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# Thermoelectric power of selected metals and binary alloy systems

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## ABSTRACT

The available experimental data and information on the thermoelectric power of the following ten binary alloy systems: aluminum-copper, aluminum-magnesium, copper-gold, copper-nickel, copper-palladium, copper-zinc, gold-palladium, gold-silver, iron-nickel, and silver-palladium, and of the following eleven metals: aluminum, copper, gold, iron, lead, magnesium, nickel, palladium, platinum, silver, and zinc have been compiled, critically evaluated, analyzed, correlated, and synthesized to generate reliable reference data (recommended values). The recommended values for each of most of the alloy systems are generated for 27 compositions: 0 (pure element), 0.5, 1, 3, 5, 10(5)95, 97, 99, 99.5, and 100% (pure element) covering the temperature range from near the absolute zero to above 1000 K for most of the alloy compositions.

## INTRODUCTION

The objective of this study has been to critically evaluate, analyze, correlate, and synthesize the available data and information on the thermoelectric power of ten selected binary alloy systems and their nine constituent elements, and to generate recommended values over the widest practicable ranges of temperature and alloy composition for each of the alloy systems. This study is a continuation of two similar studies on the thermal conductivity and electrical resistivity of the same ten binary alloy systems (Ho et al., 1978, 1983a).

The ten binary alloy systems selected are the systems of aluminum-copper, aluminum-magnesium, copper-gold, copper-nickel, copper-palladium, copper-zinc, gold-palladium, gold-silver, iron-nickel, and silver-palladium. These systems include all of the three different kinds of binary alloy systems: nontransition-metal plus nontransition-metal systems (aluminum-copper, aluminum-magnesium, copper-gold, copper-zinc, and gold-silver), nontransition-metal plus transition-metal systems (copper-nickel, copper-palladium, gold-palladium, and silver-palladium),

and a transition-metal plus transition-metal system (iron-nickel). Most of these alloy systems are among those for which the largest amounts of experimental data are available. However, even for these alloy systems serious gaps and voids still exist in the thermoelectric power data, as concerns dependence on both composition and temperature, and some of the available experimental data sets show large uncertainties or wide divergences.

For this thermoelectric power study, two more metallic elements, lead and platinum, have to be included in addition to the nine constituent elements. It is because most of the available data on the relative thermoelectric power were produced by the authors through measurements against lead, platinum, or copper, and most of the available data on the absolute thermoelectric power were calculated by the authors through conversion from their measured relative values using the then available "standard" values for the absolute thermoelectric power of lead, platinum, or copper.

Because the scale of the absolute thermoelectric power commonly used throughout the world from 1932 to about 1980 was in error, which affects most of the available thermoelectric power data in the entire worldwide literature, a revised (correct) scale of the absolute thermoelectric power from 0 to 1300 K was developed at CINDAS to replace the old scale. According to the revised scale, the available experimental data compiled from the literature were first carefully converted and then critically evaluated, analyzed, correlated, and synthesized to generate the recommended values.

Part of the results of this study - the recommended values for the absolute thermoelectric power of aluminum-copper, copper-nickel, and iron-nickel alloy systems - have been reported in graphical form (Ho et al., 1983b). The values for the remaining aluminum-magnesium, copper-gold, copper-palladium, copper-zinc, gold-palladium, gold-silver, and silver-palladium alloy systems are reported here graphically. Due to space limitations, it is not possible to report here the detailed tabulations of the recommended values, the compiled experimental data and information, the details of data analysis, correlation and synthesis, the considerations involved in arriving at the final assessment and recommendation, the comparison of the recommended values with the experimental data, the uncertainties in the recommended values for each alloy composition in different temperature ranges, etc., which will be available elsewhere (Ho et al., 1993).

## SCALE OF ABSOLUTE THERMOELECTRIC POWER

Borelius et al. (1932) established the first scale of absolute thermoelectric power through a complex indirect means of determining the Thomson coefficients of lead, tin, and a "silver-normal" alloy, which was an alloy of silver + 0.37 at.% gold + another (unknown) impurity. From the measured and derived Thomson coefficients in various ranges of temperature, they calculated the absolute thermoelectric power of lead and tin from 0 to 100 K and of the "silver-normal" alloy from 2 to 293 K, thus established for the first time the scale of absolute thermoelectric power. This 1932 scale of the absolute thermoelectric power has since been in use as international standard, though in 1958 Christian et al. (1958) modified the values of Borelius et al. for temperatures below 20 K. Despite the fact that the establishment of the first scale of absolute thermoelectric power was an

afterthought which was recognized as less than ideal by the authors, the work of Borelius et al., including the fundamental low-temperature measurement of the Thomson coefficient of the "silver-normal" alloy, survived unchallenged by investigators in the field until 1977.

Christian et al. (1958) made direct measurements relative to a $Nb_3Sn$ superconductor (mean transition temperature 17.92 K) of the absolute thermoelectric power of a zone-purified Pb sample that was "very much purer than the starting material which was already 99.999% chemically purified lead." Their determination of the absolute thermoelectric power of lead up to 18 K eliminated errors at these low temperatures in the scale of Borelius et al. which had resulted from the interpolation of the Thomson coefficient of tin between 7.2 and 20 K. The modified scale of Christian et al., which was based on their own measurements below 20 K and on the measurements of Borelius et al. from about 20 to 293 K, served as the standard for low-temperature thermoelectric measurements from 1958 to 1977.

In 1977 Roberts (1977) shook the foundations of the older scale of absolute thermoelectric power by reporting that the values for the absolute thermoelectric power of lead as determined by Borelius et al. and modified by Christian et al. were in error by as much as $0.3 \mu V\ K^{-1}$, or nearly 50%. Roberts had undertaken the long overdue low-temperature measurement of the Thomson coefficient of a 99.9999% pure lead sample, and calculated the absolute thermoelectric power of lead up to 350 K from the measured Thomson coefficients.

Immediately after getting Roberts' data for the absolute thermoelectric power of lead to 350 K in 1977, we at CINDAS were able to (1) generate the values for the absolute thermoelectric power of copper up to 350 K through the critical evaluation, analysis, and synthesis of the low-temperature data on the relative thermoelectric power of copper versus lead available from the literature, and (2) generate the values for copper up to 1300 K through the critical evaluation, analysis, and synthesis of the available high-temperature data on the Thomson coefficient of copper, the generation of recommended values for the Thomson coefficient of copper up to 1300 K, and the calculation of the absolute thermoelectric power of copper from the recommended values for the Thomson coefficient of copper. In turn, the values of the absolute thermoelectric power of platinum up to 1300 K were calculated from the relative thermoelectric power of platinum versus copper. Thus, based on Roberts' data for the absolute thermoelectric power of lead to 350 K and its own recommended values for the absolute thermoelectric power of copper to 1300 K and of platinum to 1300 K, CINDAS established a revised scale of absolute thermoelectric power to high temperatures, which has been used throughout this study.

It is interesting to note that, immediately after CINDAS' recommended values for the absolute thermoelectric power of copper and of platinum from 0 to 1300 K were presented at the Seventeenth International Thermal Conductivity Conference on 16 June 1981 (see Ho et al., 1983b), Dr. G. K. White from the audience informed us that Dr. R. B. Roberts had recently measured the Thomson coefficient of copper, among other measurements, and that his calculated values for the absolute thermoelectric power of copper over the temperature range from 273 to 900 K were reported in a manuscript which had been submitted for publication in Philosophical Magazine. Dr. White further said that even though Dr. Roberts' article was not yet available, a summary of Dr. Roberts' results was contained in a

letter sent to him, and he gave a copy of the letter to us for comparing the results. On comparison of Dr. Roberts' experimental data for the temperature range 273 to 900 K with our synthesized values for the absolute thermoelectric power of copper, we were very pleased to find that Dr. Roberts' data and our values were extremely close and almost identical, the differences, if any, being in the third significant digits. Thus, CINDAS' recommended values for the absolute thermoelectric power to high temperatures have been validated.

## ABSOLUTE THERMOELECTRIC POWER OF SELECTED BINARY ALLOY SYSTEMS AND THEIR CONSTITUENT ELEMENTS

The available experimental data and information on both the relative and the absolute thermoelectric power of the selected binary alloy systems and their constituent elements were compiled and carefully converted according to CINDAS' revised scale of absolute thermoelectric power. In the available data, serious gaps and voids exist as concerns dependence on both composition and temperature.

The resulting converted data for the absolute thermoelectric power were then critically evaluated, analyzed, correlated, and synthesized to generate a full field of internally-consistent recommended values for the absolute thermoelectric power. In analyzing and synthesizing the experimental data to generate recommended values, experimental data for both temperature dependence and composition dependence were utilized and the alloy phase diagram was constantly consulted. The phase boundaries between solid solutions and/or mechanical mixtures, the boundaries of magnetic transitions, the possibilities of forming intermetallic compounds, etc. were kept in mind, so as to be aware of any possible discontinuity or sudden change of slope in the thermoelectric power curves.

It is important to note that in this study the thermoelectric power data for ordered alloys and intermetallic compounds were ignored and the recommended values generated for the alloys are for those which are not ordered and have not been severely cold-worked or quenched.

Since the recommended values for the absolute thermoelectric power of aluminum-copper, copper-nickel, and iron-nickel alloy systems have already been reported by Ho et al. (1983b), the values for the remaining seven binary alloy systems are reported here.

### *Copper-gold alloy system*

The copper-gold alloy system forms a continuous series of solid solutions over the entire range of compositions. However, ordered structures are formed at temperatures below 663 K for compositions ranging from about 17.7 to 35.5 at.% Au and below 683 K for compositions ranging from about 35.5 to 83.5 at.% Au due to the formation of intermetallic compounds $Cu_3Au$, CuAu, and $CuAu_3$.

The recommended values for the absolute thermoelectric power of copper-gold alloy system are presented in Figure 1 for the temperature dependence of copper + gold (copper-rich) alloys, Figure 2 for the temperature dependence of gold + copper (gold-rich) alloys, and Figure 3 for the composition dependence of the entire copper-gold alloy system.

