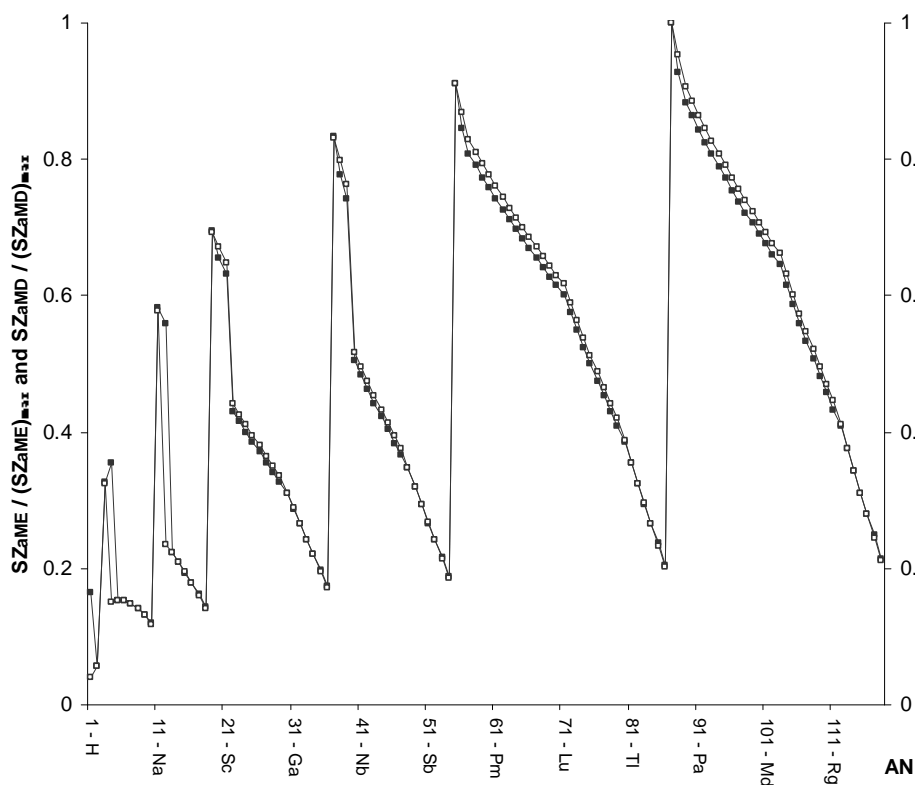
Fig. 8 Atomic size SZ_{aME} and SZ_{aMD} (normalized to $(SZ_{aME})_{max}$ and $(SZ_{aMD})_{max}$) versus atomic number AN; ■ SZ_{aME} / $(SZ_{aME})_{max}$, □ SZ_{aMD} / $(SZ_{aMD})_{max}$. For the calculation of SZ_{aME} we used the periodic number PN_{ME} of Meyer's Periodic System (Table 2a), while for the calculation of SZ_{aMD} we used the periodic number PN_{MD} of Mendeleyev's Periodic System (Table 2b).
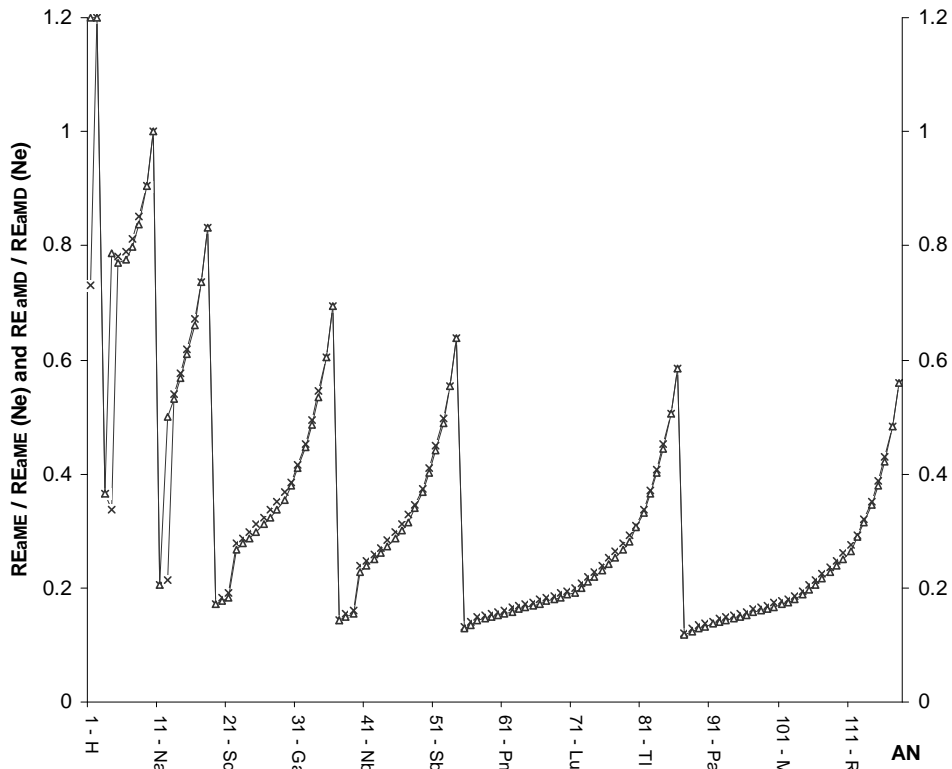
Fig. 9 Atomic reactivity RE_{aME} and RE_{aMD} (normalized to RE_{aME} (Ne) and RE_{aMD} (Ne)) versus atomic number AN; $\times$ RE_{aME} / RE_{aME} (Ne), $\triangle$ RE_{aMD} / RE_{aMD} (Ne). The calculation of RE_{aME} is based on Meyer's Periodic System (Table 2a), while the calculation of RE_{aMD} is based on Mendeleyev's Periodic System (Table 2b).
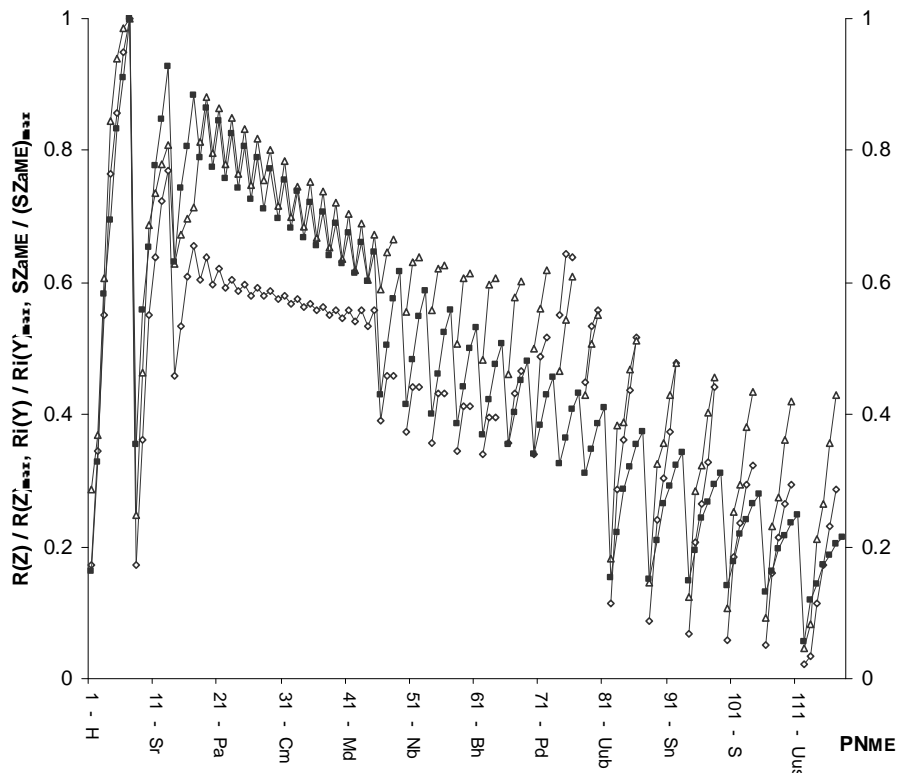
Fig. 10 Pseudo-potential radii according to Zunger $R(Z)$, ionic radii according to Yagoda $Ri(Y)$ and calculated atomic size SZ_{aME} (normalized to $R(Z)_{max}$, $Ri(Y)_{max}$, $(SZ_{aME})_{max}$, respectively) versus periodic number PN_{ME}; $\triangle$ $R(Z)$ / $R(Z)_{max}$, $\diamond$ $Ri(Y)$ / $Ri(Y)_{max}$, $\blacksquare$ SZ_{aME} / $(SZ_{aME})_{max}$. In the case of $R(Z)$ (the pseudo-potential radii according to Zunger, where no published data exist for the f-elements) we estimated the $R(Z)$ values based on a linear dependence for QN 6 between $R(Z) = 3.55$ for Ce and $R(Z) = 2.64$ for Lu, as well as for QN 7 between $R(Z) = 3.85$ for Th and $R(Z) = 2.94$ for Lr.
