RARE-EARTH-COBALT INTERMETALLIC COMPOUNDS*)

by K. H. J. BUSCHOW

Abstract
A number of rare-earth-cobalt systems were investigated by X-ray diffraction, thermal analysis and metallography. Particular features of these binary systems and those already known in the literature are discussed. The number of stable compounds observed in the rare-earth-cobalt systems varies from six to eight. The structures of most of the compounds are size-dependent. For the trivalent rare-earth elements only the compound $R_3Co$ exists throughout the whole series and undergoes no change of structure. The other compounds, among which $R_4Co_3$, $RCO_2$, $RCO_3$, $R_2Co_7$, $RCO_5$ and $R_3Co_{17}$, either become unstable with respect to the adjacent phases or change their structure or stoichiometry when the radius ratio is varied. For the series $R_2Co_7$ and $R_2Co_{17}$ two types of structures were obtained by a modification of the heat treatment. Most of the structures observed for the Co-rich compounds were found to be fairly insensitive to changes in the valence-electron concentration.

1. Introduction
The number of intermetallic compounds in rare-earth-cobalt systems reported in the literature ranges from six to eight. Each of these binary systems contains a compound near or slightly beyond 84 at.% Co which is of the hexagonal CaZn$_2$ (CaCu$_7$) structure type. The majority of the other compounds observed in each system, i.e. compounds of composition $R_2Co_{17}$, $R_2Co_7$, $RCO_3$ and even $RCO_2$, have a crystal structure which can be derived in a relatively simple manner from the CaZn$_2$ structure type. In this investigation an attempt is made to determine how the relative stability of the observed compounds changes if one varies the metallic radius of the R-component. As the metallic radius of the rare-earth elements varies rather slowly within the lanthanide series it will be sufficient for this purpose to consider only some of the rare-earth-cobalt systems. We will therefore compare only eight binary systems. Some of these systems have already been described in the literature, and some of them have been reinvestigated.

A special problem in the study of rare-earth-cobalt systems is presented by the concentration range between 30 and 40 at.% Co. In this region almost all systems investigated so far have a rather deep eutectic. This is also the region where the phase diagrams show marked differences. It is not unlikely that the method of sample preparation is rather critical for compounds occurring close

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to the eutectic composition. This aspect has therefore been given some special consideration.

2. Experimental

The cobalt and the rare-earth metals used in the present study were of 99.9% and 99.99% purity. The samples were prepared by arc-melting. The thermal analysis was performed in a purified argon atmosphere by means of an induction furnace. We employed calibrated Pt/Pt–Rh thermocouples which were protected by tubes of recrystallized alumina and which remained immersed in the sample during cooling and heating. The X-ray diagrams were obtained from powdered samples on a Philips X-ray powder diffractometer (PW 1050/30) with CuKα radiation using an X-ray monochromator. For the microscopic investigation standard techniques were used.

2.1. Lanthanum–cobalt

Six intermetallic compounds have been observed in this system. The constitutional diagram is shown in fig. 1. A detailed description has been given in ref. 1. In view of the compounds of the form R₄Co₃ reported recently by Berthet-Colominas et al. 2) we tried to re-index the X-ray pattern obtained for the phase LaₐCo which occurs in this concentration region. It appeared, however, that the phase LaₐCo does not belong to the Ho₄Co₃ structure type.
2.2. Cerium–cobalt

We observed six intermediate phases in this system. Microscopic investigation showed that only the compound \( \text{Ce}_{24}\text{Co}_{11} \) melts congruently and is surrounded by two eutectics occurring at 23.5 at.\% Co and 35 at.\% Co. In previous investigations of this system (see ref. 3) a \( \text{CeCo}_4 \) phase has been reported, whereas no indication was obtained of a compound of composition \( \text{Ce}_2\text{Co}_{17} \). As suggested already by Gschneidner 3) the true stoichiometric composition of the compound reported as \( \text{CeCo}_4 \) is \( \text{Ce}_2\text{Co}_{7} \), which is in agreement with the present results, see fig. 2. The thermal arrests due to the peritectic melting of the phase \( \text{Ce}_2\text{Co}_{17} \) have probably been overlooked as the present thermoanalytical data show that they are very close to those of the phase \( \text{CeCo}_5 \). Apart from slight differences in the concentrations at which the two eutectics occur, our results for the Ce-rich part of the diagram agree closely with the results of Ellinger et al. 4).

