SUPPLEMENTARY INFORMATION

The effect of alloying with scandium in Al-containing High-Entropy Alloys

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Table S1. Rationale of the alloys presented in this work, with the corresponding characterization. The acronyms RT, HT and LT refer to synchrotron experiments performed at room temperature, high-temperature and low-temperature respectively. The notation Thermoelectric properties includes the measurement of Seebeck coefficient, electrical conductivity and thermal diffusivity.

Alloy	Form	Performed analysis
$Al_2CoCrFeNi$	As cast pellet	SEM-EDX, Density, Vickers hardness, DSC, Disc punch test
	Annealed pellet (850 °C, 12h)	SEM-EDX, RT PXRD
	Powder from as-cast pellet	RT PXRD, HT PXRD, LT PXRD
$Al_2CoCrFeNi$ + 0.3wt.% Sc	As-cast pellet	SEM-EDX, RT PXRD, Thermoelectric properties
$Al_2CoCrFeNi$ + 0.5wt.% Sc	As-cast pellet	SEM-EDX, RT PXRD, Disc punch test
$Al_2CoCrFeNi$ + 2wt.% Sc	As-cast pellet	SEM-EDX, RT PXRD, Disc punch test
$Al_2CoCrFeNi$ + 3wt.% Sc	As-cast pellet	SEM-EDX, Density, RT PXRD, Vickers hardness, DSC
	Annealed pellet (900 °C, 12h)	SEM-EDX, RT PXRD
	Powder from as-cast pellet	RT PXRD, HT PXRD, LT PXRD
$Al_2CoCrFeNi$ + 5wt.% Sc	As-cast pellet	SEM-EDX, RT PXRD, Thermoelectric properties
$Al_{0.5}CoCrCuFeNi$	As-cast pellet	SEM-EDX, Vickers hardness, DSC
	Annealed pellet (850 °C. 12h)	SEM-EDX
	Powder from as-cast pellet	RT PXRD, HT PXRD
$Al_{0.5}CoCrCuFeNi$ + 0.5wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness
$Al_{0.5}CoCrCuFeNi$ + 2wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness
$Al_{0.5}CoCrCuFeNi$ + 3wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness, DSC
	Annealed pellet (930 °C, 6h)	SEM-EDX
	Powder from as-cast pellet	RT PXRD, HT PXRD
$AlCoCrCu_{0.5}FeNi$	As-cast pellet	SEM-EDX, Vickers hardness, DSC, Disc punch test
	Annealed pellet (850 °C, 12h)	SEM-EDX
	Powder from as-cast pellet	RT PXRD
$AlCoCrCu_{0.5}FeNi$ + 0.5wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness
$AlCoCrCu_{0.5}FeNi$ + 2wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness

$AlCoCrCu_{0.5}FeNi$ + 3wt.% Sc	As-cast pellet	SEM-EDX, Vickers hardness, DSC, Disc punch test
	Annealed pellet (930 °C, 6h)	SEM-EDX
	Powder from as-cast pellet	RT PXRD

Atomic composition and element distribution according to EDX maps.

Table S2. Atomic composition of the synthesized samples according to EDX map (x500, x2000).

	$Al_2CoCrFeNi$ at.%		**$Al_2CoCrFeNi$ + 3wt.% Sc**		**$Al_{0.5}CoCrCuFeNi$ at.%**		**$Al_{0.5}CoCrCuFeNi$ + 3wt.% Sc**		**$AlCoCrCu_{0.5}FeNi$ at.%**		**$AlCoCrCu_{0.5}FeNi$ + 3wt.% Sc**	
	As melted (±0.04)	**Anneal. (±0.04)**	**Grain at.% (±0.05)**	**Inter-grain at.% (±0.03)**	**As melted (±0.03)**	**Anneal. (±0.04)**	**Grain at.% (±0.04)**	**Inter-grain at.% (±0.04)**	**As melted (±0.03)**	**Anneal. (±0.04)**	**Grain at.% (±0.05)**	**Inter-grain at.% (±0.04)**
Al	44.4(3)	39.5(1)	29.2(6)	18.4(2)	13.2(2)	6.6(3)	7.2(6)	2.8(5)	22.8(2)	27.5(7)	10.7(1)	8.8(3)
Co	14.7(2)	15.7(7)	19.6(4)	16.2(8)	17.9(4)	19.8(3)	20.4(0)	3.2(3)	17.2(5)	16.8(0)	21.3(3)	10.6(0)
Cr	13.6(2)	13.8(2)	14.1(8)	11.1(1)	16.6(6)	18.0(6)	17.3(2)	1.6(5)	17.6(0)	16.0(6)	29.9(3)	6.3(2)
Cu	-	-	-	-	16.9(4)	11.1(9)	16.2(7)	75.7(4)	8.6(7)	6.9(7)	6.0(1)	51.8(6)
Fe	14.3(4)	15.1(4)	16.5(1)	17.7(1)	18.5(1)	20.1(4)	20.2(3)	2.4(8)	17.8(4)	16.8(9)	23.2(5)	8.6(1)
Ni	12.8(9)	15.7(7)	19.9(9)	19.4(5)	17.5(2)	17.6(8)	18.2(6)	10.6(5)	15.8(1)	15.7(1)	14.6(3)	13.0(4)
Sc	-	-	0.4(2)	17.0(2)	-	-	0.2(2)	3.4(1)	-	-	0.1(4)	0.7(5)