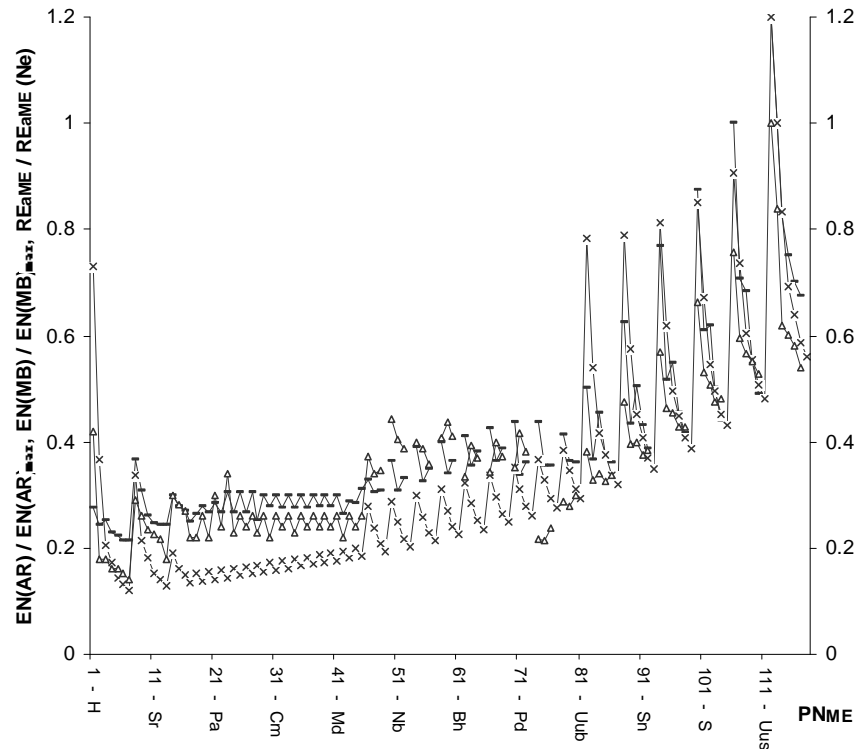
Fig. 11 Electronegativity according to Allred and Rochow EN(AR), electronegativity according to Martynov and Batsanov EN(MB), and calculated atomic reactivity RE_{aME} (normalized to $EN(AR)_{max}$, $EN(MB)_{max}$ and RE_{aME} (Ne), respectively) versus periodic number PN_{ME}; $-$ EN(AR) / $EN(AR)_{max}$, $\triangle$ EN(MB) / $EN(MB)_{max}$, $\times$ RE_{aME} / RE_{aME} (Ne).
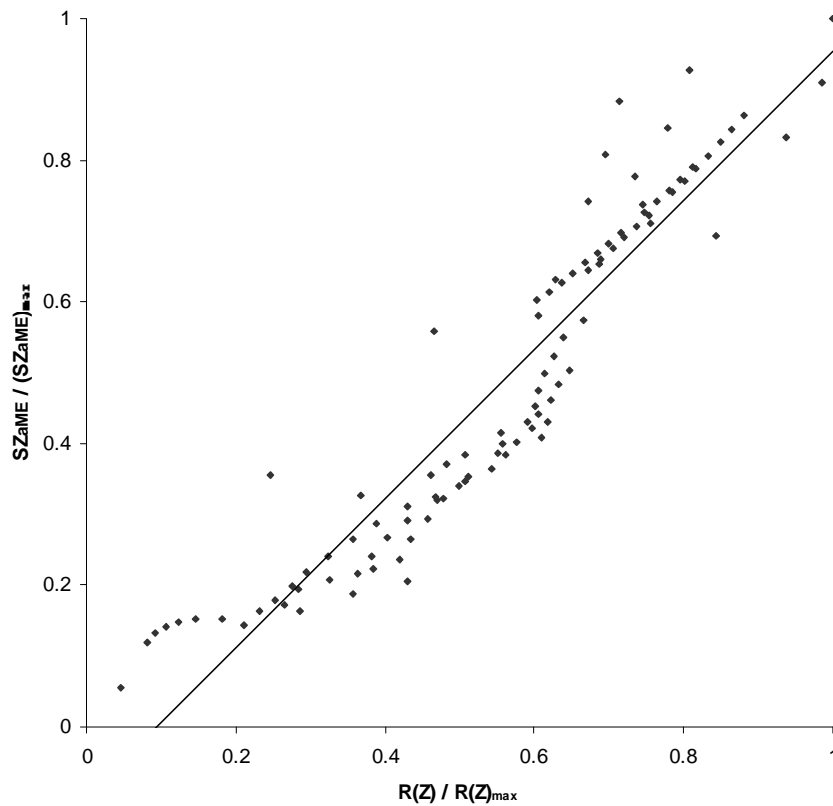
Fig. 12 Atomic size SZ_{aME} (normalized to $(SZ_{aME})_{max}$) versus pseudo-potential radii according to Zunger R(Z) (normalized to $R(Z)_{max}$) with linear regression line. In the case of R(Z), the pseudo-potential radii according to Zunger (where no published data exist for the f-elements) we estimated the R(Z) values based on a linear dependence for QN 6 between R(Z) = 3.55 for Ce and R(Z) = 2.64 for Lu, as well as for QN 7 between R(Z) = 3.85 for Th and R(Z) = 2.94 for Lr.
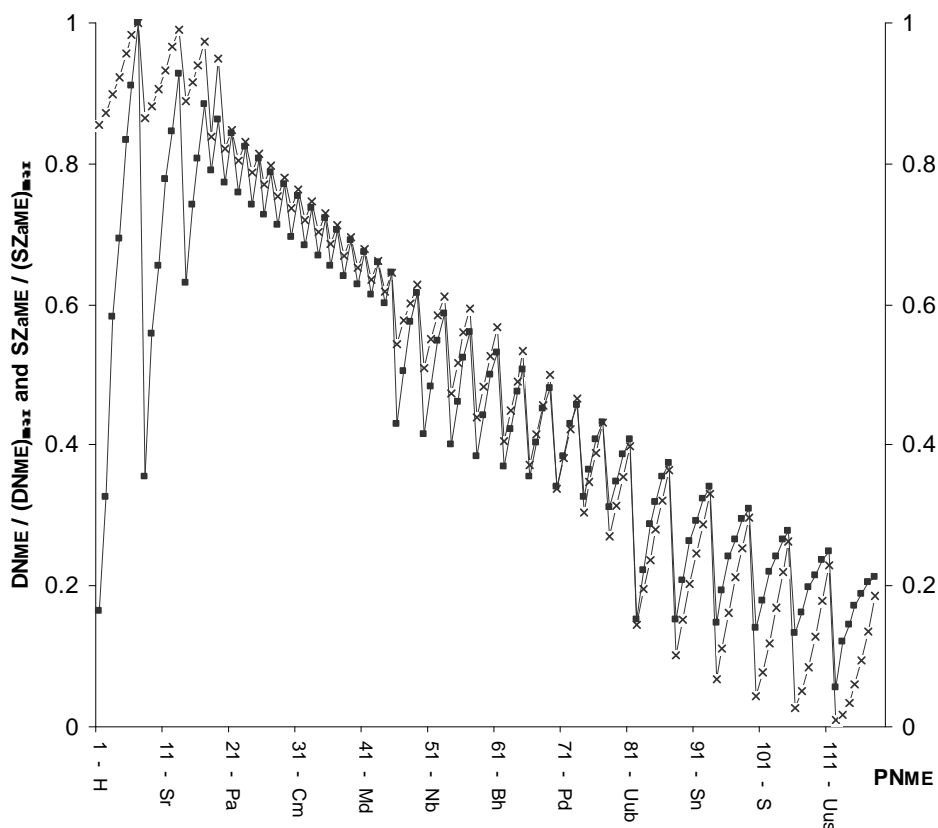
Fig. 13 Atomic size SZ_{aME} (normalized to $(SZ_{aME})_{max}$) and $DN_{ME} / (DN_{ME})_{max}$. DN_{ME} is defined by a diagonal enumeration of Meyer's Periodic System (Table 2c); ■ $SZ_{aME} / (SZ_{aME})_{max}$, × $DN_{ME} / (DN_{ME})_{max}$.