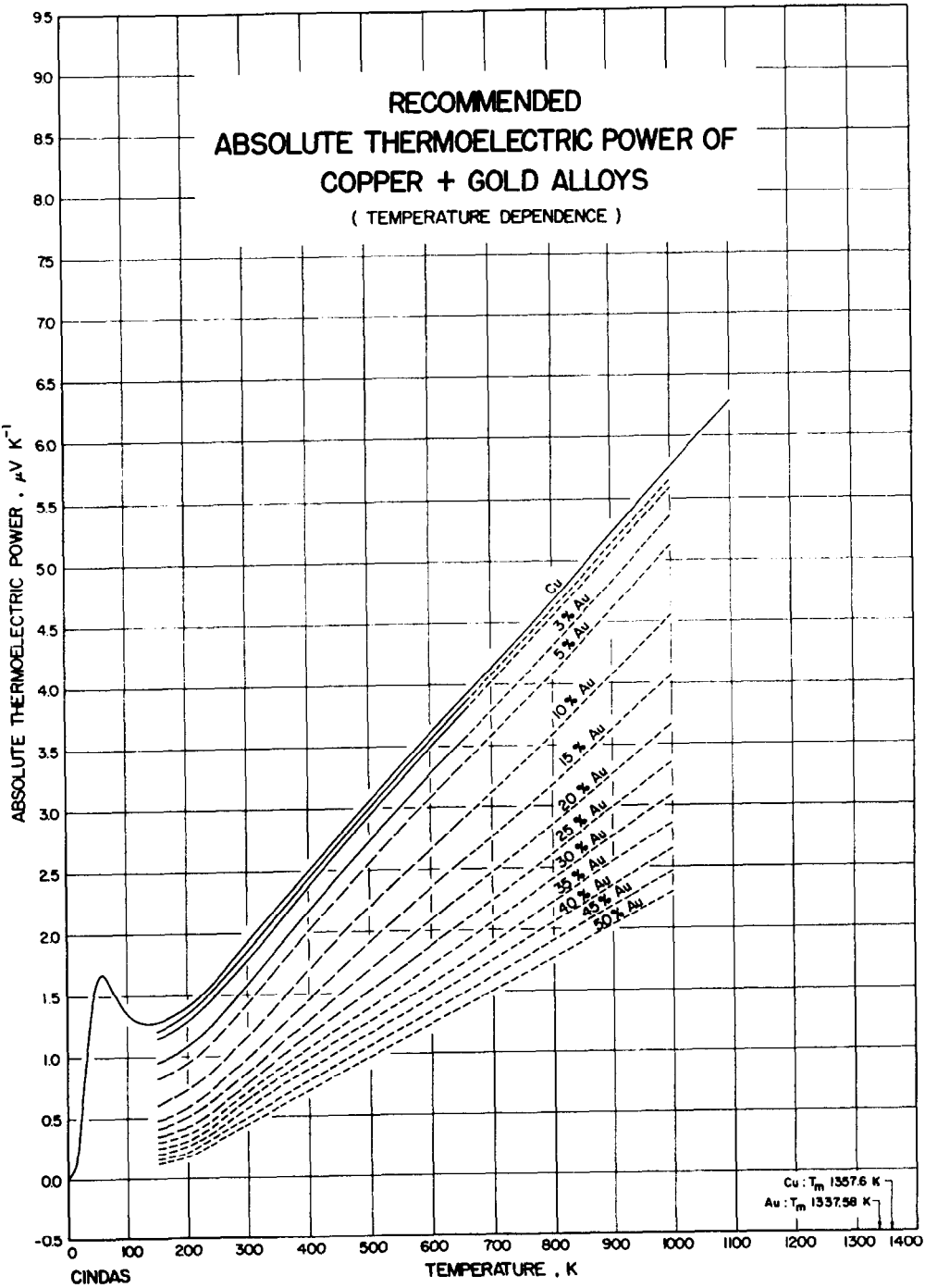



Fig. 1. Recommended absolute thermoelectric power of copper + gold alloys.

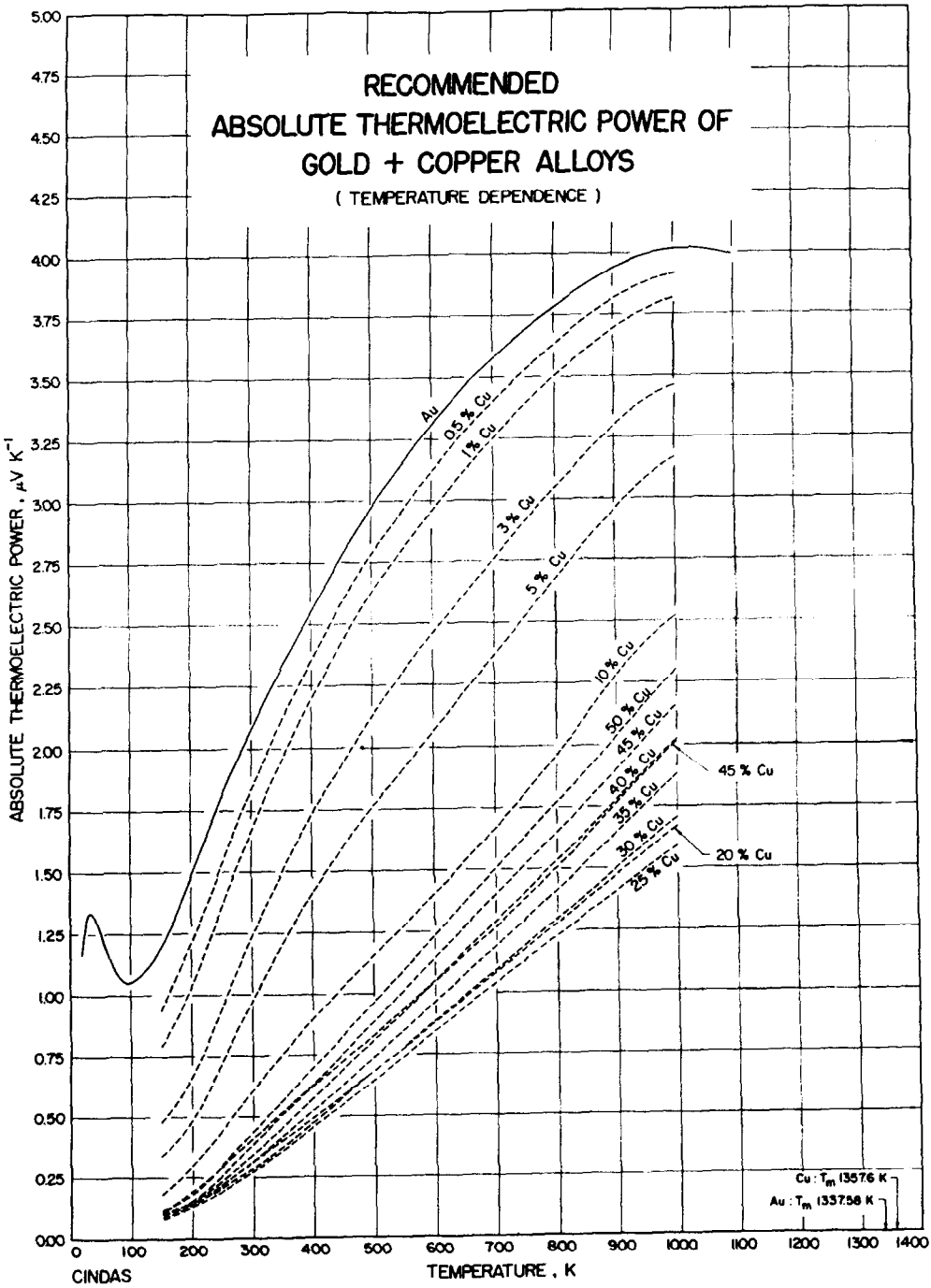



Fig. 2. Recommended absolute thermoelectric power of gold + copper alloys.

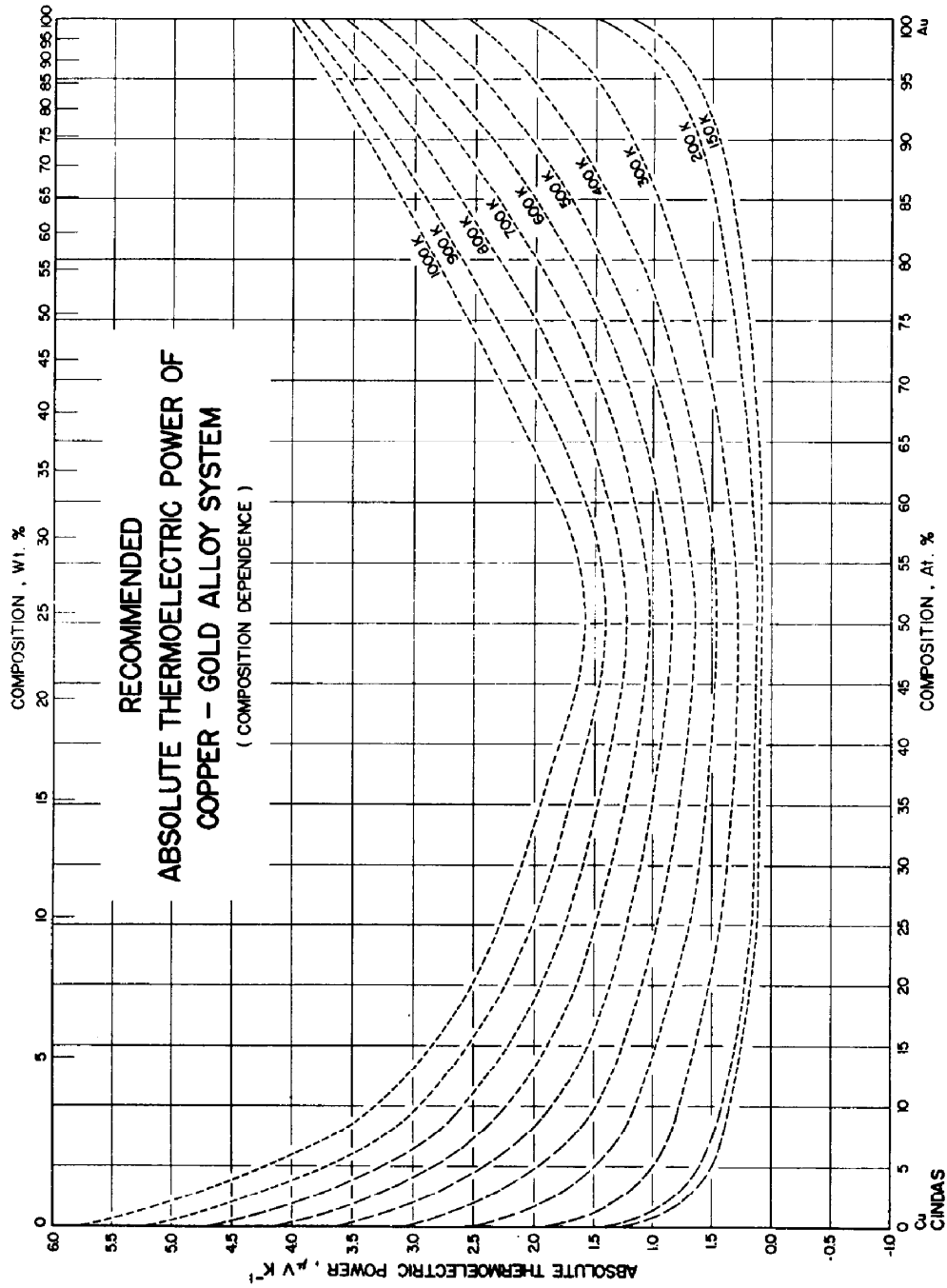



Fig. 3. Recommended absolute thermoelectric power of copper-gold alloy system.

*Copper-palladium alloy system*

At high temperatures the copper-palladium alloy system forms a continuous series of solid solutions over the entire range of compositions, with essentially random ordering of the two types of atoms over a face-centered cubic lattice. Below 875 K the situation becomes more complex, due to the appearance of phases with partial ordering of the atoms on various lattices, and of two-phase mixtures in certain ranges of temperature and composition.

The recommended values for the absolute thermoelectric power of copper-palladium alloy system are presented in Figure 4 for the temperature dependence of both copper + palladium alloys and palladium + copper alloys and Figure 5 for the composition dependence of the copper-palladium alloy system.

*Gold-palladium alloy system*

The gold-palladium alloy system forms a continuous series of solid solutions over the entire range of compositions at high temperatures. However, electron diffraction analyses and diffuse x-rays scattering studies have indicated the formation of ordered structures at lower temperatures.

The recommended values for the absolute thermoelectric power of gold-palladium alloy system are presented in Figure 6 for the temperature dependence of gold-palladium alloys, Figure 7 for the temperature dependence of palladium-gold alloys, and Figures 8 and 9 for the composition dependence of the entire gold-palladium alloy system.

*Gold-silver alloy system*

The gold-silver alloy system forms a continuous series of solid solutions over the entire range of compositions. Short-range order has been observed over a wide composition range, but long-range order has not been observed experimentally.

The recommended values for the absolute thermoelectric power of gold-silver alloy system are presented in Figure 10 for the temperature dependence of gold-silver alloys, Figure 11 for the temperature dependence of silver-gold alloys, and Figure 12 for the composition dependence of the entire gold-silver alloy system.

*Silver-palladium alloy system*

The preponderance of available evidence suggests that the silver-palladium alloy system forms a continuous series of solid solutions over the entire range of compositions. The evidence for the formation of two intermetallic compounds is inconclusive.

The recommended values for the absolute thermoelectric power of silver-palladium alloy system are presented in Figure 13 for the temperature dependence of silver-palladium alloys, Figure 14 for the temperature dependence of palladium-silver alloys, and Figures 15 and 16 for the composition dependence of the entire silver-palladium alloy system.