![Fig. 2. Phase diagram of the Ce–Co system.](image)

2.3. Samarium–cobalt

This system contains seven intermetallic compounds. It has been described in more detail in ref. 5. The phase diagram is shown in fig. 3. Recent investigations of the Gd–Co system 6) have shown that for \( \text{Gd}_2\text{Co}_7 \) a hexagonal and a rhombohedral structure exists. This is also the case for \( \text{La}_2\text{Co}_7 \) and should therefore also apply to \( \text{Sm}_2\text{Co}_7 \). The missing rhombohedral modification has now indeed been observed in quenched samples. The rhombohedral modification, however, was always accompanied by large amounts of the hexagonal form; annealing at various temperatures hardly changed the amounts at which
the two forms occurred in the sample. This is probably closely connected with the heavily twinned microstructure observed for samples of this composition.

2.4. Gadolinium–cobalt

A description of this system is given in ref. 6. The phase diagram, which contains eight intermetallic compounds, is shown in fig. 4. Previous investiga-
2.5. Dysprosium–cobalt

This system has been investigated by Wood and Conard\(^7\), predominantly by electron-microprobe methods. They concluded that there exist at least seven intermediate phases and at least three eutectics. Thermoanalytical measurements were carried out by them only in the Dy-rich region up to 800 °C approximately. In the present investigation thermoanalytical measurements have been performed in the whole concentration region. We also reinvestigated this system by means of X-ray diffraction and metallography. We were able to confirm the existence of the seven compounds reported by Wood and Conard, although we observed three of them to occur at a slightly different Co concentration: Dy\(_2\)Co, DyCo\(_{1.2}\) and Dy\(_2\)Co\(_{1.7}\) instead of Dy\(_3\)Co\(_2\), DyCo\(_5\) and DyCo\(_9\).

The compound Dy\(_x\)Co is isotypic with the compound Gd\(_x\)Co\(^7\); the value for \(x\) is close to 4/7; its structure is still unknown. From the results of metallography, X-ray diffraction and thermoanalysis a tentative phase diagram was constructed which is shown in fig. 5. This diagram also contains the phase Dy\(_4\)Co\(_3\). The presence of this phase could not be detected by metallography and X-ray diffraction on as-cast samples, nor on samples annealed at 700 °C. Single-phase samples of the compound Dy\(_4\)Co\(_3\) could be obtained, however, by splat-cooling. No decomposition of this phase was observed on keeping the splat-cooled sample for several hours at 700 °C and lower temperatures; this

![Fig. 5. Phase diagram of the Dy–Co system.](image-url)
indicates that Dy₄Co₃ is probably not metastable. The eutectic near 40 at. % Co was clearly shown by X-ray diffraction and metallography to consist of the phases DyₓCo and DyCo₂. We did not observe thermal arrests in the range 40–60 at. % Co which could possibly be identified with the peritectic melting point of the phase Dy₄Co₃. It is therefore concluded that Dy₄Co₃ is formed by a peritectoid reaction from the two adjacent phases, i.e. from DyₓCo and DyCo₂. Once these two phases have crystallized from the melt, diffusion proceeds probably too slowly to give rise to the formation of observable amounts of the compound Dy₄Co₃, even after annealing for several days at 700 °C and lower temperatures. It is seen in fig. 5 that the compound Dy₄Co₃ occurs quite close to the eutectic composition. Regions near a eutectic very likely give rise to pronounced supercooling, especially when the melt is cooled rapidly, as for instance by splat-cooling. In that case solidification may start at a temperature below the peritectoid temperature of the compound Dy₄Co₃, so that this phase crystallizes primarily. This would explain why normal arc-casting techniques, which involve a slower cooling rate than splat-cooling, may fail to give rise to a compound Dy₄Co₃.