From the different elemental-property parameter patterns we could so far formulate the atomic size and the atomic reactivity as functions of AN and PN. We do get the impression that all additional patterns like e.g. the atomic affinity pattern and the atomic density pattern can be formulated as function of AN and PN. As an example the density (= atomic mass / atomic volume) may easily be derived from the patterns discussed above, in this case from the atomic number pattern and the atomic size pattern. As can be seen in Fig. 3 the atomic affinity pattern group members show for the GN 1-3 and GN 13-18 elements the general trend from the atomic reactivity pattern, while for the GN 4-12 the trend follows the atomic size pattern group.

In our analysis of the atomic environment types AET [19,20] we observed that in a crystal structure the atomic size, the atomic reactivity and the atomic affinity determine the AET. The analysis of over 200,000 atomic positions in binary, ternary and quaternary compounds revealed that less than 60 geometrically different AET's (the majority belonging to only 24 AET's) are observed in experimental data. The AET's are governed by the size of the constituent chemical elements A, B, C, …, the interactions A-B, A-C, B-C, …, as well as A-A, B-B, C-C, … (which are represented by the corresponding shortest interatomic distances).

Elemental-property parameters used for parameterization of materials properties

Previous papers, such as that on the prediction of compound formers/non-formers in binary, ternary and quaternary systems [17], as well as that on the atomic environment (AET) prediction for equiatomic binary compounds [18], do well support the postulate: "Structure-sensitive properties of materials are quantitatively described by the elemental-property parameters of the constituent chemical elements." Now as we obtained a direct link between the fundamental elemental parameters AN and PN and the derived elemental-property parameters SZ_a and RE_a, we can generalize our postulate to the following statement: "Structure-sensitive material properties are quantitatively determined by the elemental-property parameters AN and PN (or simple mathematical functions of them) of the constituent chemical elements." This generalization is an important link to strategically explore structure-sensitive properties of materials.

In [17] we gained from different compilations [21-27] the following numbers of distinct information about compound-forming, respectively non-forming systems: 2016 binary systems, 6382 ternary systems and 7021 quaternary systems. For that work, 44 binary non-forming systems were discarded because either

we found contradictory information or the information was based on one schematic phase diagram (either from old experimental work or from calculated phase diagrams (CALPHAD work): Ag–U, Al–Cs, Al–K, Al–Na, Au–B, Au–Ge, Au–P, B–Bi, B–Cd, B–Cu, B–Ga, B–Ge, B–Hg, B–Pb, B–Sb, B–Sn, B–Tl, B–Zn, Ba–Ni, Be–Li, Be–Na, Bi–Si, C–Co, C–Ge, C–Ir, C–Ni, Cs–Li, Cs–Mg, Cu–Li, K–Li, K–Mg, Li–Mg, Li–Ni, Li–Pu, Li–Rb, Mg–Na, Mg–Rb, Mg–U, Re–Sb, S–Te, Sb–W, Si–Sn, Si–Tl, Si–Zn. The remaining three data sets: 2318 binary systems (with the halides added), 6266 ternary systems and 6913 quaternary systems are used throughout the present work.

Below we give the two conditions (criteria), which we used to decide whether a binary, ternary or quaternary system belongs to a former or non-former system. These two definitions (here explicitly explained for the ternary case) for separating formers from non-formers are based on the following fundamentals:

- Description of crystal structure within the framework of space group theory,
- Gibbs' phase rule.

As the majority of the data are gained under normal conditions (T = 298 K, and atmospheric pressure) and phase equilibrium, we take this as our standard reference.

Definition 1: A compound-forming system possesses at least one ternary compound separated by three two-phase regions involving three adjacent chemical element(s) and/or binary compound(s) and/or ternary compound(s) in at least one complete isothermal section. In those cases where no phase diagram for an A–B–C system is known, but a ternary compound with a ternary crystal structure is reported, then this system is of course accepted as a compound former.

Definition 2: A ternary system is a non-former when no three-phase region or complete solid solution respectively, is shown in at least one complete isothermal section. In those cases where no complete isothermal section for an A–B–C system is known, a ternary system will be a non-former when all its three binary boundary systems are non-formers. Based on this criterion we derived 2118 ternary and 4274 quaternary non-formers from 634 published binary non-former phase diagrams. This criterion has been confirmed, so far, by all published ternary isothermal sections.

Based on the criterion that a ternary system is a former when at least one ternary 'line' phase with a ternary crystal structure is published, we found 4264 ternary formers. Note that this simple criterion excludes pseudo-ternary phases, which are solid solutions of binary compounds.

In order to test the parameterization power of SZ_a and RE_a we applied the program "DISCOVERY" [28] on the above-mentioned data sets. The aim of this process is to search systematically for salient 2D- and 3D-features sets, to visualize them and to correlate quantitatively the materials properties of these chemical elements (in this case: formers/non-formers) with the elemental-property parameters of their constituent chemical elements. This is done in a three-step procedure:

1) Collection of published (experimental and theoretical) elemental-property parameters and calculation of SZ_a and RE_a (Tables 1 and 2).

2) Application of an automatic generator for 2D- and 3D-features sets resulting from combinations of elemental-property parameters and mathematical operators. We introduced the operators +, –, *, /, max. and min. to link the elemental-property parameters (EP) of the different constituents, say A, B, C for ternary systems, to form global elemental-property parameters EP1(tot) = EP1(A) op EP1(B) op EP1(C).

3) Automatic high-quality separation detection and its visualization.

The general idea is very simple: Assume that we consider 2000 data points (1000 ternary formers and 1000 ternary non-formers) in a plot of a selected 3D-features set. We check now for each point whether their n nearest neighbors in the plot are of the same class or not, here formers or non-formers, and make a statistical analysis for all 2000 data points (n = 1, …, 50, depending on the standard wanted).

As a first example, we ran the program "DISCOVERY" on all elemental-property parameters belonging to each pattern group listed in Tables 1a, 1b. Using all mathematical operators mentioned above, we give the best separation results listed in Table 3a. The success of the separation is indicated by the hit rate, e.g. 90% (n = 1) → 44% (n = 50). This means that 90% of all data points have a nearest neighbor of the same kind and 44% of all data points have 50 nearest neighbors of the same kind taking the reference data point as the center of a circular area. Table 3b shows the results of the best 2D-features sets using AN, PN, and the calculated elemental-property parameters SZ_a and RE_a. The 6266 ternary systems mentioned above were used as test material.