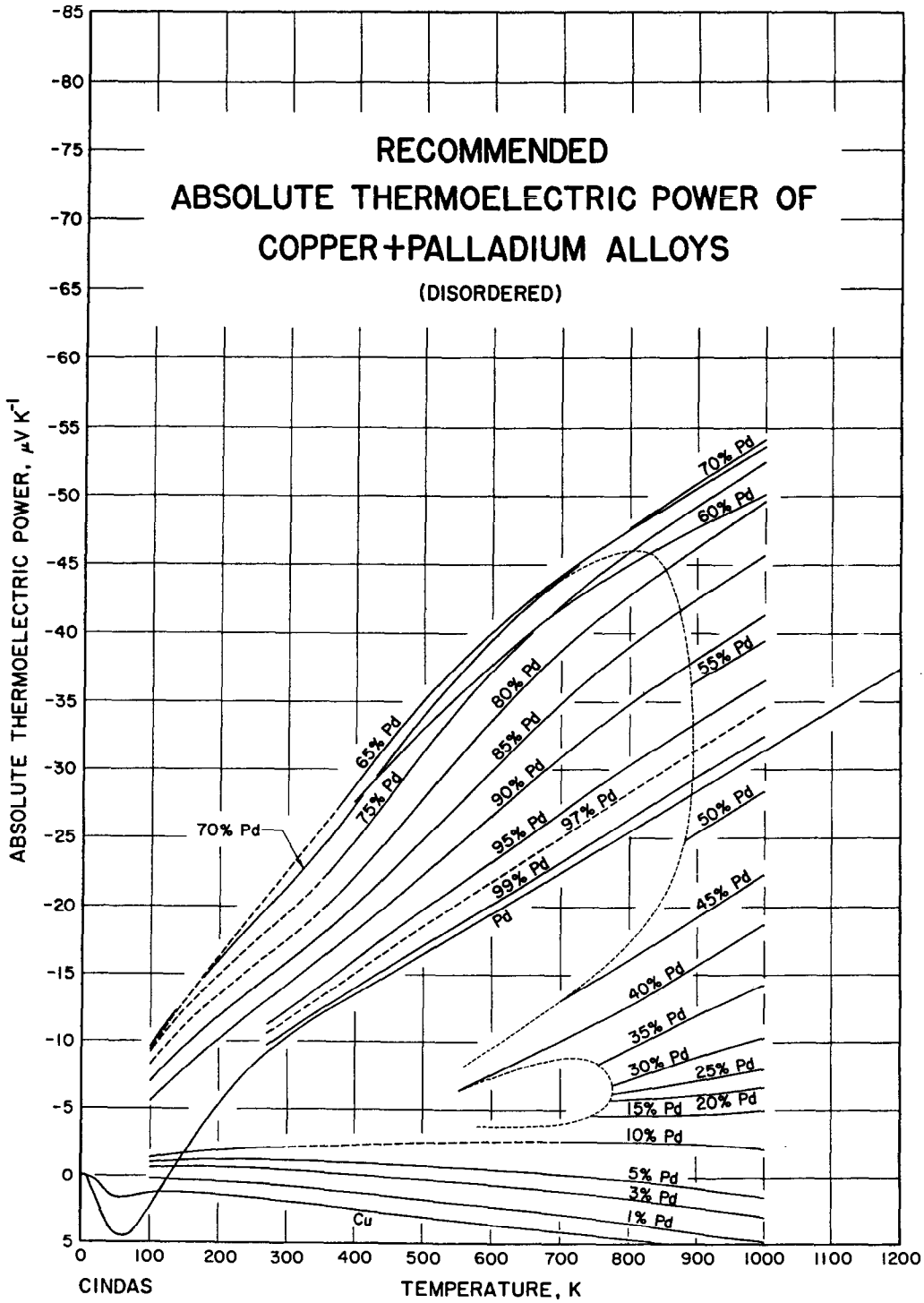
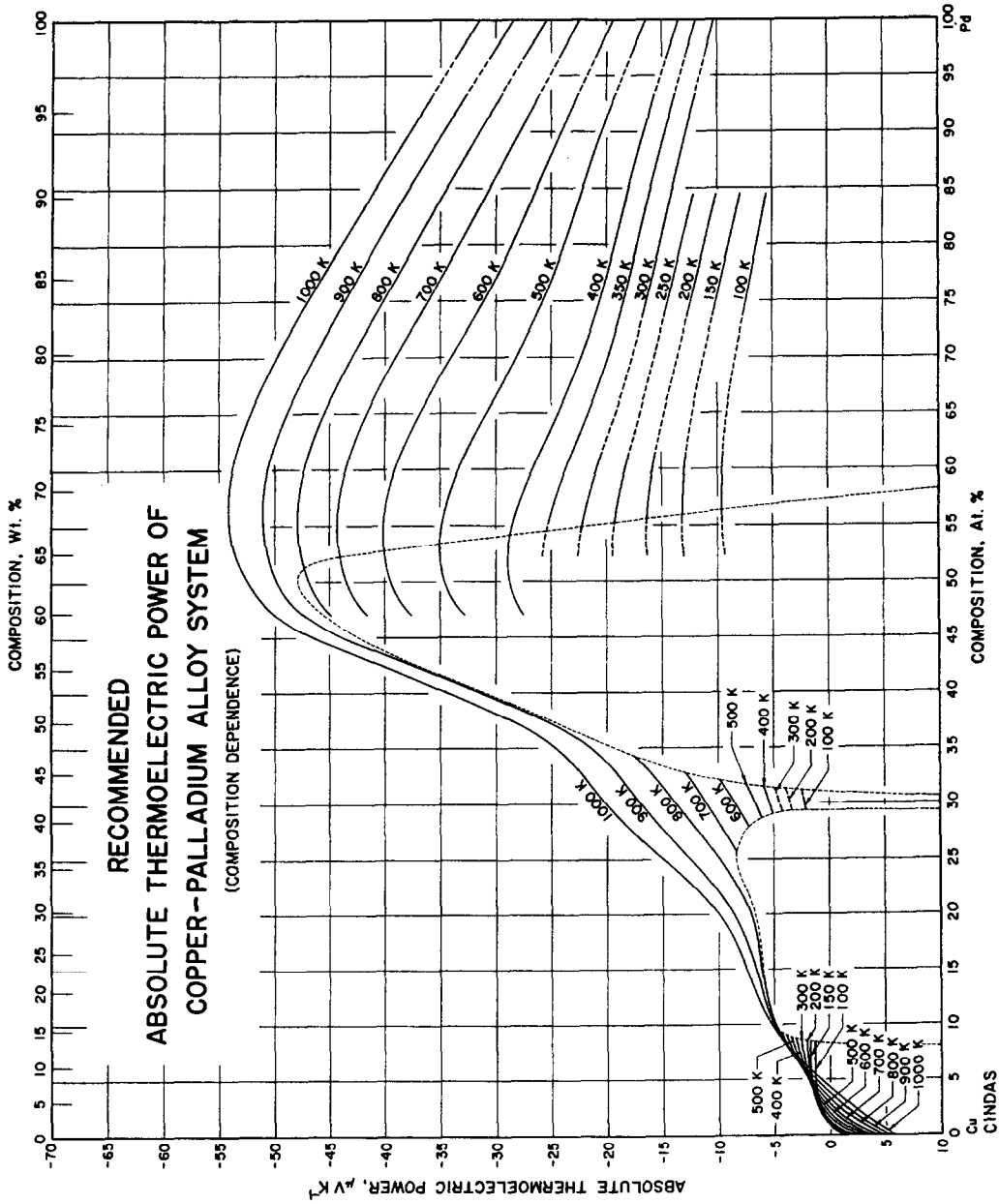



Fig. 4. Recommended absolute thermoelectric power of copper + palladium alloys.


RECOMMENDED
ABSOLUTE THERMOELECTRIC POWER OF
COPPER-PALLADIUM ALLOY SYSTEM
(COMPOSITION DEPENDENCE)
COMPOSITION, Wt. %
COMPOSITION, At. %
ABSOLUTE THERMOELECTRIC POWER, μV K⁻¹
Cu
CINDAS
Pd
1000 K
900 K
800 K
700 K
600 K
500 K
400 K
350 K
300 K
250 K
200 K
150 K
100 K