Element distribution of the as-cast and annealed alloys according to EDX maps

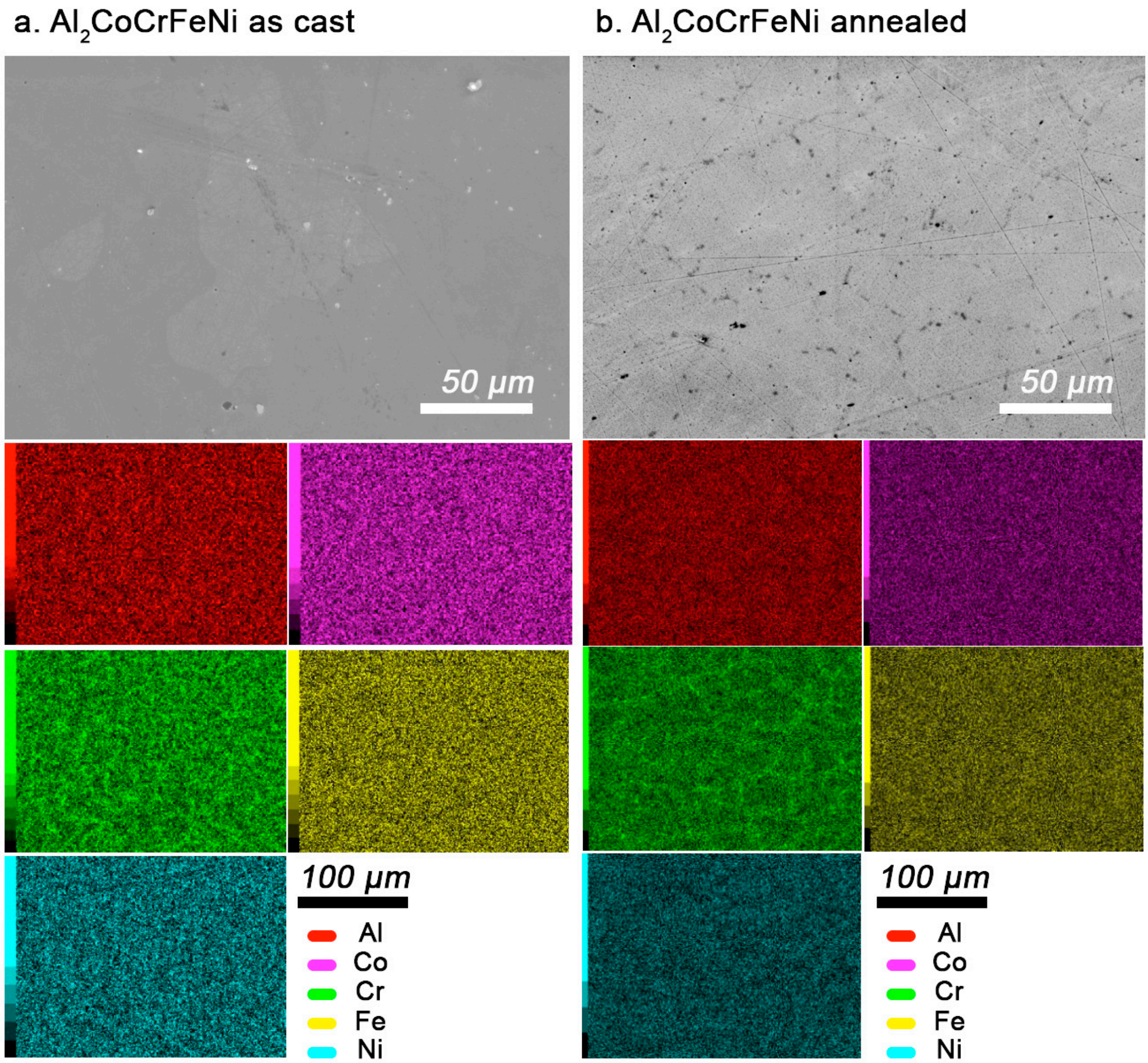

Figure S1. BSE images and element distribution of as cast and annealed $Al_2CoCrFeNi$ samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Fe, Ni.