A detailed comparison for the separation efficiency of the various elemental-property parameters of the atomic size and atomic reactivity patterns groups is given in Tables 4 and 5, respectively. It is evident that SZ_{aME} (AN, PN_{ME}) as well as RE_{aME} (AN, PN_{ME}) (in all cases the normalized values are used) improve the separation between compound-forming and non-forming systems. Similar improvements have been achieved for binary and quaternary data sets.

Figs. 14-16 show the best "overall results" 2D-features sets for binary, ternary and quaternary chemical systems. We are aware that by using a 2D-features set, instead of a 3D-features set, we are loosing some information. Nevertheless by using the appropriate 2D-projection of the 3D-features set we loose only about 3% accuracy, but get the advantage that the given figures are easy to interpret. Based on our statistical analysis several 2D- and 3D-features

Table 3 (a) Result of the best 3D-features sets using the published elemental-property data sets (Tables 1a, 1b). The hit rate per ternary system decreases from e.g. 97% with n = 1 correct neighbor to 71% with n = 50 correct nearest neighbors for the case of the reactivity pattern group. (b) Result of the best 2D-features sets using the calculated elemental-property data sets (Tables 2a, 2b). The hit rate per ternary system decreases from e.g. 99% with n = 1 correct neighbor to 86% with n = 50 correct nearest neighbors for the case of RE_{aME}.

<div align="center">(<i>a</i>)</div>

Using all data sets listed in Tables 1a, 1b for each group	Elemental-property parameter as listed in Tables 1a, 1b	Separation result in the best 3D-features set
I) Atomic number pattern group	AN, QN, AM, nc(C), nc(S), cma(Mo), cma(Cu), aes	90% (n = 1) → 44% (n = 50)
II) Periodic number pattern group	PN_{ME}, PN_{MD}, PN(P), VE, GN	99% (n = 1) → 88% (n = 50)
i) Atomic size pattern group	R(Z), Ri(Y), Rc(P), Rm(WG), Rve(S), Rce(S), R(M), R(VD)	98% (n = 1) → 75% (n = 50)
ii) Atomic reactivity pattern group	EN(MB), EN(P), EN(AR), EN(abs), IE(first), CP(M), wf, n(WS)	97% (n = 1) → 71% (n = 50)

<div align="center">(<i>b</i>)</div>

Belonging to patterns listed in Tables 1a, 1b	Derived elemental-property parameters as listed in Tables 2a, 2b	Separation result in the best 2D-features set
I) Atomic number pattern group	AN	88% (n = 1) → 40% (n = 50)
II) Periodic number pattern group	PN_{ME}, PN_{MD}	99% (n = 1) → 84% (n = 50)
i) Atomic size pattern group	SZ_{aME}, SZ_{aMD}	99% (n = 1) → 84% (n = 50)
ii) Atomic reactivity pattern group	RE_{aME}, RE_{aMD}	99% (n = 1) → 86% (n = 50)

Table 4 Results of the best 2-D features sets using only elemental parameters belonging to the atomic size pattern. The hit rate per ternary system decreases from e.g. 98.9% with n = 1 correct neighbor to 83.2% with 50 correct nearest neighbors for the case of SZ_{aME}.

i) Atomic size pattern group	Best separation result
R(VD): Radii according to Villars and Daams (derived from their V compilation)	81.3% (n = 1) → 5.5% (n = 50)
R(M): Radii according to Miedema (derived from his $V^{2/3}$ compilation)	81.3% (n = 1) → 5.7% (n = 50)
Rve(S): Valence electron distance according to Schubert	84.3% (n = 1) → 15.6% (n = 50)
Rce(S): Core electron distance according to Schubert	85.6% (n = 1) → 22.5% (n = 50)
Rm(WG): Metal radii according to Waber and Gschneidner	81.9% (n = 1) → 33.2% (n = 50)
Rc(P): Covalent radii according to Pauling	87.3% (n = 1) → 38.3% (n = 50)
Ri(Y): Ionic radii according to Yagoda	88.7% (n = 1) → 48.1% (n = 50)
R(Z): Pseudo-potential radii according to Zunger	97.9% (n = 1) → 75.2% (n = 50)
SZ_{aME} (AN, PN_{ME}): Atomic size, present calculation	98.9% (n = 1) → 83.2% (n = 50)

sets have better separation results (in the range of 3-5%). The plots shown here reflect clearly the validity of Pauling's electronegativity- and Laves' geometrical-concepts shown by its atomic reactivity differences (y-axis) and its atomic size ratios (x-axis), respectively.

These plots demonstrate that for compound formation the atomic reactivity difference is more important than the atomic size ratio. Furthermore in plots using SZ_a and RE_a Meyer's Periodic System is more efficient than Mendeleyev's System. However, in plots where we use PN, it is Mendeleyev's System that is more efficient (Figs. 17, 18).

By using "Pauling's and Laves' axes" we reach for the binary, ternary and quaternary systems the following hit rates: 78.5% (n = 1) → 39.2% (n = 50), 97.8% (n = 1) → 81.0% (n = 50), 99.9% (n = 1) → 97.9% (n = 50), respectively.

Table 5 Results of the best 2-D features sets using only elemental parameters belonging to the atomic reactivity pattern. The hit rate per ternary system decreases from e.g. 98.7% with 1 correct neighbor to 84.9% with 50 correct nearest neighbors for the case of RE_{aME}.