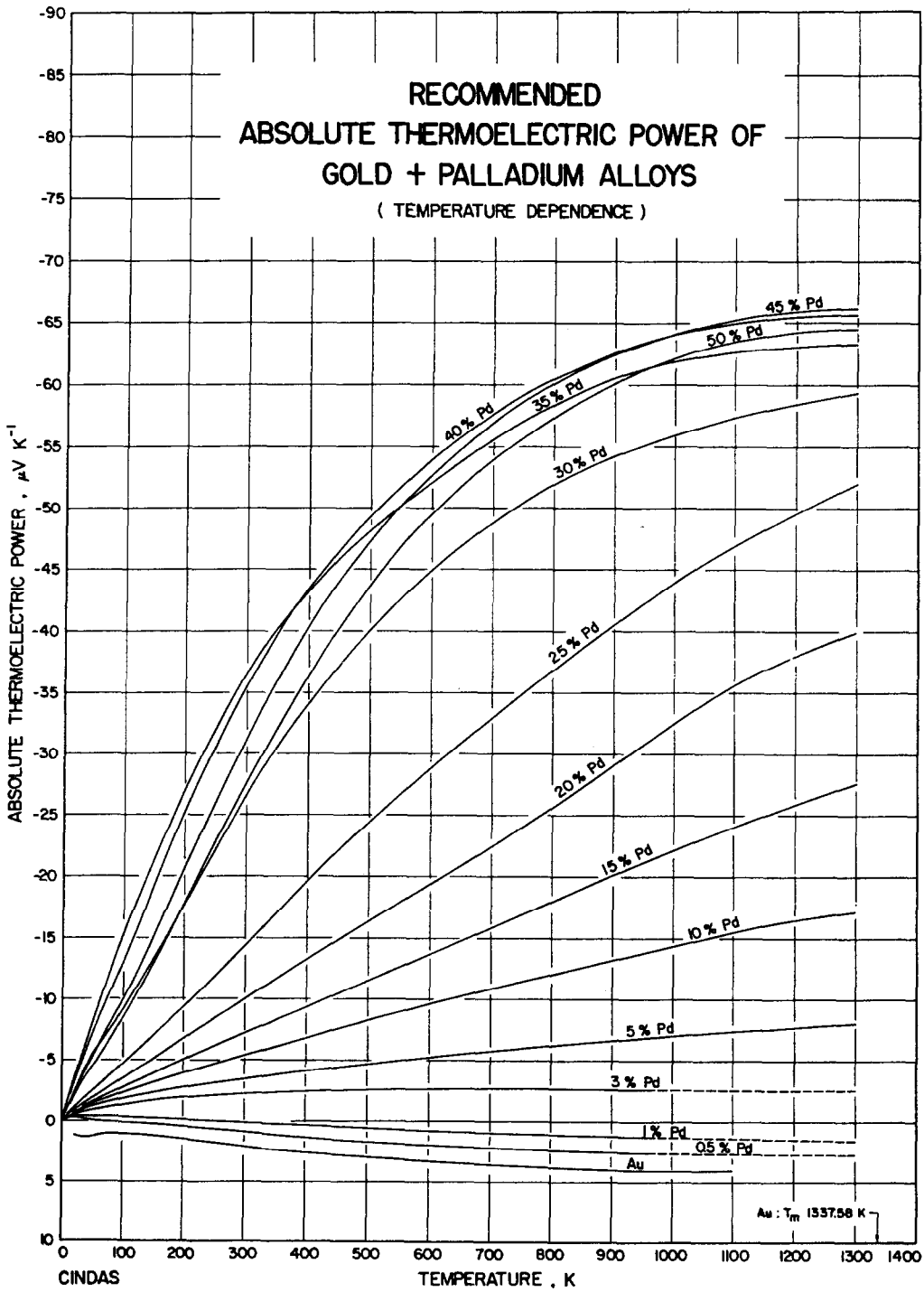



**Fig. 6.** Recommended absolute thermoelectric power of gold + palladium alloys.

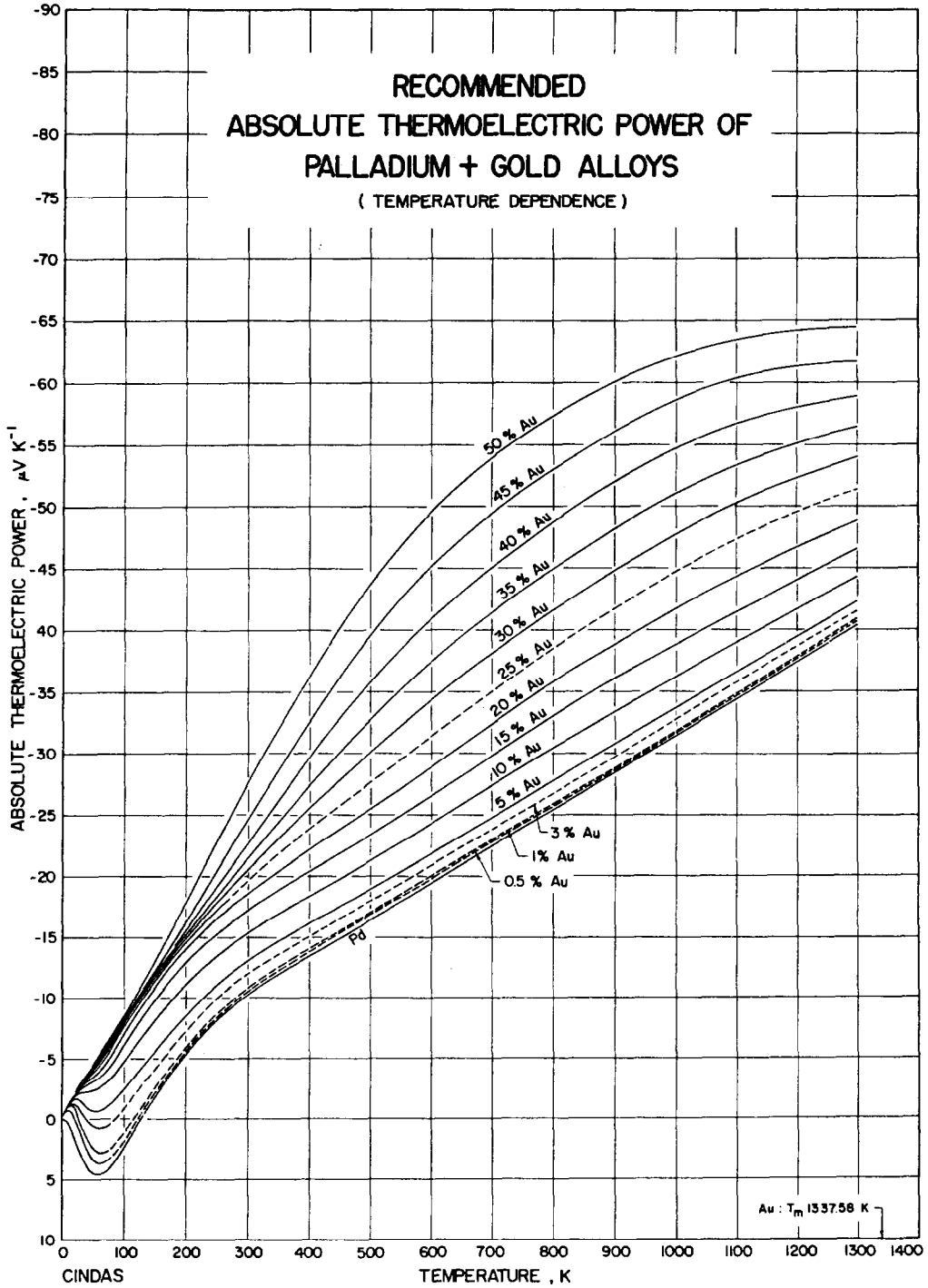
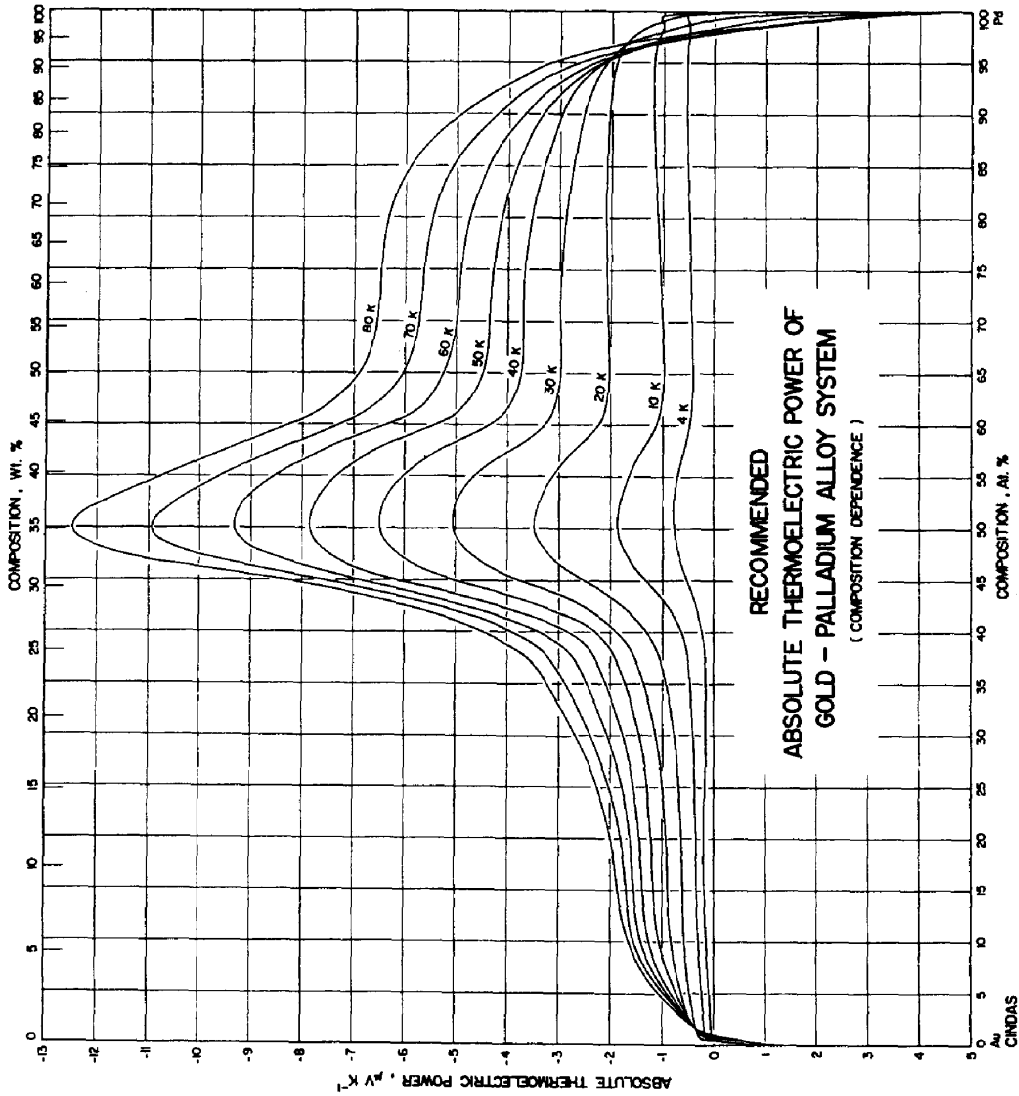
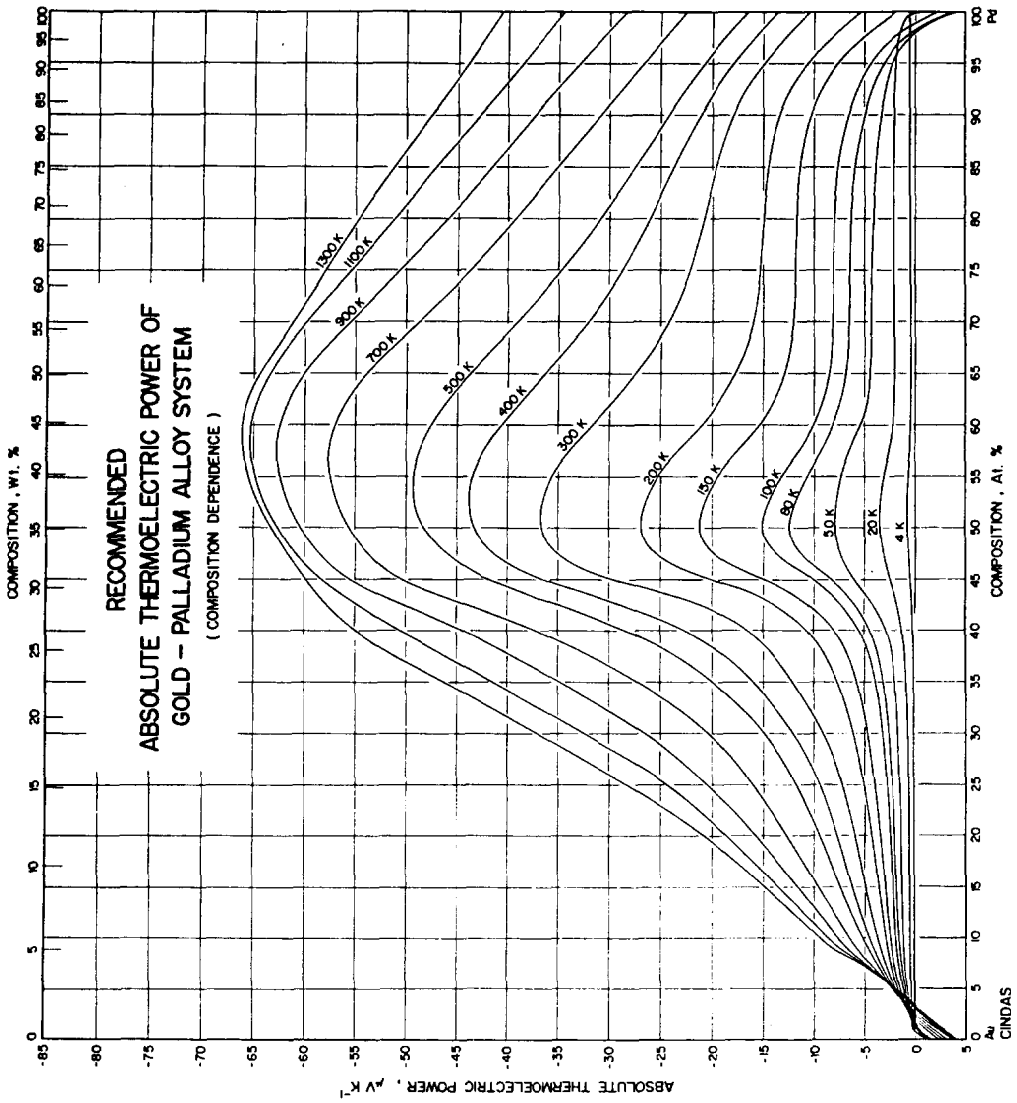



Fig. 7. Recommended absolute thermoelectrical power of palladium + gold alloys.