2.6. Holmium-cobalt

A detailed description of this binary system has been given elsewhere ⁸). The number of intermediate phases, their stoichiometry and their crystal structure are exactly the same as in the Dy-Co system except for the compound with CaZn₅ structure, which occurs at a slightly higher Co content. The two constitutional diagrams are also similar: the compounds HoₓCo, HoCo₃ and Ho₂Co₁₇ melt congruently and the eutectics occur at nearly the same Co concentration. The compound Ho₄Co₃ is formed peritectoidally from HoₓCo and HoCo₂ approximately 20 °C below the HoₓCo–HoCo₂ eutectic temperature. It can be formed directly from the melt by rapid quenching of the melt, involving supercooling. The Ho–Co phase diagram is shown in fig. 6.

2.7. Erbium-cobalt

In a previous investigation ⁹) we did not observe a compound of the form Er₄Co₃ in this binary system. In addition we encountered some difficulties in interpreting the results of thermal analysis and metallography in the concentration region from 20 to 40 at. % Co. The investigation of the binary systems described above has made it clear that especially in this region supercooling may occur. We therefore reinvestigated this concentration region. It was found that the compound ErₓCo melts congruently and that the rather finely divided Co-rich eutectic occurring at approximately 33 at. % Co has been overlooked. The thermal analysis in this concentration region was repeated with larger samples and at slower cooling rates. The compound ErₓCo melts at 810 °C; the temperatures of the adjacent eutectics are 800 °C and 795 °C, respectively.
The weak thermal arrest at 840 °C reported formerly proved to be absent, which agrees with the observations of the X-ray-diffraction measurements that the compound Er₃Co does not show a phase transformation. The phase Er₄Co₃ was not present in detectable amounts in as-cast samples of this composition, nor in samples annealed for 2 weeks at 700 °C. Splat-cooled samples on the other hand contained large amounts of this phase together with small amounts
of the phases $\text{ErCo}_2$ and $\text{Er}_x\text{Co}$. The phase $\text{Er}_4\text{Co}_3$ did not disappear on annealing the splat-cooled sample 24 h at 700 °C. We also kept the splat-cooled sample several hours at 780 °C and then cooled it quickly. In this case we observed a decrease of the amount of the phase $\text{Er}_4\text{Co}_3$ together with an increase of the phases $\text{ErCo}_2$ and $\text{Er}_x\text{Co}$. It is concluded therefore that near 40 at. % Co a similar situation exists as described above for the Dy–Co system but that the $\text{Er}_4\text{Co}_3$ peritectoid reaction temperature is appreciably below the $\text{Er}_x\text{Co}$–$\text{ErCo}_2$ eutectic temperature. The Er–Co phase diagram is shown in fig. 7.

2.8. Yttrium–cobalt

Although yttrium is a III-A element it very closely resembles the lanthanide elements and has a metallic radius nearly equal to that of Gd. Binary systems in which one of the components is Y therefore show a very close resemblance to binary systems in which one component is one of the heavy rare-earth elements. The Y–Co system has been investigated by Pelleg and Carlson \(^{10}\) and by Strnat et al. \(^{11}\). At some points the two reported constitutional diagrams are at variance. We therefore reinvestigated this binary system by X-ray diffraction, metallography and thermal analysis. From the results a phase diagram was constructed (fig. 8). Apart from minor differences in some of the melting temperatures the Co-rich part is in fair agreement with the results reported by Strnat et al. A homogeneity region has been reported by these authors only for the phase $\text{Y}_2\text{Co}_{17}$. In addition to $\text{Y}_2\text{Co}_{17}$ we observed also a homogeneity region for the phase $\text{YCo}_5$ at high temperatures with limit compositions.
Fig. 9. Micrograph of a sample $\text{YCo}_{4.75}$ which after annealing was quenched from 1200 °C; etched with $\text{HNO}_3$ aq.

Fig. 10. Micrograph of a sample $\text{YCo}_{4.75}$ which after annealing was quenched from 1300 °C; etched with $\text{HNO}_3$ aq.