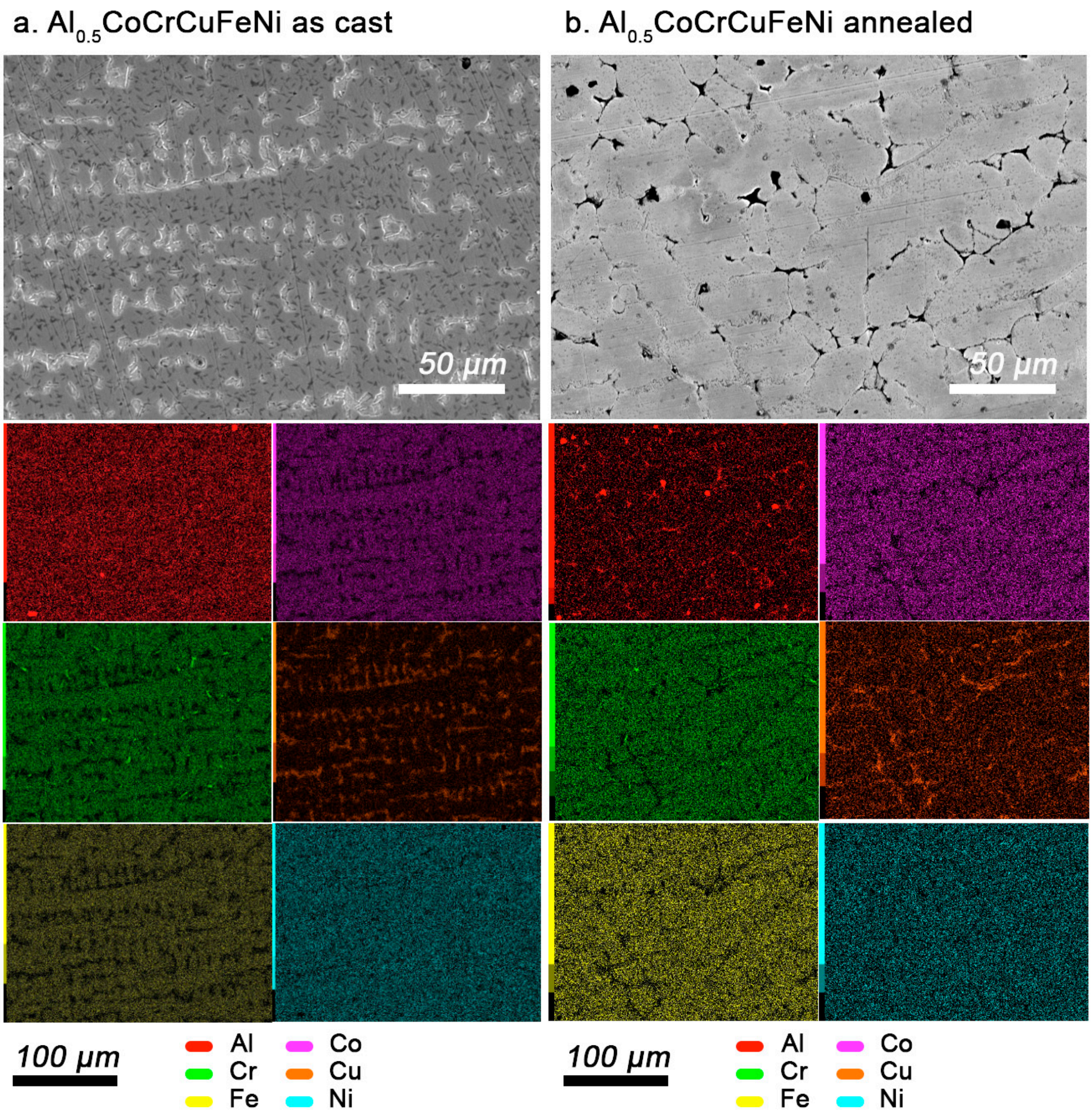

Figure S2. BSE images and element distribution of as cast and annealed $Al_{0.5}CoCrCuFeNi$ samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni.

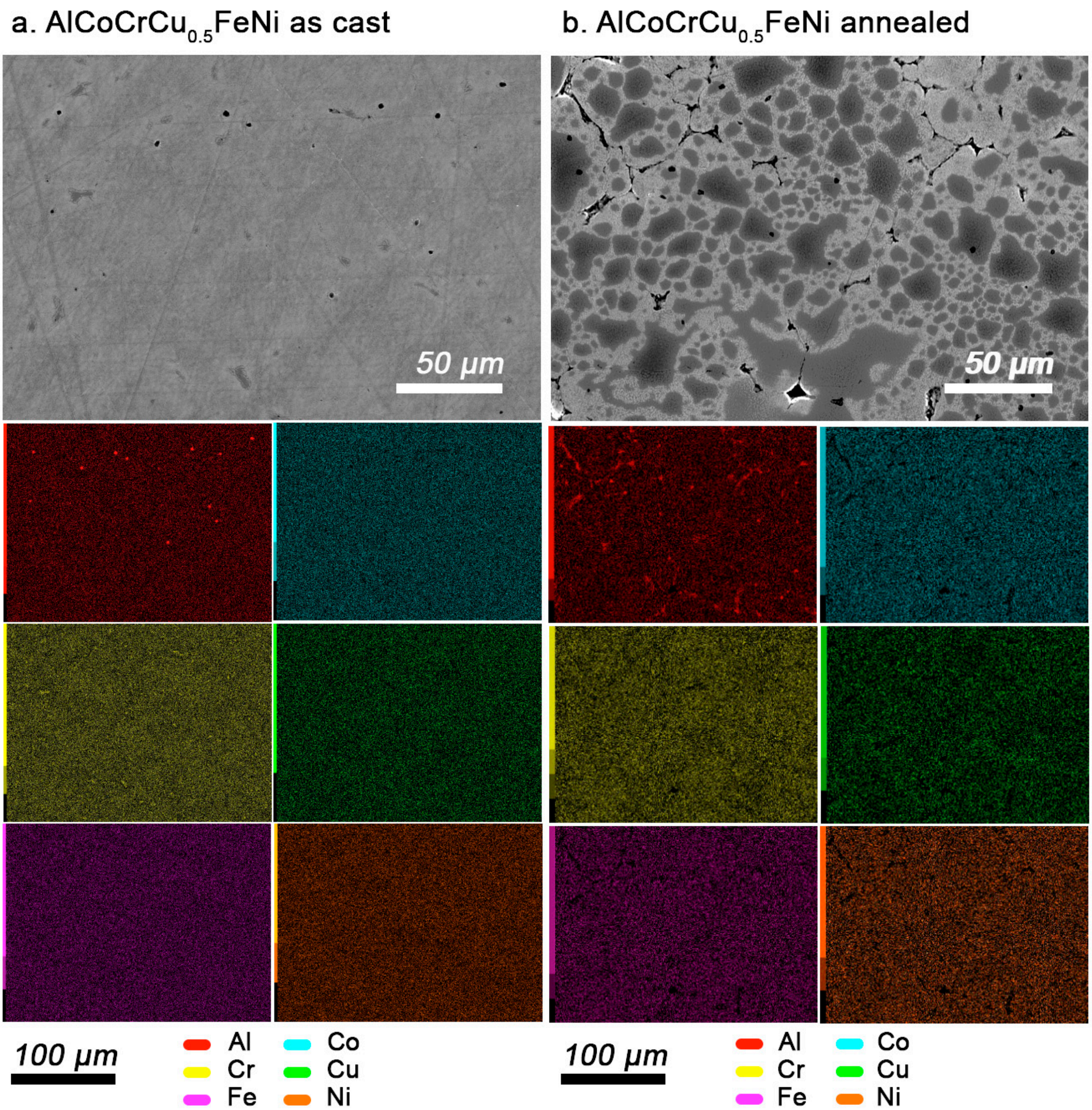

Figure S3. BSE images and element distribution of as cast and annealed $AlCoCrCu_{0.5}FeNi$ samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni.