RECOMMENDED
ABSOLUTE THERMOELECTRIC POWER OF
GOLD – PALLADIUM ALLOY SYSTEM
( COMPOSITION DEPENDENCE )
80 K
70 K
60 K
50 K
40 K
30 K
20 K
10 K
4 K
COMPOSITION , WT. %
COMPOSITION , AT. %
ABSOLUTE THERMOELECTRIC POWER , μV K⁻¹
Au
CINDAS


RECOMMENDED
ABSOLUTE THERMOELECTRIC POWER OF
GOLD – PALLADIUM ALLOY SYSTEM
( COMPOSITION DEPENDENCE )
1300 K
1100 K
900 K
700 K
500 K
400 K
300 K
200 K
150 K
100 K
80 K
50 K
20 K
4 K
COMPOSITION , Wt. %
COMPOSITION , At. %
ABSOLUTE THERMOELECTRIC POWER , μV K⁻¹
Au
Pd
CINDAS

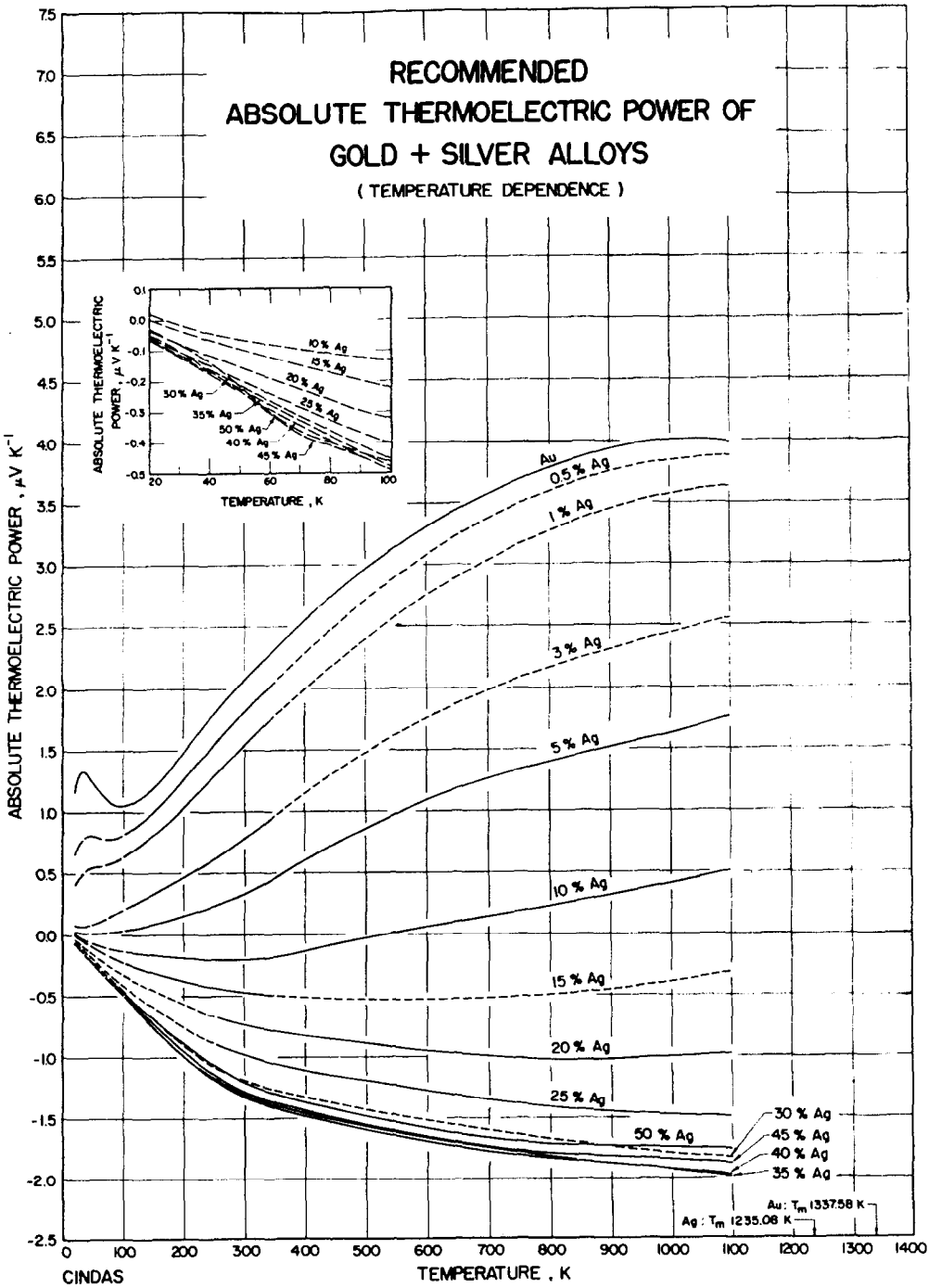



Fig. 10. Recommended absolute thermoelectric power of gold + silver alloys.

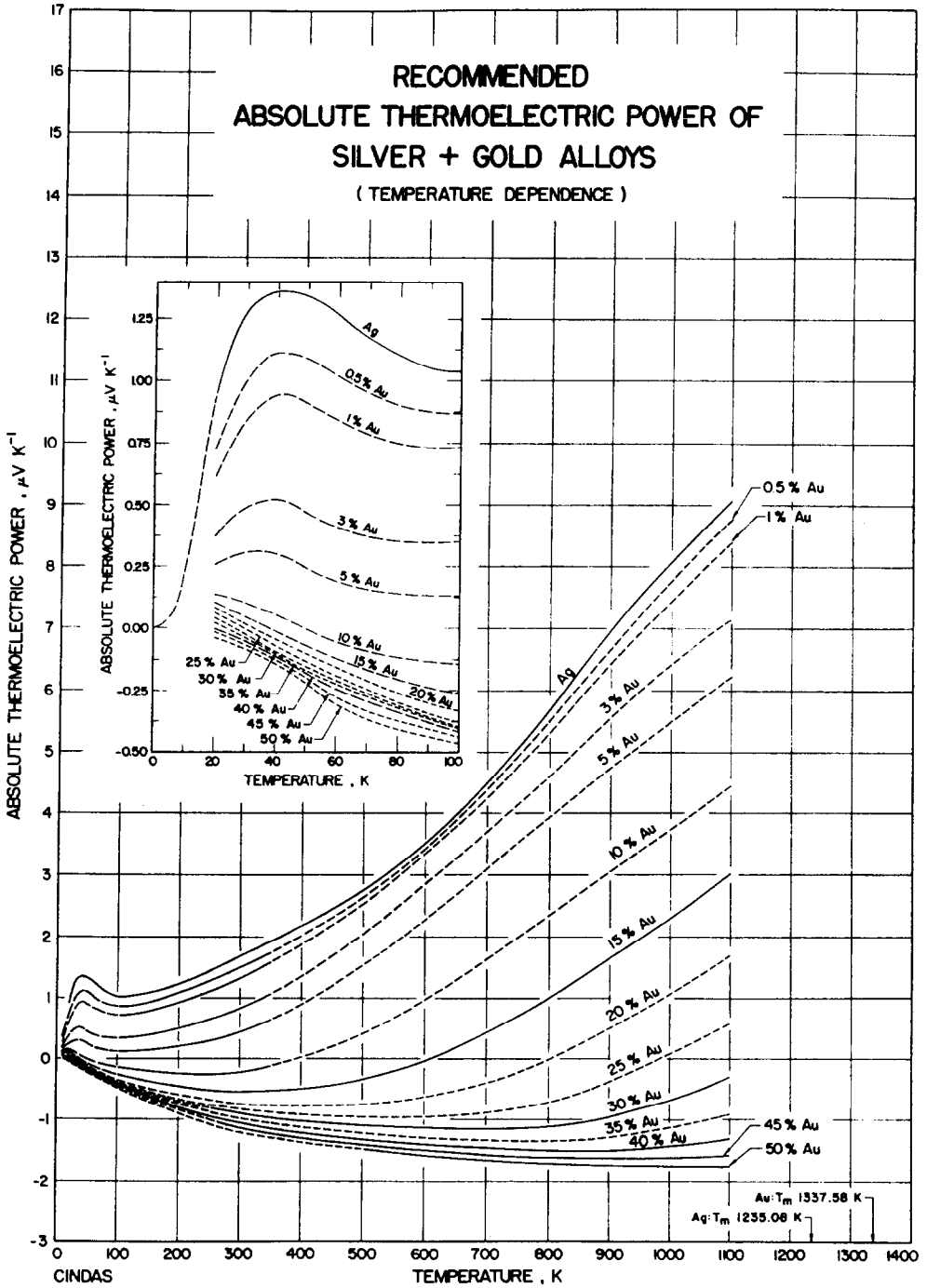
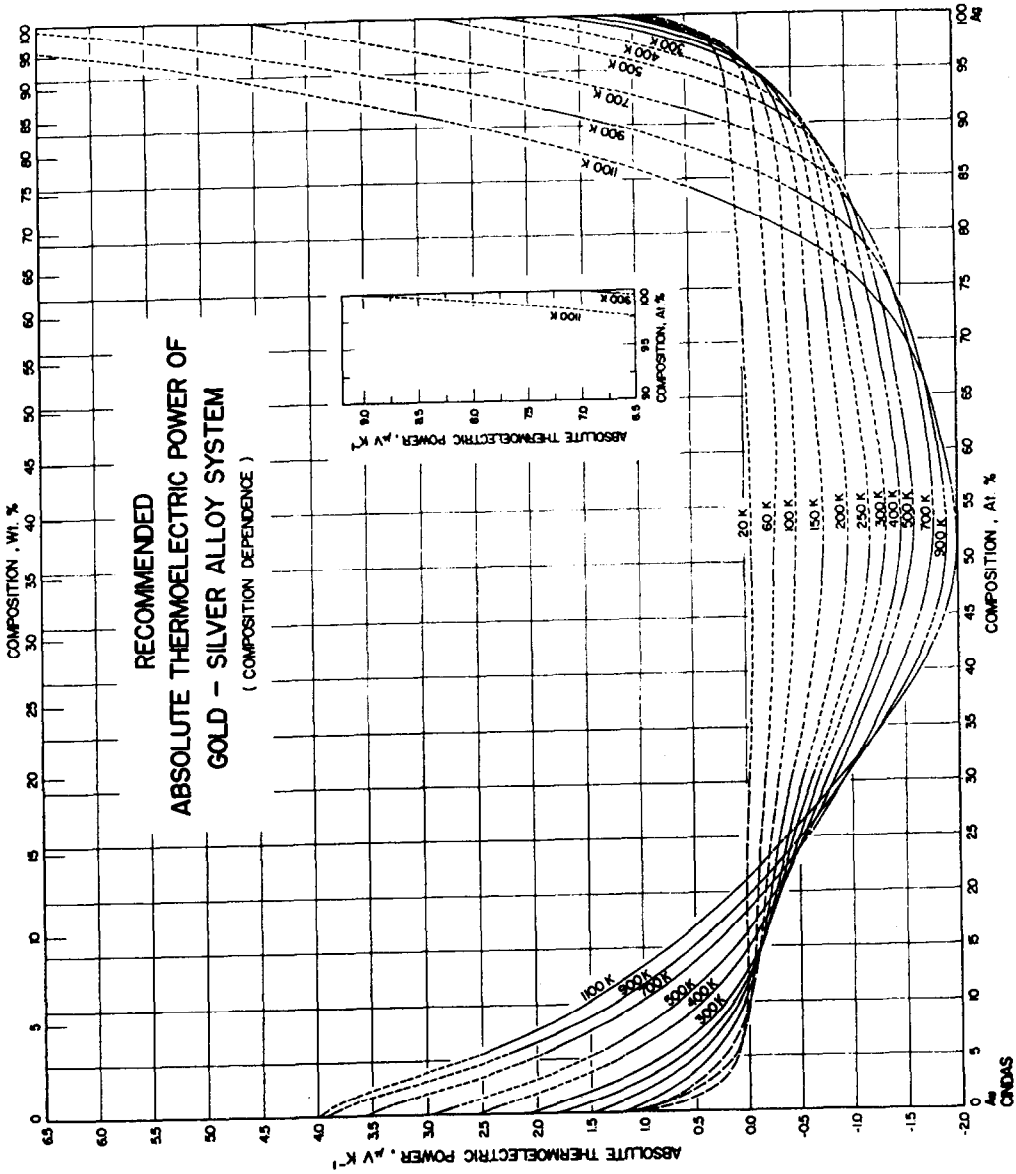



Fig. 11. Recommended absolute thermoelectric power of silver + gold alloys.