ii) Atomic reactivity pattern group	Best separation result
n(WS): n(Wigner and Seitz) according to Miedema	90.8% (n = 1) → 34.3% (n = 50)
EN(MB): Electronegativity according to Martynov & Batsanov	93.7% (n = 1) → 40.9% (n = 50)
wf: Work function	83.0% (n = 1) → 49.4% (n = 50)
EN(abs): Absolute electronegativity	94.4% (n = 1) → 52.9% (n = 50)
EN(P): Electronegativity according to Pauling	76.6% (n = 1) → 55.9% (n = 50)
IE(first): First ionization energy	94.7% (n = 1) → 54.7% (n = 50)
CP(M): Chemical potential according to Miedema	96.1% (n = 1) → 59.3% (n = 50)
EN(AR): Electronegativity according to Allred & Rochow	96.6% (n = 1) → 70.6% (n = 50)
RE_{aME} (AN, PN_{ME}): Atomic reactivity, present calculation	98.7% (n = 1) → 84.9% (n = 50)
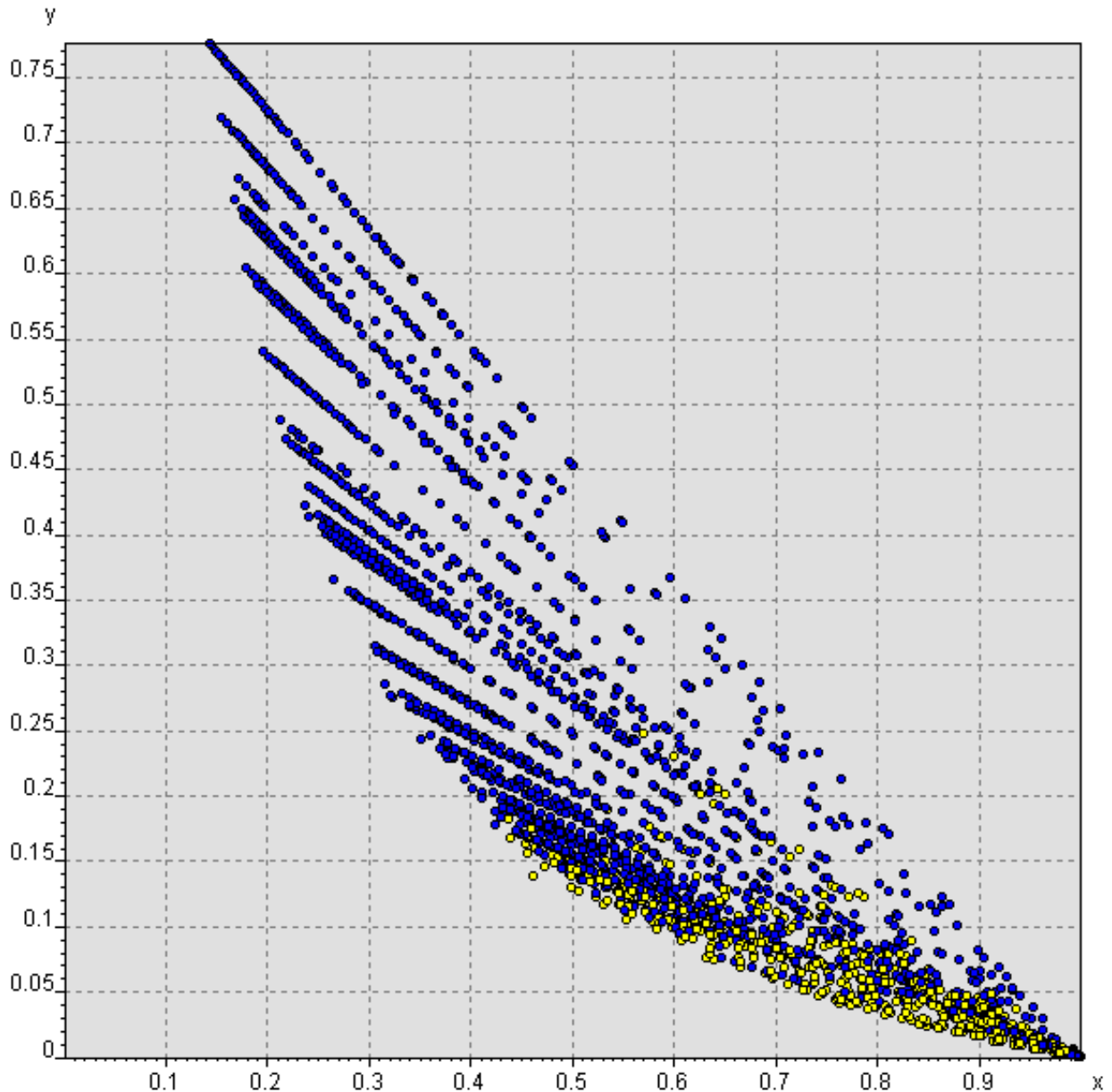
Fig. 14 Separation of 2318 binary systems into compound formers (●) and non-formers (○) based on a plot of the absolute difference (y-axis) $|RE_{aME}$ (A) $- RE_{aME}$ (B)$|$ / (2 RE_{aME} (Ne)) versus the ratio (x-axis) SZ_{aME} (A) / (2 SZ_{aME} (B)). For the binary systems the absolute difference = (|EP (A) – EP (B)|) / (n(n-1)/2) and for the ratio [EP (A) / EP (B)] / (n(n-1)/2), EP (A) < EP (B) and EP (A), EP (B) ≤ 1. EP (A), EP (B) = elemental-property parameters of element A and B, respectively, and n = number of elements = 2.
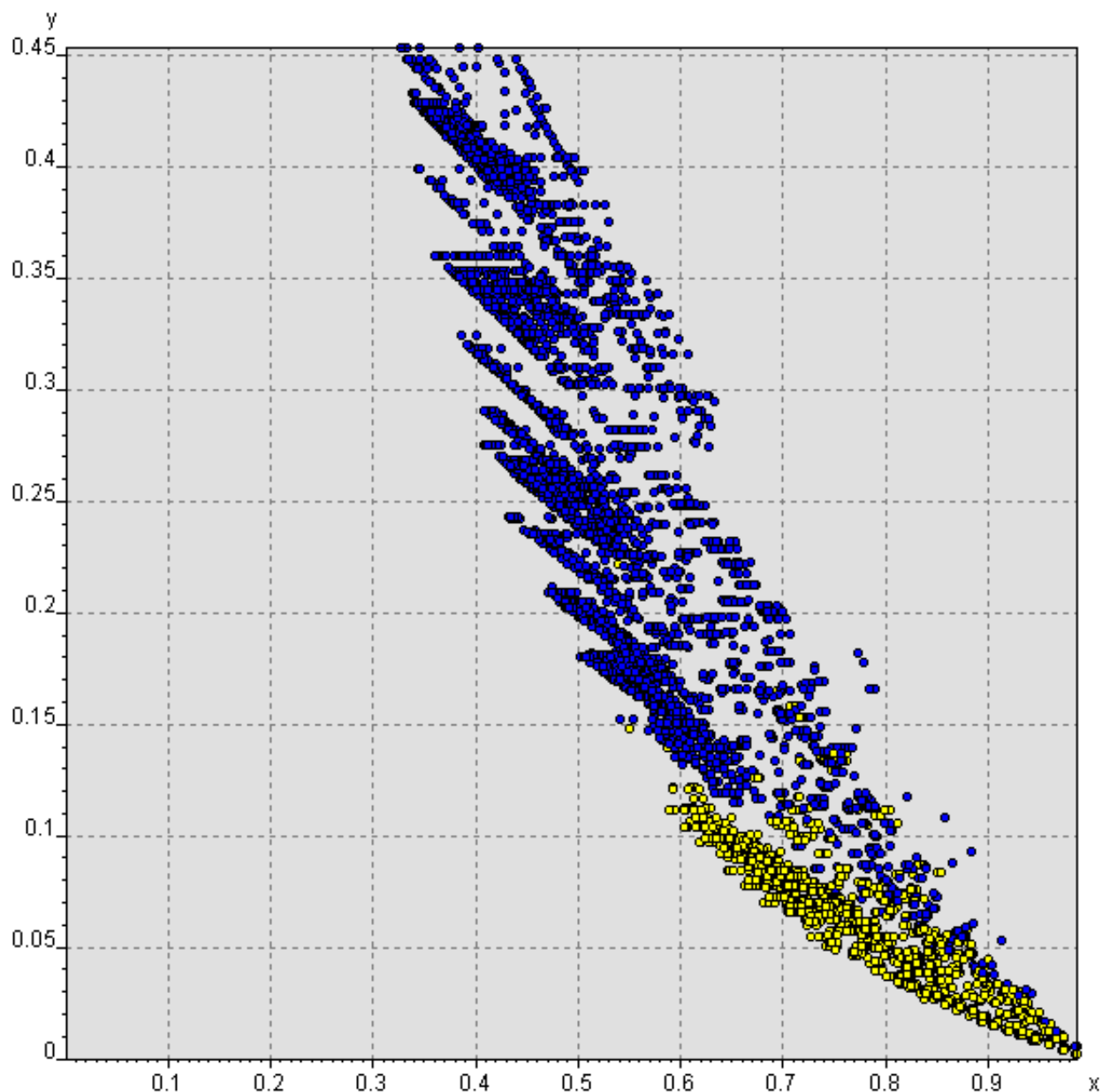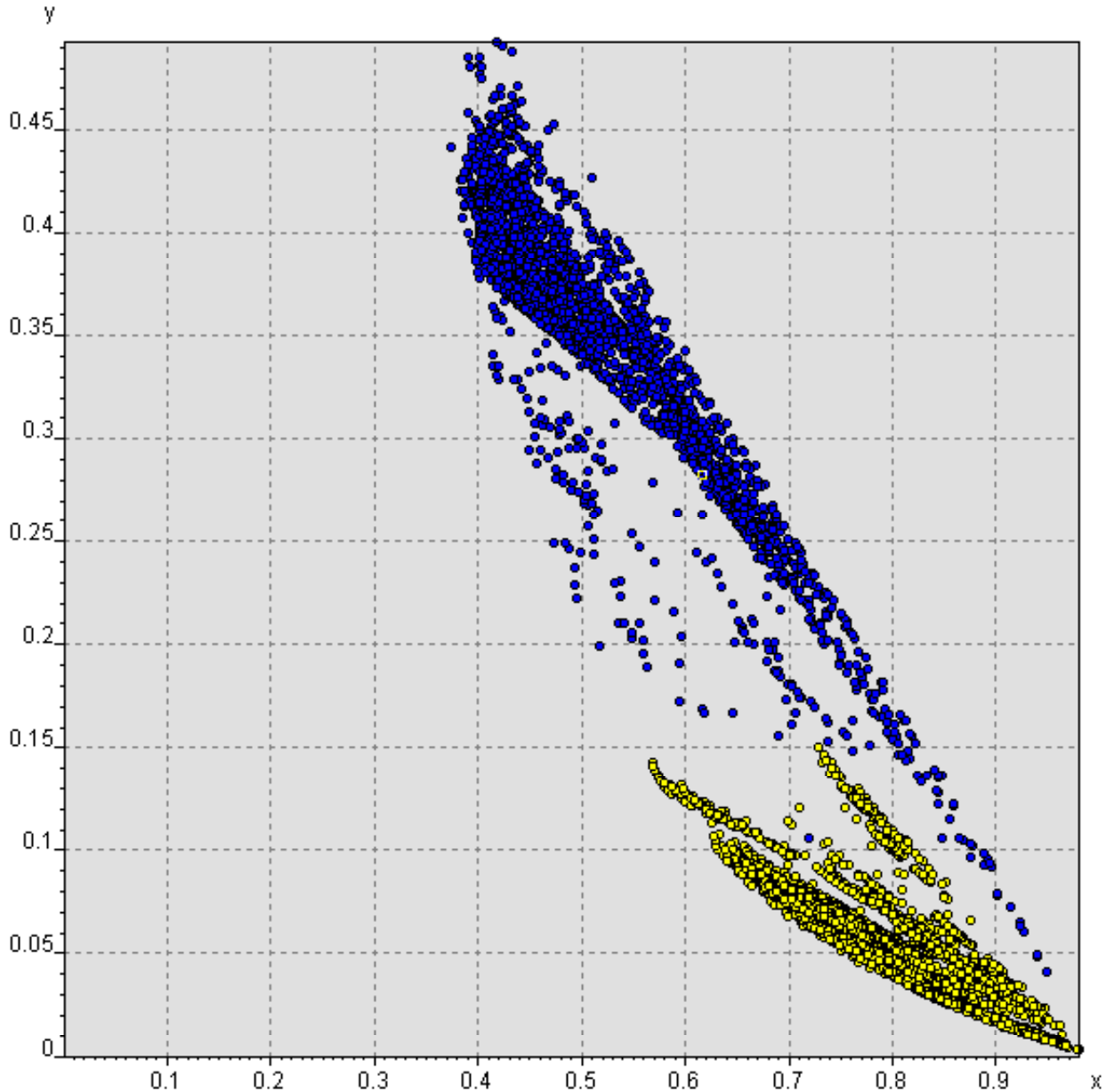
Fig. 15 Separation of 6266 ternary systems into compound formers (●) and non-formers (○) based on a plot of the absolute difference (y-axis) $2/3[(RE_{aME}(A) - RE_{aME}(C)) / RE_{aME}(Ne)]$ versus the ratio (x-axis) $[SZ_{aME}(A) / SZ_{aME}(B) + SZ_{aME}(A) / SZ_{aME}(C) + SZ_{aME}(B) / SZ_{aME}(C)]/3$. For the ternary systems the absolute difference $= (|EP(A) - EP(B)| + |EP(A) - EP(C)| + |EP(B) - EP(C)|) / (n(n-1)/2)$ and for the ratio $[EP(A) / EP(B) + EP(A) / EP(C) + EP(B) / EP(C)] / (n(n-1)/2)$, with $EP(A) < EP(B) < EP(C)$ and $EP(A)$, $EP(B)$, $EP(C) \leq 1$. $EP(A)$, $EP(B)$, $EP(C)$ = elemental-property parameters of element A, B and C, respectively and n = number of elements = 3.