Element distribution of the as-cast Sc-containing alloys according to EDX maps

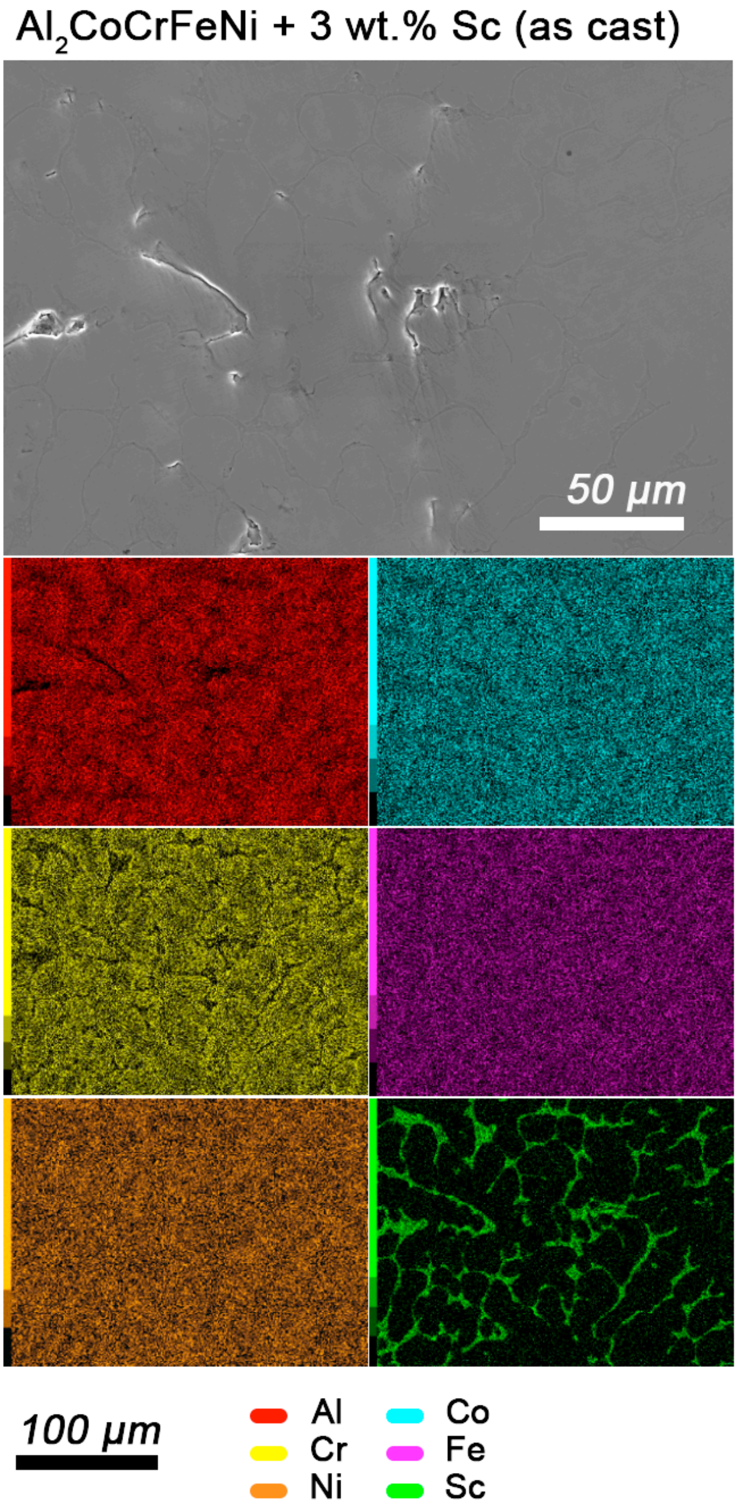

Figure S4. BSE image and element distribution of as cast $Al_2CoCrFeNi$ +3wt.%Sc samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Fe, Ni and Sc.