Fig. 11. Micrograph of a sample $\text{YCo}_{4.9}$ which after annealing was quenched from 1200 °C; etched with $\text{HNO}_3$ aq.
tions $\text{YCo}_{4.9}$ and $\text{YCo}_6$. A homogeneity region for this phase has also been reported recently by Schweizer and Tasset $^{22}$ with limit compositions $\text{YCo}_{4.75}$ and $\text{YCo}_6$. According to our results the cobalt deficiency of the compound $\text{YCo}_5$ is less pronounced. This is shown most clearly by conventional metallography, which method is very likely more accurate than X-ray diffraction and X-ray fluorescence in concentration regions where compounds occur close together. Figures 9–11 show micrographs of the samples $\text{YCo}_{4.75}$ and $\text{YCo}_{4.9}$ which first were homogenized in vacuum at 1000 °C for 10 days and subsequently were kept at 1200 °C and 1300 °C for 30 h followed by rapid quenching in water. It is seen that for the sample $\text{YCo}_{4.75}$ appreciable amounts of the phase $\text{Y}_2\text{Co}_7$ are still present (dark regions) even at 1300 °C which is close to the melting point.

For the compounds $\text{YCo}_3$ and $\text{Y}_2\text{Co}_7$ we observed a different structure type than reported in ref. 11 (see sec. 4). Except for the structure of the compound $\text{Y}_3\text{Co}$ (see sec. 4) our results for the Y-rich part (0–30 at. % Co) compare favourably with those of Pelleg and Carlson and with those of Strnat et al. The agreement for Co concentrations between 30 and 60 at. % Co is only a very rough one. There are indications that in addition to the thermal arrests observed in this investigation at 725 °C there is a second thermal arrest lying very close to it; the vicinity of the eutectic makes its observance rather difficult. Our X-ray data point to the existence of three phases in this concentration region. For two of these phases we could not reach a conclusion as to the stoichiometry and the structure of the corresponding compounds and their relation to the observed thermal arrests. One of the phases could be identified, however, as the hexagonal compound $\text{Y}_4\text{Co}_3$ whose structure was reported in ref. 2. We obtained this compound nearly as a single phase by annealing the arc-cast sample for 4 days at 700 °C. It was almost absent in as-cast and splat-cooled samples. This seems to indicate that $\text{Y}_4\text{Co}_3$ melts peritectically and that the situation is similar to that observed for $\text{Gd}_4\text{Co}_3$ rather than for $\text{Dy}_4\text{Co}_3$, $\text{Ho}_4\text{Co}_3$ and $\text{Er}_4\text{Co}_3$.

### 3. Survey of the phase diagrams

In figs 1 to 8 the constitutional diagrams of the R–Co systems are shown, arranged according to the decreasing size of the metallic radius of the rare-earth elements, including Y. In the figures the compounds are indicated as line compounds. It should be kept in mind, however, that appreciable homogeneity regions may exist in the Co-rich compounds especially $^{5,6,11}$.

Inspection of the figures shows that the stability of the group of compounds derived from the CaZn$_2$ type of structure is increasing steadily as the radius ratio ($r_R/r_{\text{Co}}$) becomes smaller. Within this group the compound $\text{RCO}_3$ seems to benefit more from the radius-ratio decrease than for instance the compound $\text{R}_2\text{Co}_{17}$. This seems plausible if one compares the structure of these compounds.
The structure of the compounds $\text{RCO}_3$ may be derived from the structure of the $\text{RCO}_5$ compounds by an ordered substitution of R atoms at Co sites $^{16}$. This substitution takes place in alternate $\text{RCO}_5$ unit cells and is accompanied by a slight layer shift. In view of the fact that the R atoms are much larger than the Co atoms this substitution should proceed more easily as the radius ratio becomes smaller. The structure of the compounds $\text{R}_2\text{Co}_{17}$ is derived from the $\text{RCO}_5$ structure by an ordered substitution of a pair of Co atoms at one third of all R sites. This latter substitution is less critical with respect to the radius ratio. With regard to the La–Co system we note that here the radius ratio is such that a compound of the cubic NaZn$_{13}$ type becomes stable. The phase $\text{La}_2\text{Co}_{17}$ is probably stable with respect to $\text{LaCo}_5$ and Co but not with respect to $\text{LaCo}_5$ and $\text{LaCo}_13$. If one thinks of the cubic Laves phase $\text{RCO}_2$ as being derived from the hexagonal $\text{RCO}_5$ compounds by a substitution of an R atom at a Co site in every $\text{RCO}_5$ unit cell, it becomes clear that the relative stability of the compounds $\text{RCO}_2$ should even be more dependent on the radius ratio than the compounds $\text{RCO}_3$. The fact that $\text{RCO}_2$ is absent in the La–Co system together with the compound $\text{RCO}_3$ is therefore not at all surprising.