We like to stress the fact that although the atomic reactivity is the reciprocal of the atomic size, it is justified to use in some cases both in the same plot. In a ternary system A–B–C in general the element with the maximal atomic size has the minimal atomic reactivity. This means e.g. in a max.$(SZ_a(A,B,C))$ versus max.$(RE_a(A,B,C))$ plot that only the B-element is not taken into consideration, otherwise the B- and C-elements would be ignored.

For binary systems, comparing the results of Fig. 14 with those of Fig. 17, a much more satisfactory separation is achieved using a max.$[PN_{ME}(A) / (PN_{ME})_{max}, PN_{ME}(B) / (PN_{ME})_{max}]$ versus $[PN_{ME}(A) / (PN_{ME})_{max} \times PN_{ME}(B) / (PN_{ME})_{max}]$ plot, with a separation of 95.6% (n = 1) → 44.5% (n = 50). This plot is further improved (see Fig. 18) when the Meyer numbers PN_{ME} are replaced by the Mendeleyev numbers PN_{MD} to 95.6% (n = 1) → 55.3% (n = 50).

Fig. 16 Separation of 6913 quaternary systems into compound formers (●) and non-formers (○) based on a plot of the absolute difference (y-axis) [3(RE$_{aME}$ (A) − RE$_{aME}$ (D)) + (RE$_{aME}$ (B) − RE$_{aME}$ (C))] / 6(RE$_{aME}$ (Ne)) versus the ratio (x-axis) [SZ$_{aME}$ (A) / SZ$_{aME}$ (B) + SZ$_{aME}$ (A) / SZ$_{aME}$ (C) + SZ$_{aME}$ (A) / SZ$_{aME}$ (D) + SZ$_{aME}$ (B) / SZ$_{aME}$ (C) + SZ$_{aME}$ (B) / SZ$_{aME}$ (D) + SZ$_{aME}$ (C) / SZ$_{aME}$ (D)] / 6. For the quaternary systems the absolute difference = (|EP (A) − EP (B)| + |EP (A) − EP (C)| + |EP (A) − EP (D)| + |EP (B) − EP (C)| + |EP (B) − EP (D)| + |EP (C) − EP (D)|) / (n(n-1)/2), with EP (A) < EP (B) < EP (C) < EP (D) and EP (A), EP (B), EP (C), EP (D) ≤ 1. EP (A), EP (B), EP (C), EP (D) = elemental-property parameters of element A, B, C, and D, respectively, and n = number of elements = 4.