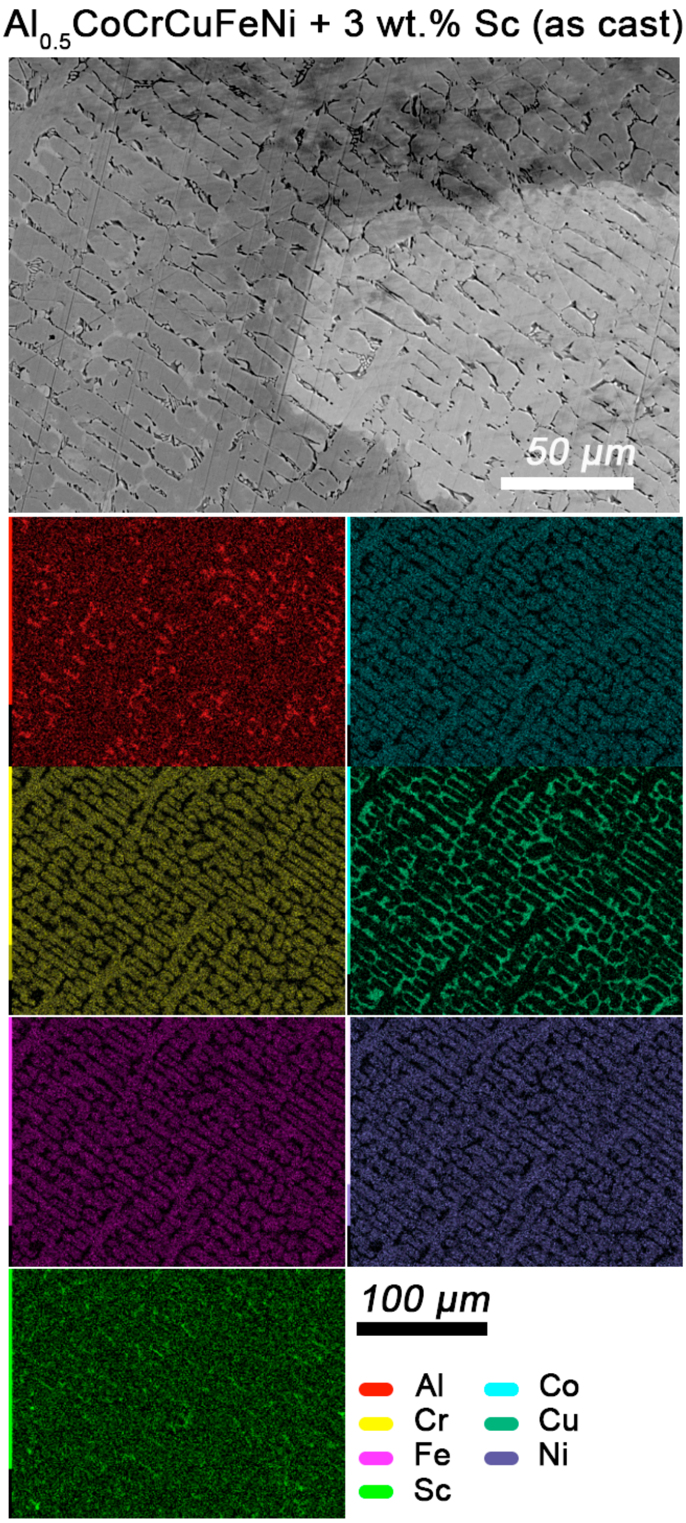

Figure S5. BSE image and element distribution of as cast $Al_{0.5}CoCrCuFeNi$ +3wt.%Sc samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni and Sc.

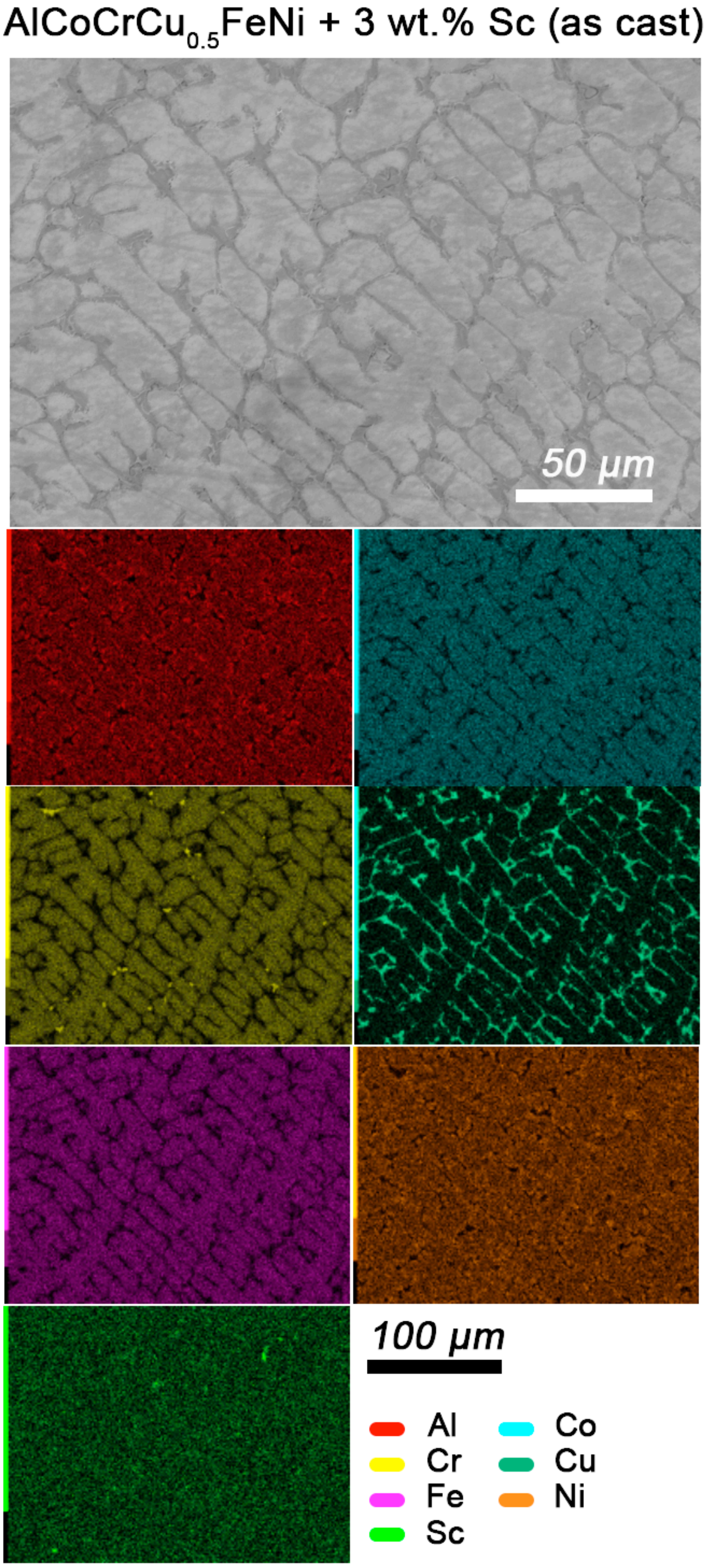

Figure S6. BSE image and element distribution of as cast $AlCoCrCu_{0.5}FeNi$ +3wt.%Sc samples according to EDX. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni and Sc.