RECOMMENDED
ABSOLUTE THERMOELECTRIC POWER OF
GOLD – SILVER ALLOY SYSTEM
( COMPOSITION DEPENDENCE )
COMPOSITION , Wt. %
COMPOSITION , At. %
ABSOLUTE THERMOELECTRIC POWER , μV K⁻¹
20 K
60 K
100 K
150 K
200 K
250 K
300 K
400 K
500 K
700 K
900 K
1100 K
Au
CINDAS

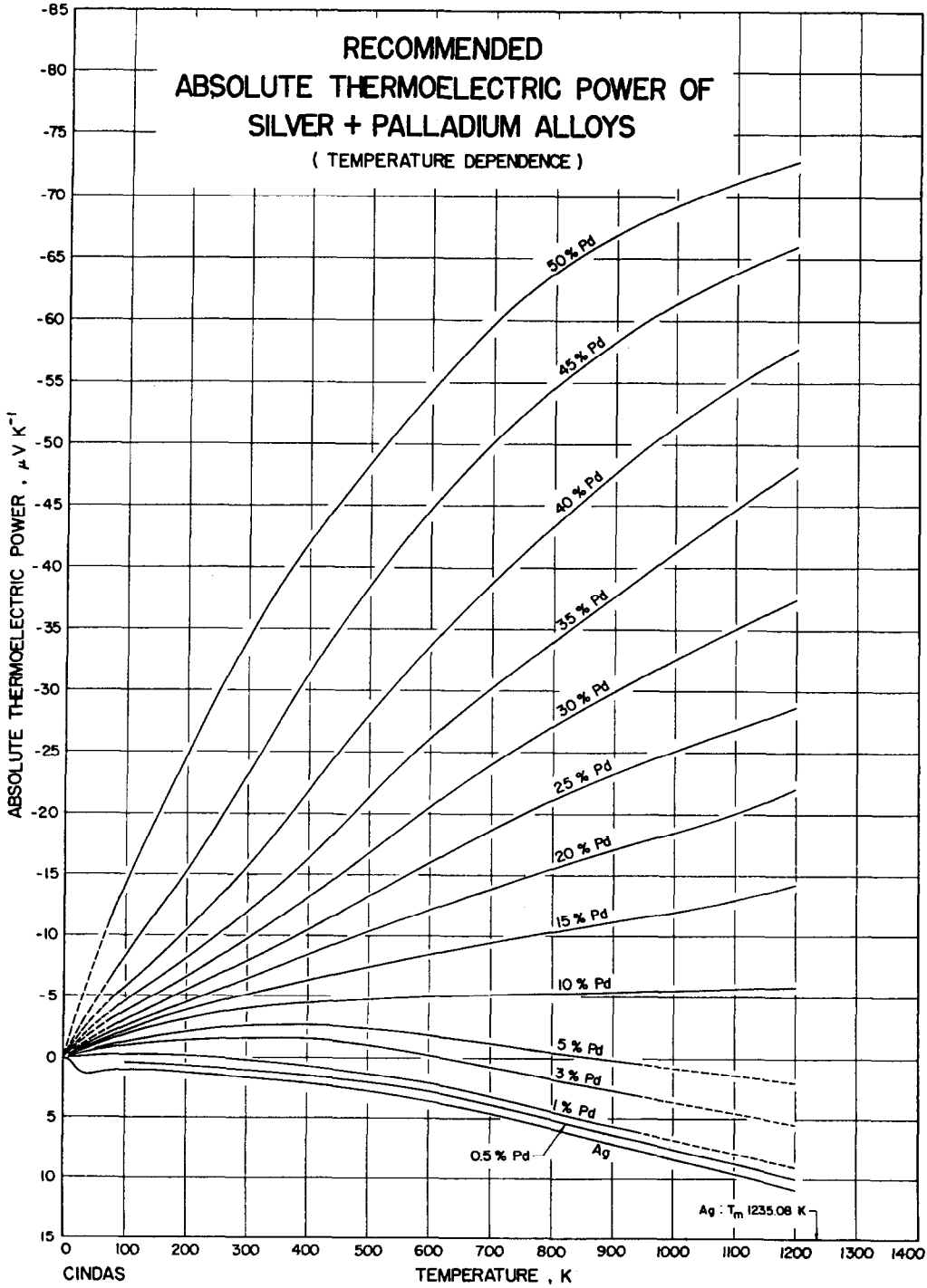



Fig. 13. Recommended absolute thermoelectric power of silver + palladium alloys.

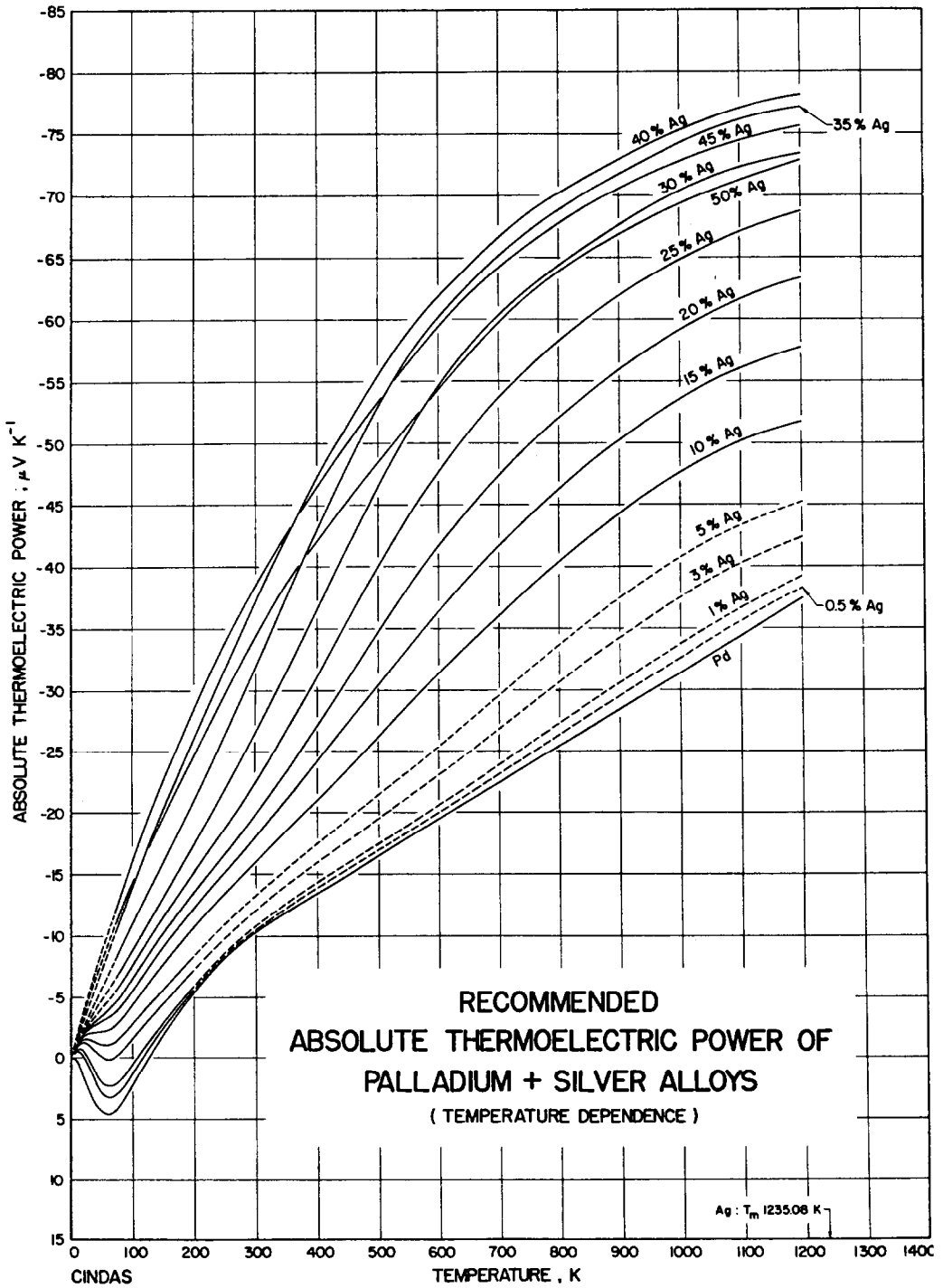



Fig. 14. Recommended absolute thermoelectric power of palladium + silver alloys.

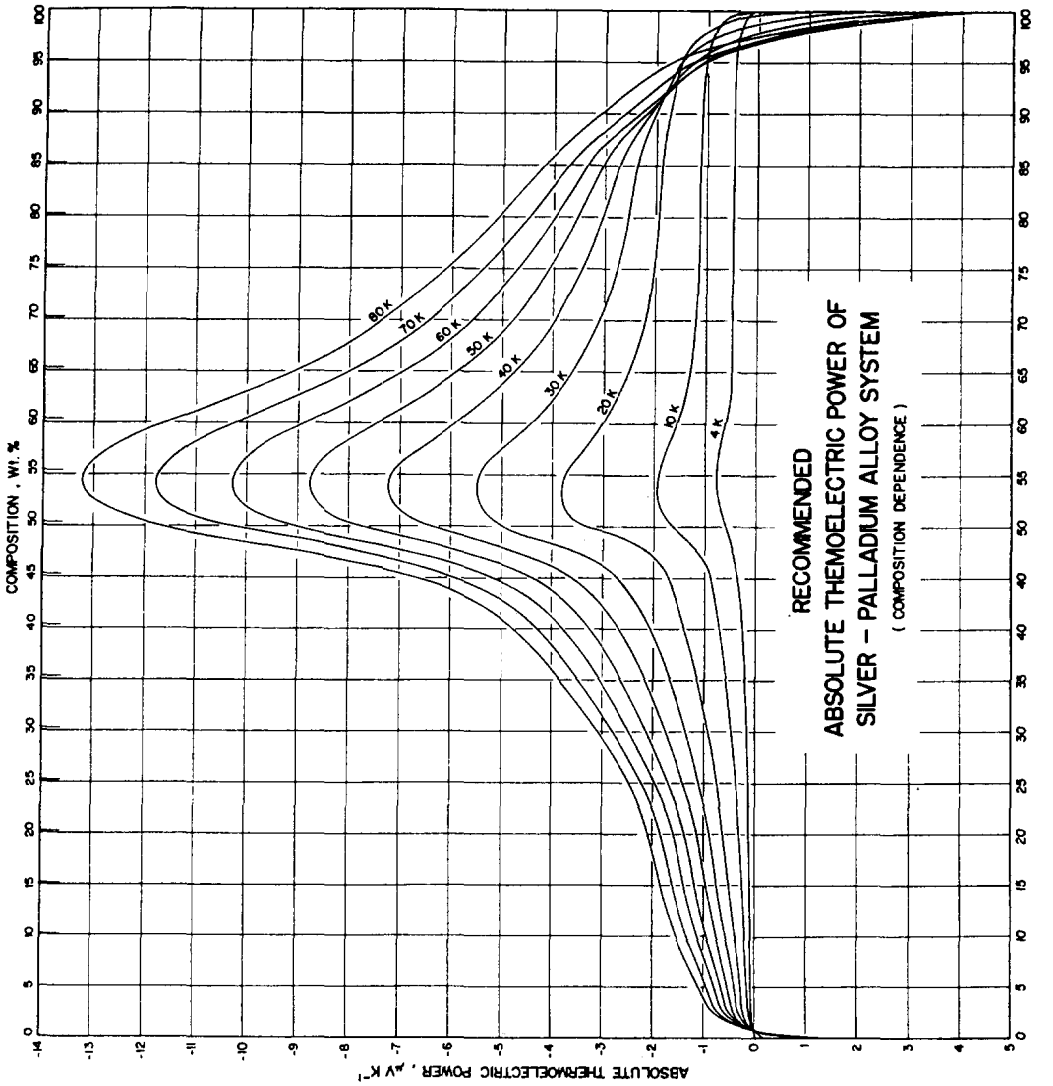



Fig. 15. Recommended absolute thermoelectric power of silver-palladium alloy system.