The hit rate as a function of n shows for the binary systems that the hit rates decreases by the same size in the intervals n = 1-15 and n = 16-50, therefore it is appropriate to give 95.6% (n = 1) → 75% (n = 15). We even do not take into account that the binary systems data set is about one third of the ternary or quaternary data set. The clearest violation is the system Li−Sr, which contains five phases and is clearly located in the non-formers area. On the opposite side, the system Se−Te is clearly located in the former area, but several established, independently determined, phase diagrams show no binary phase. Here we dare to predict the existence of a binary phase, in analogy to the S–Se system. Two additional violations in the non-forming area: Hf–V and V–Zr have one phase crystallizing in the prototype 'MgCu$_2$,
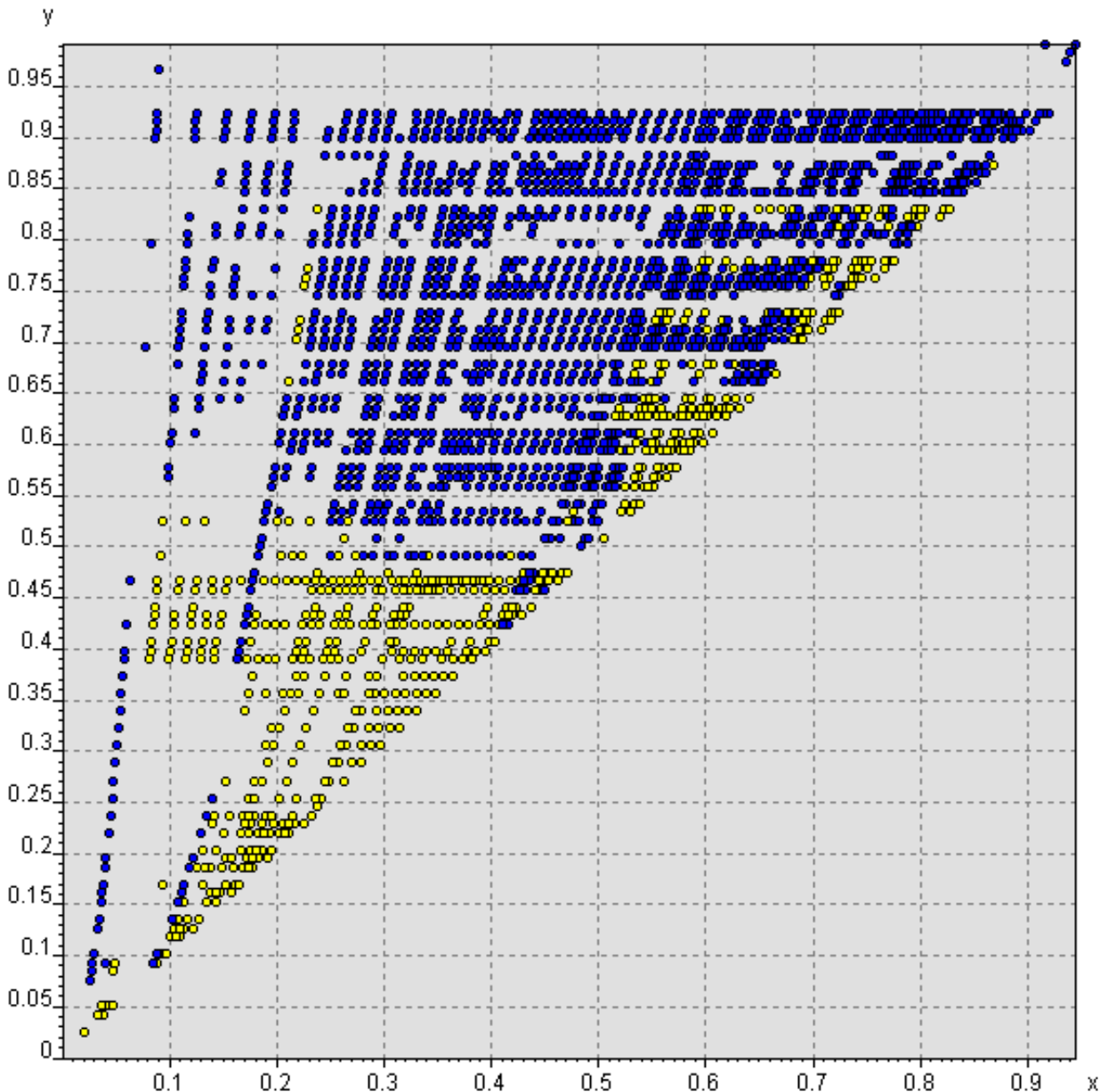
Fig. 17 Separation of 2318 binary systems into compound formers (●) and non-formers (○) based on a plot of max. [(PN$_{ME}$ (A) / (PN$_{ME}$)$_{max}$, PN$_{ME}$ (B) / (PN$_{ME}$)$_{max}$)] (y-axis) versus PN$_{ME}$ product [PN$_{ME}$ (A) / (PN$_{ME}$)$_{max}$ * PN$_{ME}$ (B) / (PN$_{ME}$)$_{max}$] (x-axis) using Meyer's Periodic System.

cF24, 227' (Pauling File prototype notation [26,27]), which are very close to a 'binary' close-packed structure.

In Figs. 14-18 the most efficient operators seem to be difference and maximum. It is a peculiarity of ternary systems that for the feature max.(EP (A,B,C)), only the value of EP (A), of the chemical element A with the largest EP value is taken into account. The chemical elements B and C are irrelevant. For the difference: |(EP (A) – EP (B)| + |(EP (A) – EP (C)| + |(EP (B) – EP (C)| (equal to EP$_{max}$ (A,B,C) – EP$_{min}$ (A,B,C)) only the maximum and minimum values of EP are taken into account. The intermediate value EP appear to be irrelevant.

Conclusions

This work revealed the following new knowledge:
1) An adequate description of the derived elemental-property parameters requires the introduction of the periodic number PN in addition to the well-established atomic number AN. AN and PN represent fundamental elemental-property parameters which are independent from each other.
2) The derived elemental-property parameters atomic size SZ$_a$, and its reciprocal, the atomic reactivity RE$_a$, can be expressed as functions of AN and PN. Other elemental-property parameter functions, like the mass density, can be described by combination of one or
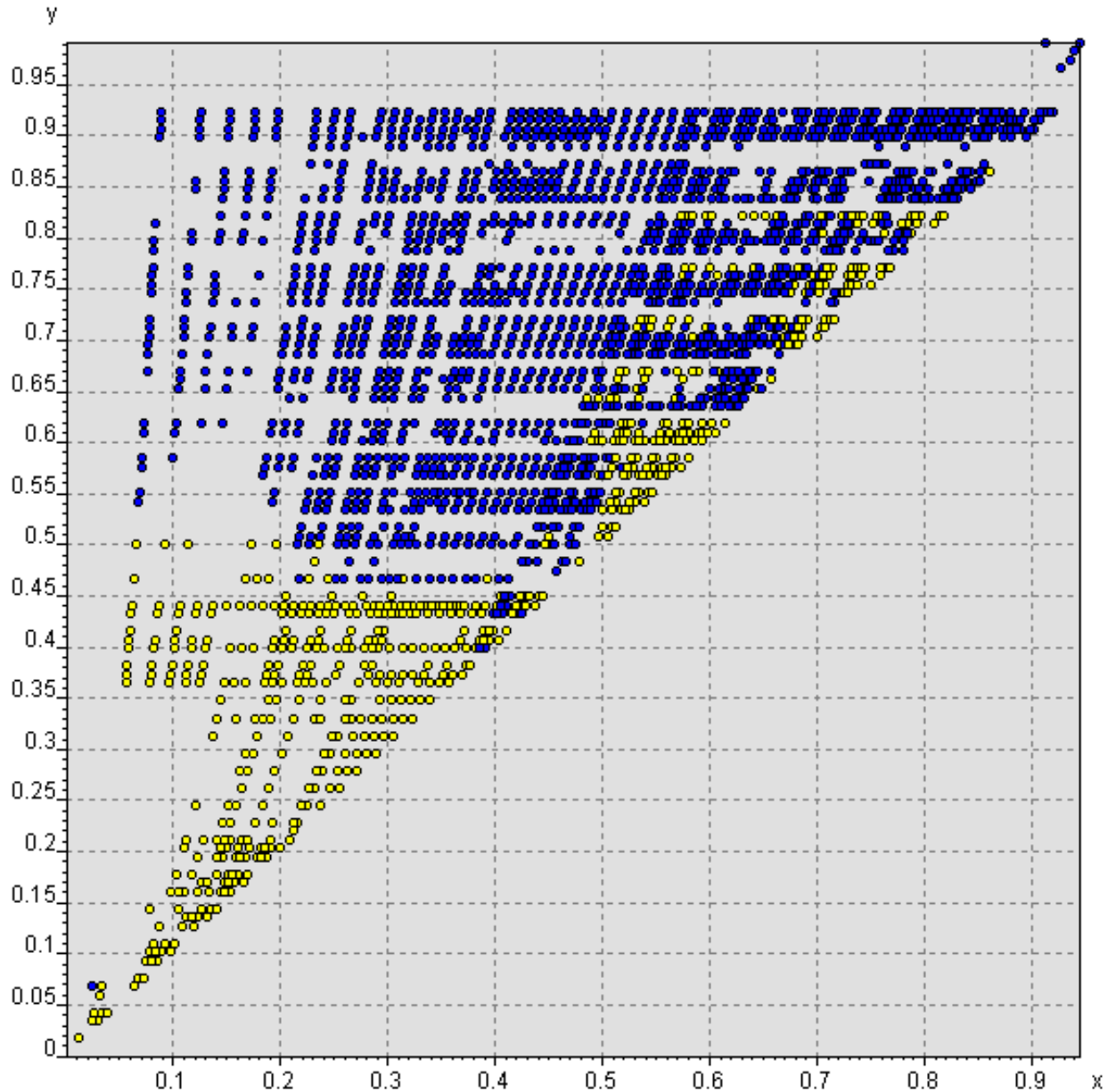
Fig. 18 Separation of 2318 binary systems into compound formers (●) and non-formers (○) based on a plot of max. [(PN$_{MD}$ (A) / (PN$_{MD}$)$_{max}$, PN$_{MD}$ (B) / (PN$_{MD}$)$_{max}$)] (y-axis) versus product [PN$_{MD}$ (A) / (PN$_{MD}$)$_{max}$ * PN$_{MD}$ (B) / (PN$_{MD}$)$_{max}$] (x-axis), using Mendeleyev's Periodic System.

more patterns (for the mass density it is a combination of atomic number pattern and atomic size pattern).