Mechanical properties: disk-punch test

Applied force was plotted against disk's displacement. To normalize the disks for the standard 0.5 mm thickness, the following equation(s) were used:

Ultimate tensile strength for brittle materials [MPa = Nmm^{-2}] [1]

$$Cl = A - (D + 2t) = 8 - (4 + 2t)$$

$$UTS = \frac{F_m}{t(0.14D - 0.82Cl + 2.17u_m + 0.6)}$$

Where F_m is the maximum load during PT, t is the disk thickness in mm, D is the punch diameter (4 mm), Cl is the die clearance in mm and u_m is the displacement at failure. In the first equation, A is the diameter of the lower die (8 mm).

Punch test results obtained after normalizing thickness to 0.5mm: [2]

The inflexion point is a constant in the following equations.

For $P_{test} < P_{inflexion}$

$$P_{0.5} = 0.5^2\left(\frac{P_{test}}{t^2}\right)$$

For $P_{test} > P_{inflexion}$

$$P_{0.5} = 0.5\left(\frac{P_{test}}{t}\right) + 0.5\,P_{inflexion}\left(\frac{0.5 - t}{t^2}\right)$$

Ultimate tensile strength [MPa = Nmm^{-2}]

$$UTS = \frac{0.4964\,F_m}{u_m h_o} - 94.146$$

Yield stress

$$YS = \frac{0.4454\,F_e}{h_o^2} + 86.866$$

Where F_m is the maximum load during PT, u_m is the displacement related to the maximum load F_m, h_o is the normalized thickness (0.5mm) and F_e is the load illustrating the conversion between linearity and yield zone (intersection between zone 1 and 2). [3]

Fracture stress for brittle materials

$$\sigma_f = 130\frac{F_m}{t^2} - 320$$

Fracture toughness for brittle materials

$$K_{IC} = 0.07(\,\sigma_f)^{2/3}$$

The units of KIC are MPa, F_m is in N and t is the initial thickness in mm. [4]

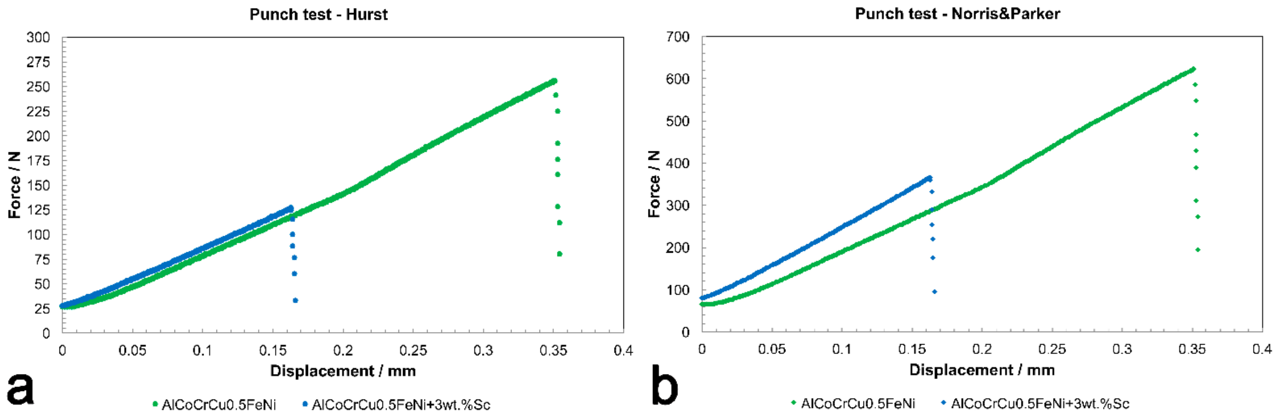

Figure S7. Disk punch test for the $AlCoCrCu_{0.5}FeNi$ HEA with 0 and 3wt.% Sc, data elaborated according to the equations reported by Norris and Parker (*right*) and Hurst (*left*) [1], [3].

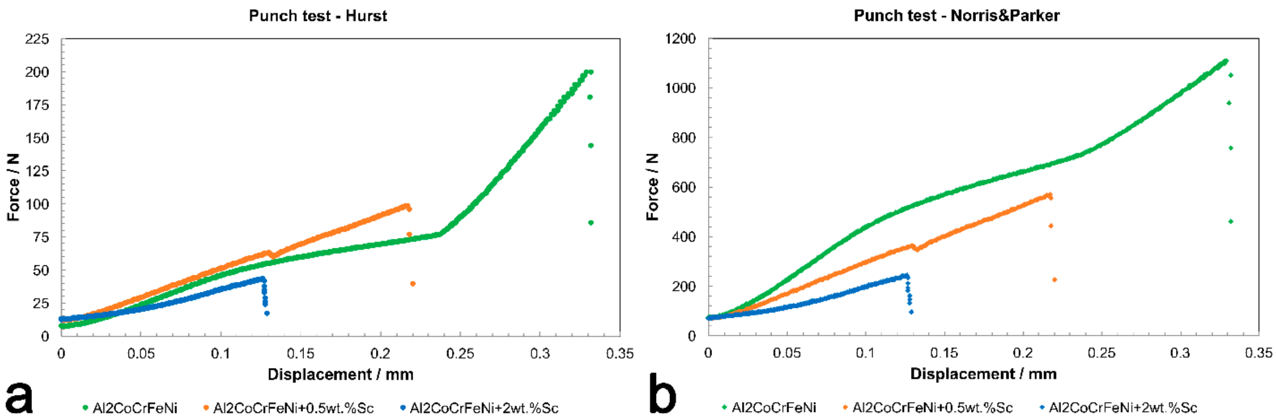

Figure S8. Disk punch test for the $Al_2CoCrFeNi$ alloy with 0, 0.5 and 2wt.% Sc, data elaborated according to the equations reported by Hurst (*left*) and Norris and Parker (*right*) [1], [3].