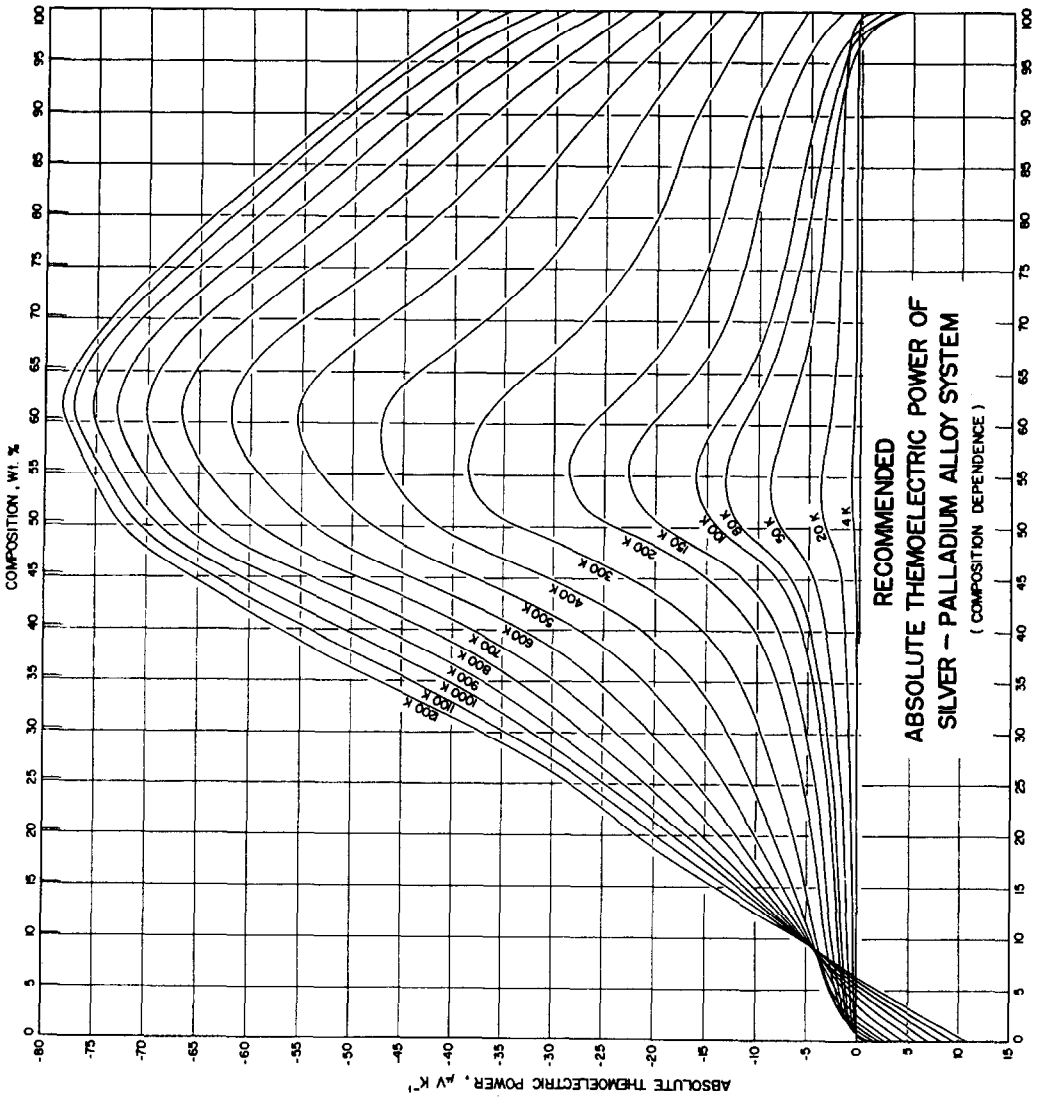



Fig. 16. Recommended absolute thermoelectric power of silver-palladium alloy system

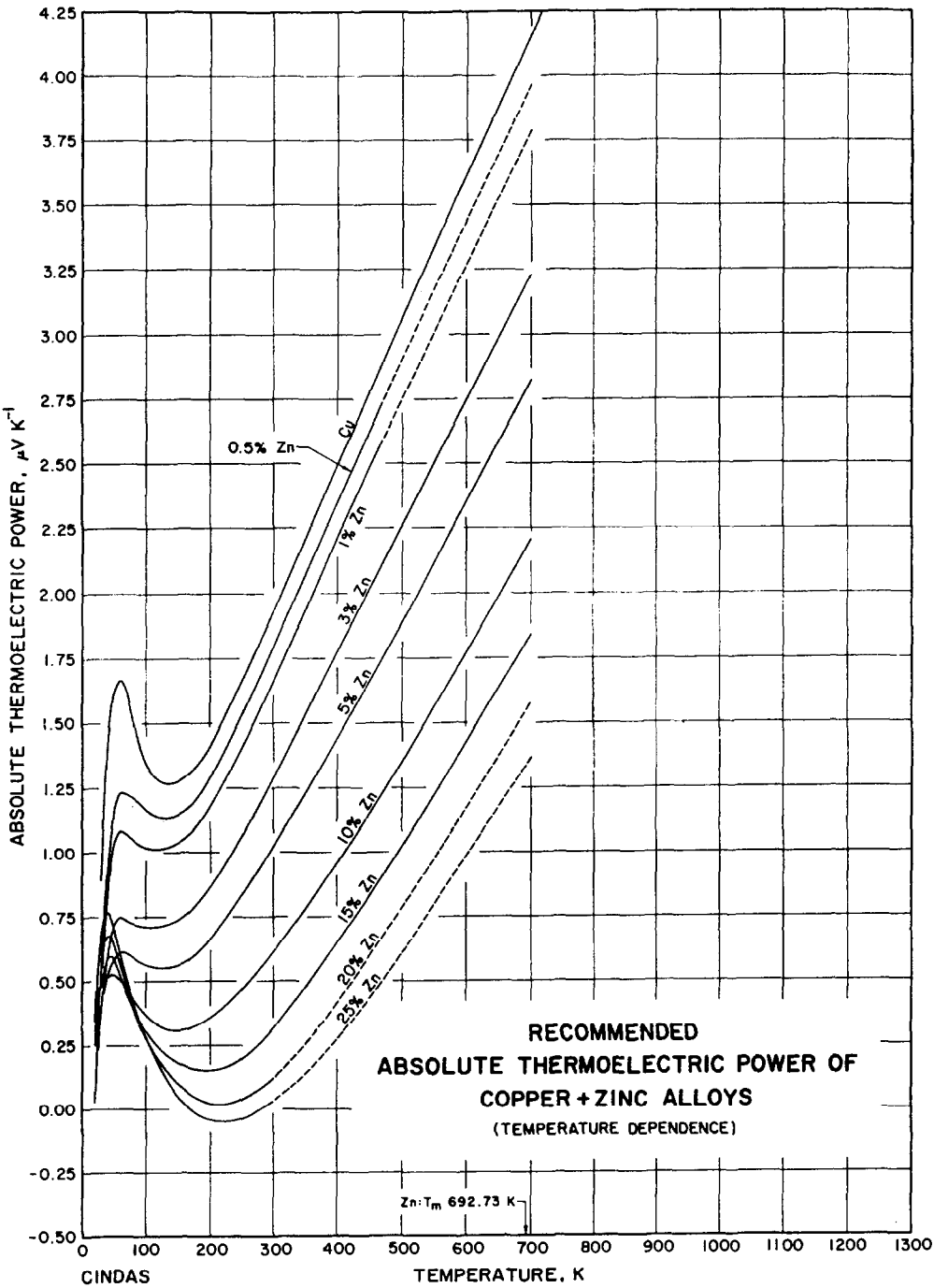



Fig. 17. Recommended absolute thermoelectric power of copper + zinc alloys.

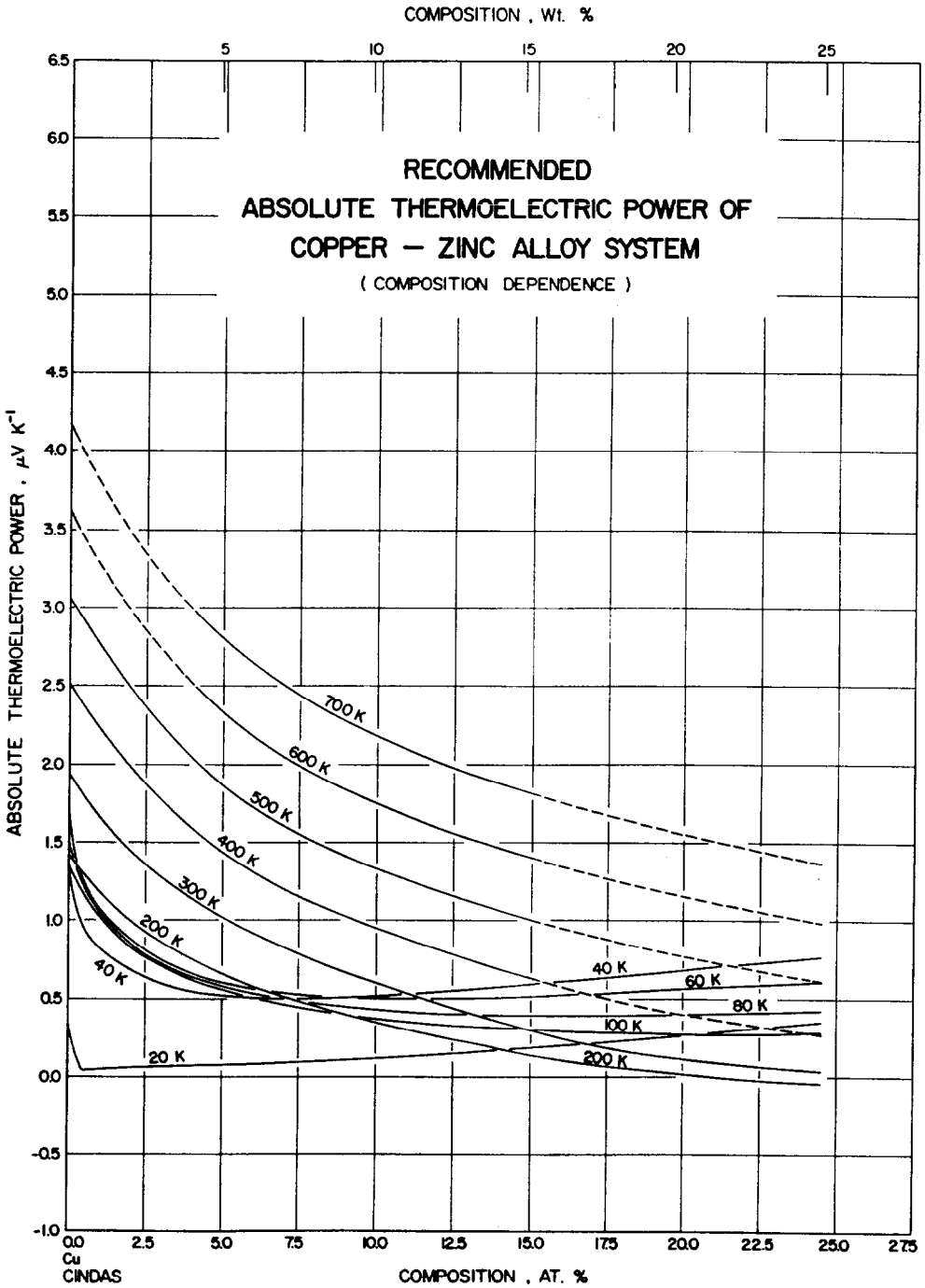



Fig. 18. Recommended absolute thermoelectric power of copper-zinc alloy system.