The results obtained in this investigation have shown that the compound $\text{R}_4\text{Co}_3$ is obtained rather easily for $R = \text{Gd}$ but presents more difficulties for the other heavy rare-earth elements. The results described above suggest furthermore that in order to obtain this compound an increasing degree of supercooling is necessary as one proceeds from Gd to Er. In this connection it is interesting to mention the results of Berthet-Colominas et al. $^2$ who obtained the compounds $\text{R}_4\text{Co}_3$ for nearly all heavy rare-earth elements without difficulty. These authors prepared their samples by levitation melting following by rapid quenching. Contrary to arc-casting and our method of splat-quenching $^{12}$) this method of alloy preparation starts from a melt which is not in contact with any solid material prior to quenching. It will therefore contain no or relatively few nuclei for solidification and may hence give rise to a large degree of supercooling.

The phase diagrams show furthermore that the heavy rare-earth elements for a compound $\text{R}_4\text{Co}$. These compounds are isotypic for $R = \text{Gd}, \text{Dy}, \text{Ho}$ and Er. It does not seem to be formed by yttrium and the light rare-earth elements. The compound $\text{R}_3\text{Co}$ on the other hand is formed by all rare-earth elements except for cerium, which seems to adopt an abnormal valency when combined with cobalt.

Finally we wish to mention the compounds formed by alloying Th and Co. Thorium is an actinide element; it has a metallic radius close to Gd and is therefore expected to give rise to intermetallic compounds similar to those observed for the rare-earth elements. It has one more valence electron, however, and is for this reason very well suited for studying the influence of electron concentration on compound formation.
The Th–Co system has been investigated by Thomson \(^{13}\)). The following compounds were observed: Th\(_7\)Co\(_3\), ThCo, ThCo\(_3\), ThCo\(_5\) and Th\(_2\)Co\(_7\). Some uncertainty was reported concerning the exact stoichiometry and structure of the compound ThCo\(_4\). The evidence cited in ref. 13 indicates a composition richer in Co than 75 at.\%, while moreover difficulties were encountered in indexing all the lines of the X-ray pattern. We reinvestigated this concentration region and only found evidence for the existence of a compound of composition Th\(_2\)Co\(_7\). We observed two modifications of this compound. The lattice constants of both modifications are reported below. No evidence was obtained for the existence of the compounds ThCo\(_3\) and ThCo\(_2\). This suggests that the stability of the compounds RCo\(_3\) and RCo\(_2\) discussed above is not only size-dependent but depends also on the electron concentration. For the compounds RCo\(_2\) this has already been discussed in ref. 14. With higher Co concentrations the enhancement in electron concentration due to the substitution of Th for R becomes gradually smaller. The stoichiometry and structure for the cobalt-rich Th compounds is virtually the same as those reported for the rare-earth elements of comparable size.

4. The crystal structures

R\(_2\)Co\(_{17}\)

The structure of rare-earth compounds of this composition is size-dependent. At small radius ratios the hexagonal Th\(_2\)Ni\(_{17}\) type is stable. When the radius ratio is increased this structure type occurs at elevated temperatures. The structure stable at room temperature is in this case the rhombohedral Th\(_2\)Zn\(_{17}\) type. The lattice constants of the R\(_2\)Co\(_{17}\) compounds have been given elsewhere \(^{12}\).