Element distribution of the annealed alloys according to EDX maps

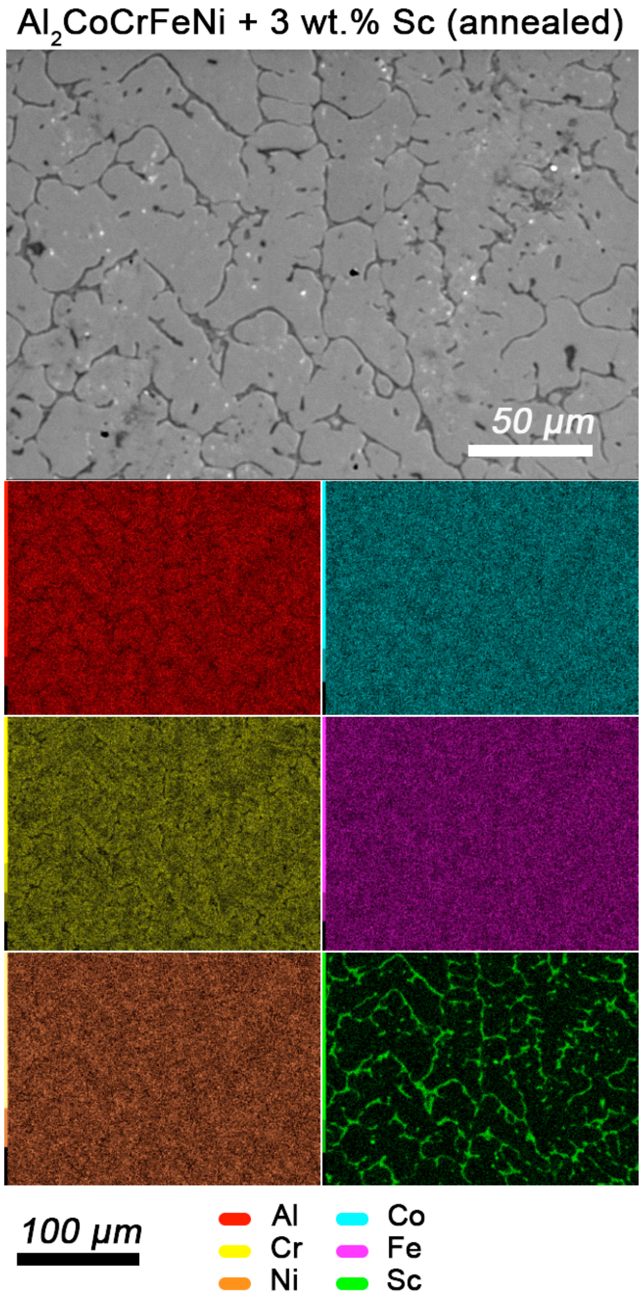

Figure S9. BSE image and element distribution of $Al_2CoCrFeNi$+3wt.%Sc samples after annealing at 900°C, 12h. Data from EDX maps. From top to bottom and left to right, elements are: Al, Co, Cr, Fe, Ni and Sc.

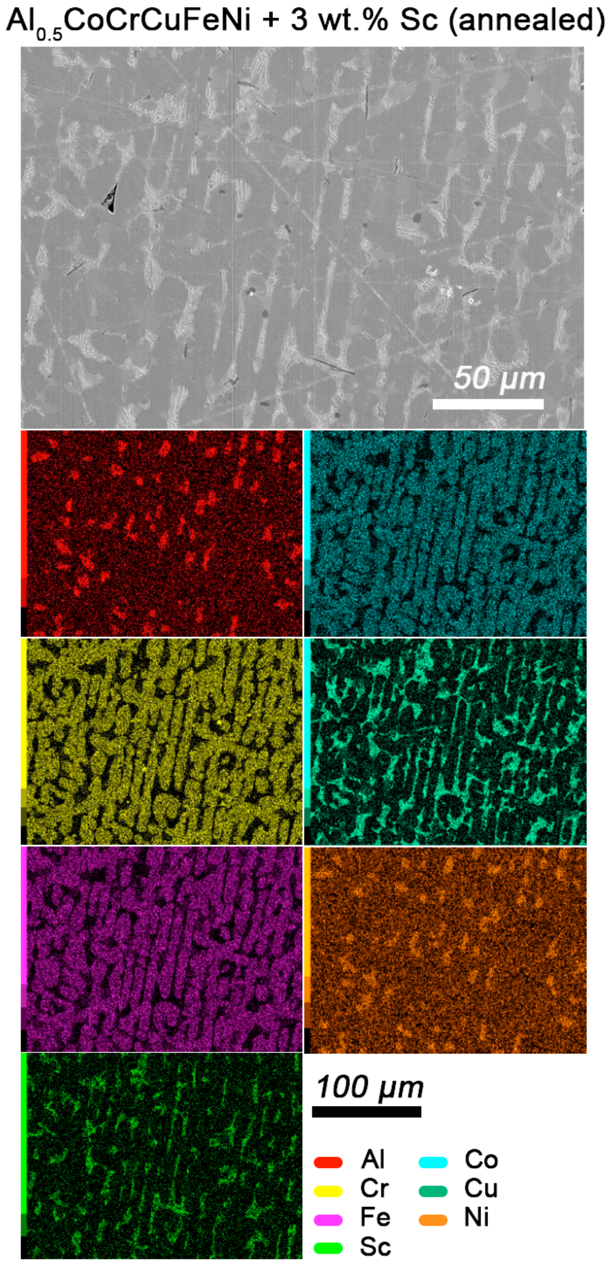

Figure S10. BSE image and element distribution in $Al_{0.5}CoCrCuFeNi$ +3wt.%Sc samples after annealing at 930°C, 6h, respectively. Data from EDX maps. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni and Sc.