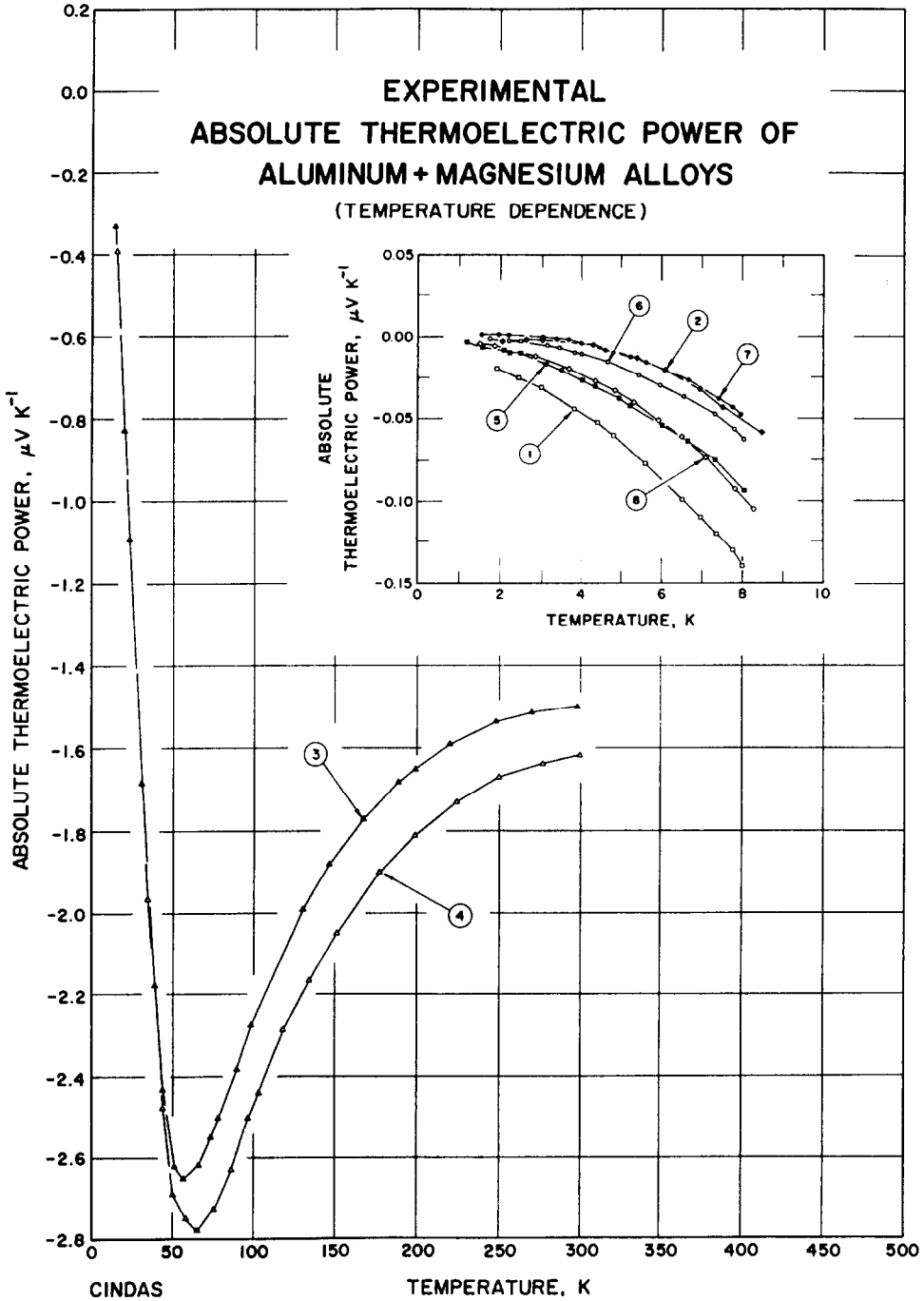



Fig. 19. Experimental absolute thermoelectric power of aluminum + magnesium alloys.

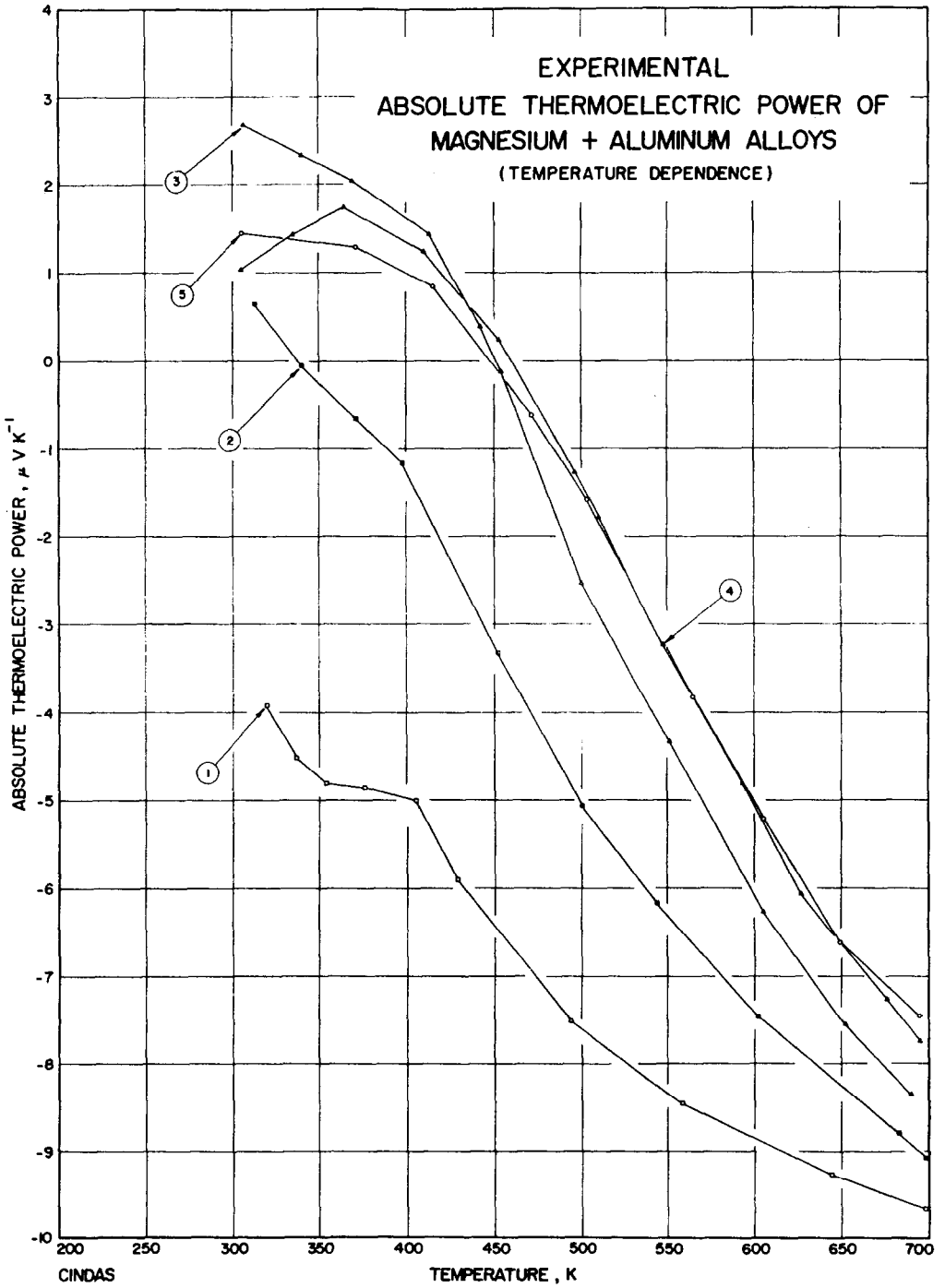



Fig. 20. Experimental absolute thermoelectric power of magnesium + aluminum alloys.

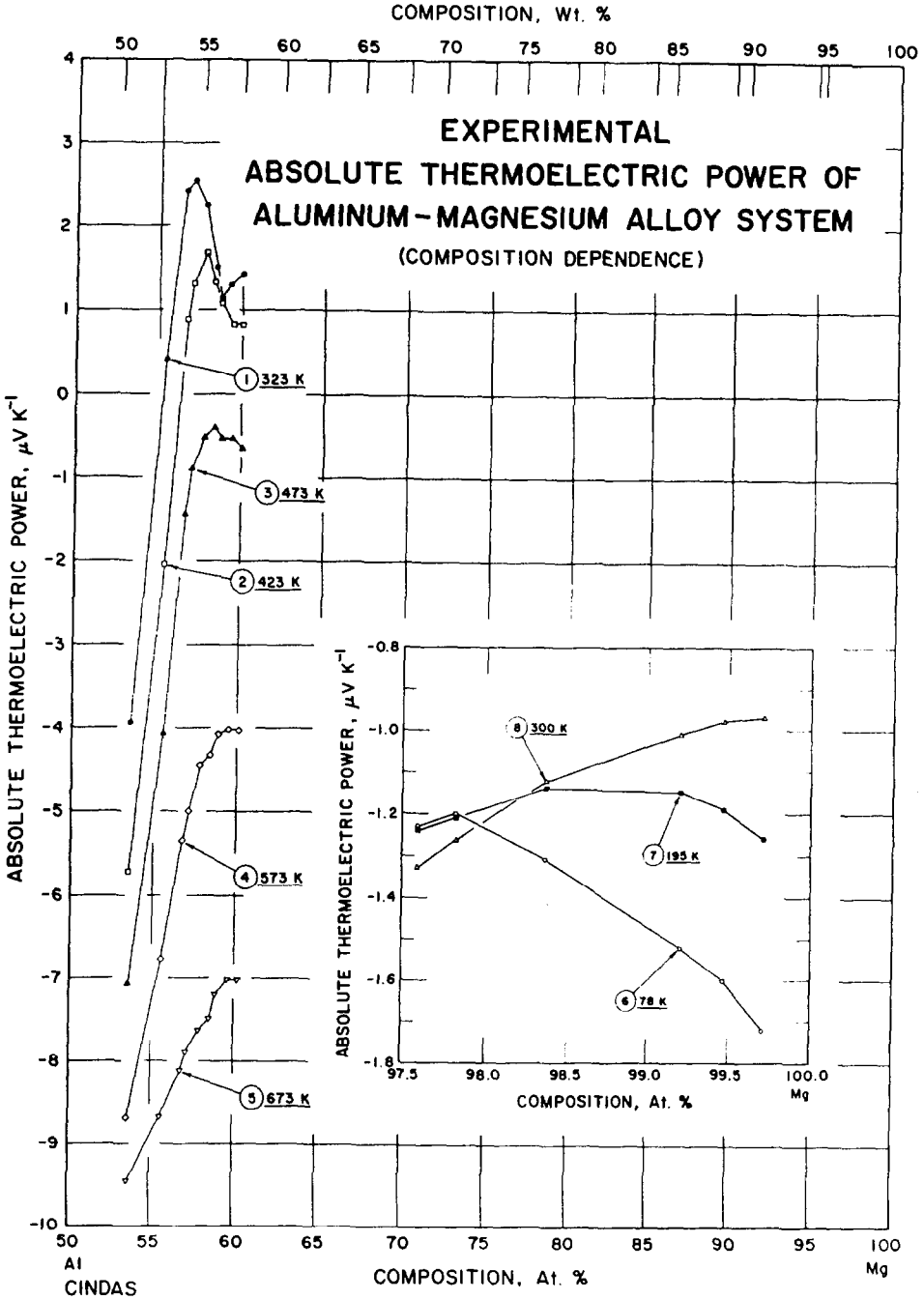



Fig. 21. Experimental absolute thermoelectric power of aluminum-magnesium alloy system.

*Copper-zinc alloy system*

The copper-zinc alloy system has a very complex phase diagram. Experimental data on the thermoelectric power are very scattered except in the $\alpha$-brass and $\gamma$-brass regions.

The recommended values for the absolute thermoelectric power of copper-zinc alloy system were generated only up to 25 wt.% Zn and up to 700 K as presented in Figure 17 for temperature dependence and Figure 18 for composition dependence, though the recommended values for the absolute thermoelectric power of pure zinc have been generated for the temperature range from 2 to 693 K for solid zinc and from 693 to 1000 K for molten zinc.

*Aluminum-magnesium alloy system*

The aluminum-magnesium alloy system has a complicated phase diagram. However, since there exist only very limited data on the absolute thermoelectric power of these alloys, any reference to phase boundaries is hardly required.

Due to the large scatter of the available data for a physically complex system and the lack of adequate theoretical guidance, no recommended values were generated for this alloy system except for pure aluminum from 2 to 931 K and pure magnesium from 2 to 1000 K. Therefore, instead of recommended values, the available experimental data for the absolute thermoelectric power of aluminum + magnesium alloys, magnesium + aluminum alloys, and the aluminum-magnesium alloy system are presented in Figures 19, 20, and 21, respectively.

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