RCo\(_5\)

The crystal structure of these compound is of the hexagonal CaCu\(_5\) type. Strong deviations from stoichiometric composition may occur. This was first observed for the compound in which R is Er \(^9\)) and for which the Er deficiency has been interpreted as a replacement of Er by Co. A study of the variation of the lattice constants \(a\) and \(c\) upon an increase of the Co concentration within the homogeneity range of the compound SmCo\(_5\) soon revealed that the R deficiency in R–Co compounds with CaCu\(_5\) structure is more likely due to a replacement of R atoms by pairs of Co atoms \(^5\). This latter interpretation has successfully been applied since to account for the deviation of the stoichiometry for quite a number of CaCu\(_5\)-type rare-earth–cobalt compounds \(^6,8,22,23\)). In general a large homogeneity region is only observed at high temperatures. Within the homogeneity regions the lattice constants vary with Co content in a similar way as shown for GdCo\(_x\) in fig. 12 (in this case the homogeneity region extends from about \(x = 4·9\) to \(x = 6·0\)). At lower temperatures mos
RCO₅ compounds have no or at least only a rather small homogeneity region. The lattice constants for the CaCu₅-type rare-earth–cobalt compounds have been reported elsewhere.

For the light rare-earth elements the crystal structure assumed by these compounds is predominantly of the hexagonal Ce₂Ni₇ type described by Cromer and Larson. At smaller radius ratios the compounds adopt the rhombohedral Gd₂Co₇ structure described by Bertaut et al. The difference in stability of the two structure types, especially as regards the medium-sized rare-earth elements, seems to be very small. The lattice constants obtained in these investigations are summarized in table I.

R₂Co₇

For the light rare-earth elements the crystal structure assumed by these compounds is predominantly of the hexagonal Ce₂Ni₇ type described by Cromer and Larson. At smaller radius ratios the compounds adopt the rhombohedral Gd₂Co₇ structure described by Bertaut et al. The difference in stability of the two structure types, especially as regards the medium-sized rare-earth elements, seems to be very small. The lattice constants obtained in these investigations are summarized in table I.

RCO₃

It was shown recently by Bertaut et al. that the rare-earth compounds of this composition crystallize in the rhombohedral PuNi₃ structure. The lattice constants arrived at in the present investigation are summarized in table I. They show good agreement with the values reported in ref. 17. It is seen that the value observed for the a-axis of the compound CeCo₃ is smaller than any of the other values listed for the compounds RCo₃. Evidently Ce assumes a valency in this compound which is close to 4. In doing so its metallic radius heavily decreases and compares favourably with that of (tetravalent) Th. It is understandable therefore why CeCo₃ exists and why ThCo₃ does not exist.
<table>
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**TABLE II**

Lattice constants for the compounds R₄Co₃

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The X-ray patterns of the splat-cooled samples of this composition have been indexed according to the unit-cell dimensions of the Ho$_4$Co$_3$ structure described by Berthet-Colominas et al. The lattice constants collected in table II are in satisfactory agreement with their results.

Compounds of this composition assume the orthorhombic Al$_3$Ni type of structure. A structure determination has been given by Cromer and Larson for La$_3$Co. In order to obtain a better insight into the intensity distributions of the X-ray reflections of R$_3$Co compounds in which R is a heavy rare-earth metal, we carried out a structure determination for Ho$_3$Co. This led to a reindexing of the numerous reflections of the R$_3$Co X-ray diagrams for some of these compounds. The lattice constants are collected in table III.

### TABLE III

Lattice constants for some rare-earth–cobalt compounds

| compound | lattice constants (Å) |  
| --- | --- | --- |
|  | a  | b  | c  |
| La$_3$Co | 7.277 | 10.020 | 6.575 |
| Pr$_3$Co | 7.143 | 9.780 | 6.410 |
| Nd$_3$Co | 7.107 | 9.750 | 6.386 |
| Sm$_3$Co | 7.055 | 9.605 | 6.342 |
| Gd$_3$Co | 7.031 | 9.496 | 6.302 |
| Tb$_3$Co | 6.985 | 9.380 | 6.250 |
| Dy$_3$Co | 6.965 | 9.341 | 6.233 |
| Ho$_3$Co | 6.920 | 9.293 | 6.213 |
| Er$_3$Co | 6.902 | 9.191 | 6.189 |
| Y$_3$Co | 7.026 | 9.454 | 6.290 |

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