3) The result of equation (2), the frequently used RE$_a$ = k$_{SZ}$ k$_{RE}$ (SZ$_a$)$^{-1}$, is most outstanding. This means that the atomic reactivity (different electronegativity scales belong to this pattern group) of an element is the reciprocal value of its atomic size. This may be the reason why the 'electronegativity concept' was never really accepted in physics. Nevertheless in chemistry its ability to reflect the reactivity of the elements with each other is accepted and frequently used. To our knowledge the result that the atomic reactivity is

simply the reciprocal of the corresponding atomic size is not found in the literature.

4) On the example of the problem of separating formers from non-formers in binary, ternary and quaternary systems, we verify and reformulate our previous postulate: "Structure-sensitive properties of materials are quantitatively described by the elemental-property parameters of the constituent chemical elements". The reformulation of the postulate results in: "Structure-sensitive material properties are quantitatively described by the elemental-property parameters AN and PN (or simple

mathematical functions of them) of the constituent chemical elements". This generalization is an important step to strategically explore structure-sensitive properties of materials.

5) It is worth mentioning some observations in context with the noble gases (= inert gases). It is known that noble gases create compounds only in very exceptional cases. Figs. 17 and 18 show that the systems containing noble gases are all located in the compound-forming area. The eight listed elemental-property parameters belonging to the atomic reactivity patterns show only values for the absolute electronegativity EN(abs), and the first ionization potentials IE(first). They follow the general trend within GN 18, like our calculated atomic reactivity RE_a. Analogous observations are made for the elemental-property parameters Rve(S), Rce(S), and R(VD), belonging to the atomic size, which are also in general agreement with our calculated atomic size SZ_a. Looking at the binaries it appears that, from the about 500 noble-gas-containing systems, only 10 systems have been investigated. In these few investigated systems the following seven compounds are known: XeO_3, $Ar(H_2)_2$, KrF_2, XeF_6, XeF_4, XeF_3, and XeF_2. Obviously the conditions of existence for these noble-gas-containing compounds are quite different from those of the majority of the compounds considered in this work (T = 298 K and atmospheric pressure). Nevertheless, the above-mentioned observations allow us to conclude that many more noble-gas-containing compounds can be expected.

Acknowledgements

References

[1] G. Busch, H. Schade, in: *Vorlesungen über Festkörperphysik*, Birkhäuser, 1973.

[2] G.V. Samsonov, in: *Handbook of the Physicochemical Properties of the Elements*, IFI/Plenum Data Corporation, New York, 1968.

[3] J.C. Slater, in: *Theory of Alloy Phases*, American Society for Metals, Cleveland, OH, 1956.

[4] *L. Meyer's Periodic Table*, E.H. Sargent & Co, 1964.

[5] *D.I. Mendeleyev's Periodic Table*, in: F.R. de Boer, R. Boom, W.C.M. Mattens, A.R. Miedema, A.K. Niessen, in: *Cohesion in Metals*, Vol. 1, F.R. de Boer, D.G. Pettifor (Eds.), North-Holland, Amsterdam, 1988, p. 45.

[6] D.G. Pettifor, in: *Intermetallic Compounds, Principles and Practice*, Vol. 1, J.H. Westbrook, R.L. Fleischer (Eds.), Wiley, New York, 1994, pp. 419-438.

[7] A. Zunger, in: *Structure and Bonding in Crystals*, M. O'Keefe, A. Navrotsky (Eds.), Academic Press, New York, 1981, p. 73.

[8] E. Teatum, K. Gschneidner, J. Waber, LA-2345, U.S. Department of Commerce, Washington, DC, 1960.

[9] K. Schubert, private communication, 1990.

[10] F.R. de Boer, R. Boom, W.C.M. Mattens, A.R. Miedema, A.K. Niessen, in: *Cohesion and Structure*, Vol. 1, F.R. de Boer, D.G. Pettifor (Eds.), Elsevier Science Publishers B.V., Amsterdam, 1988, pp. 715-716.

[11] P. Villars, J.L.C. Daams, *J. Alloys Compd.* 197 (1993) 177-196.

[12] S.S. Batsanov, *Sov. Sci. Rev. B. Chem.* 15 (1990), 1-79.

[13] C. Kittel, *Einführung in die Festkörperphysik*, Oldenbourg Verlag, München, 1980, pp. 92-104.

[14] R.T. Sanderson, *Inorganic Chemistry*, Reinhold, New York, 1967, used in: G. Busch, H. Schade, *Vorlesungen über Festkörperphysik*, Birkhäuser, 1973.

[15] D.G. Pettifor, *J. Solid State Commun.* 51 (1984) 31.

[16] P. Villars, *Dissertation Nr. 6787*, ETH, Zürich, Switzerland, 1981.

[17] P. Villars, K. Brandenburg, M. Berndt, S. LeClair, A. Jackson, Y.-H. Pao, B. Igelnik, M. Oxley, B. Bakshi, P. Chen, S. Iwata, *J. Alloys Compd.* 317-318 (2001) 26-38.

[18] P. Villars, K. Cenzual, J. Daams, Y. Chen, S. Iwata, *J. Alloys Compd.* 367 (2004) 167-175.

[19] P. Villars, in: *Intermetallic Compounds, Principles and Practice*, Vol. 1, J.H. Westbrook, R.L. Fleischer (Eds.), Wiley, New York, 1994, pp. 227-275.

[20] J.L.C. Daams, in: *Intermetallic Compounds, Principles and Practice*, Vol. 1, J.H. Westbrook, R.L. Fleischer (Eds.), Wiley, New York, 1994, pp. 363-383.

[21] *ICSD*, FIZ/NIST, Fachinformations Zentrum Karlsruhe, Germany, 1999.

[22] P. Villars, L.D. Calvert, *Pearson's Handbook of Crystallographic Data for Intermetallic Phases*, 2nd Ed., Vols. 1-4, ASM International, Materials Park, OH, 1991.

[23] P. Villars, *Pearson's Desk Edition*, Vols. 1-2, ASM International, Materials Park, OH, 1997.

[24] T.B. Massalski, H. Okamoto, *Binary Alloy Phase Diagrams*, 2nd Ed., ASM International, Materials Park, OH, 1994, and on CD-ROM, 1998.

[25] P. Villars, A. Prince, H. Okamoto, *Handbook of Ternary Alloy Phase Diagrams*, Vols. 1-10, ASM International, Materials Park, OH, 1994, and on CD-ROM, 1996.

[26] P. Villars, K. Cenzual, J. Daams, F. Hulliger, T. Massalski, H. Okamoto, K. Osaki, A. Prince, *Pauling File, Binaries Edition*, on CD-ROM, ASM International, Materials Park, OH, 2002.

[27] P. Villars, K. Cenzual, H. Okamoto, J. Daams, K. Osaki, A. Prince, *"Pauling File (LPF)", Multinaries Edition* (in preparation).

[28] K. Brandenburg, M. Berndt, P. Villars, *"DISCOVERY"*, Crystal Impact, Bonn, Germany, 2000.