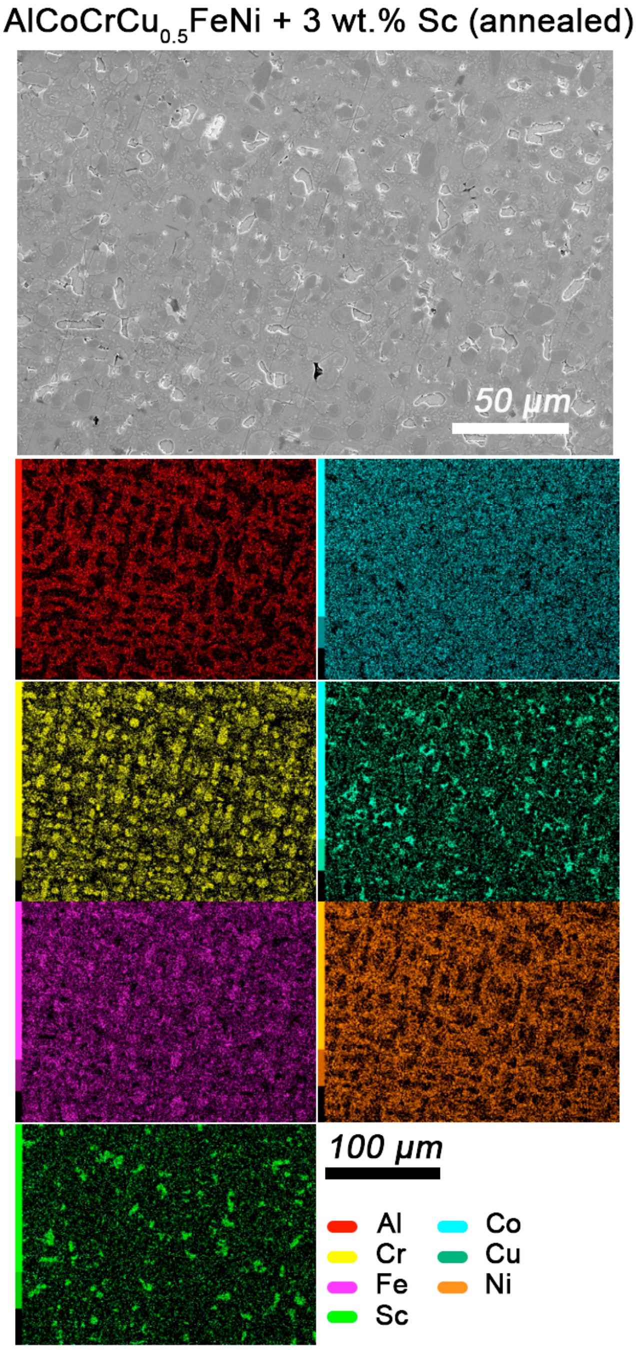

Figure S11. BSE image and element distribution of $AlCoCrCu_{0.5}FeNi$ +3wt.%Sc samples after annealing at 930°C, 6h, respectively. Data from EDX maps. From top to bottom and left to right, elements are: Al, Co, Cr, Cu, Fe, Ni and Sc.

Electrical and thermal transport measurements

Table S3. Density and Dulong-Petit heat capacity used for the calculation of thermal conductivity.

Composition	Density (in g cm^{-3})	C_p (Dulong-Petit, in J g^{-1} K^{-1})
$Al_2CoCrFeNi$ + 5 w% Sc	5.99	0.53651
$Al_2CoCrFeNi$ + 0.3 w% Sc	6.35	0.53565

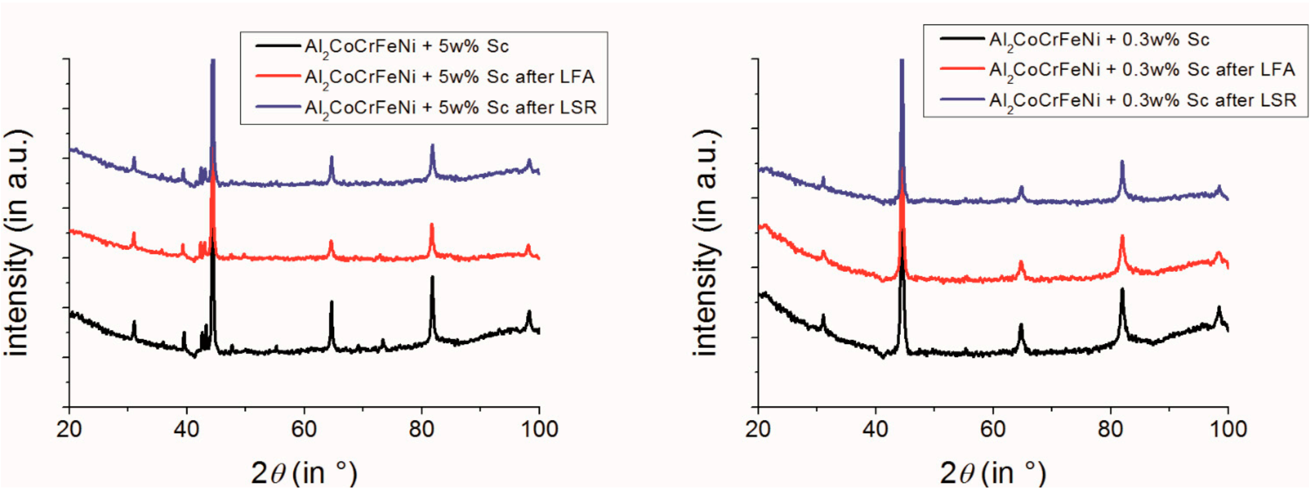

Figure S12. PXRD data of Al2CoCrFeNi + 5 w% Sc (left) and Al2CoCrFeNi + 0.3 w% Sc (right) before thermoelectric measurements (black) and after LFA (red) and LSR (blue), respectively.

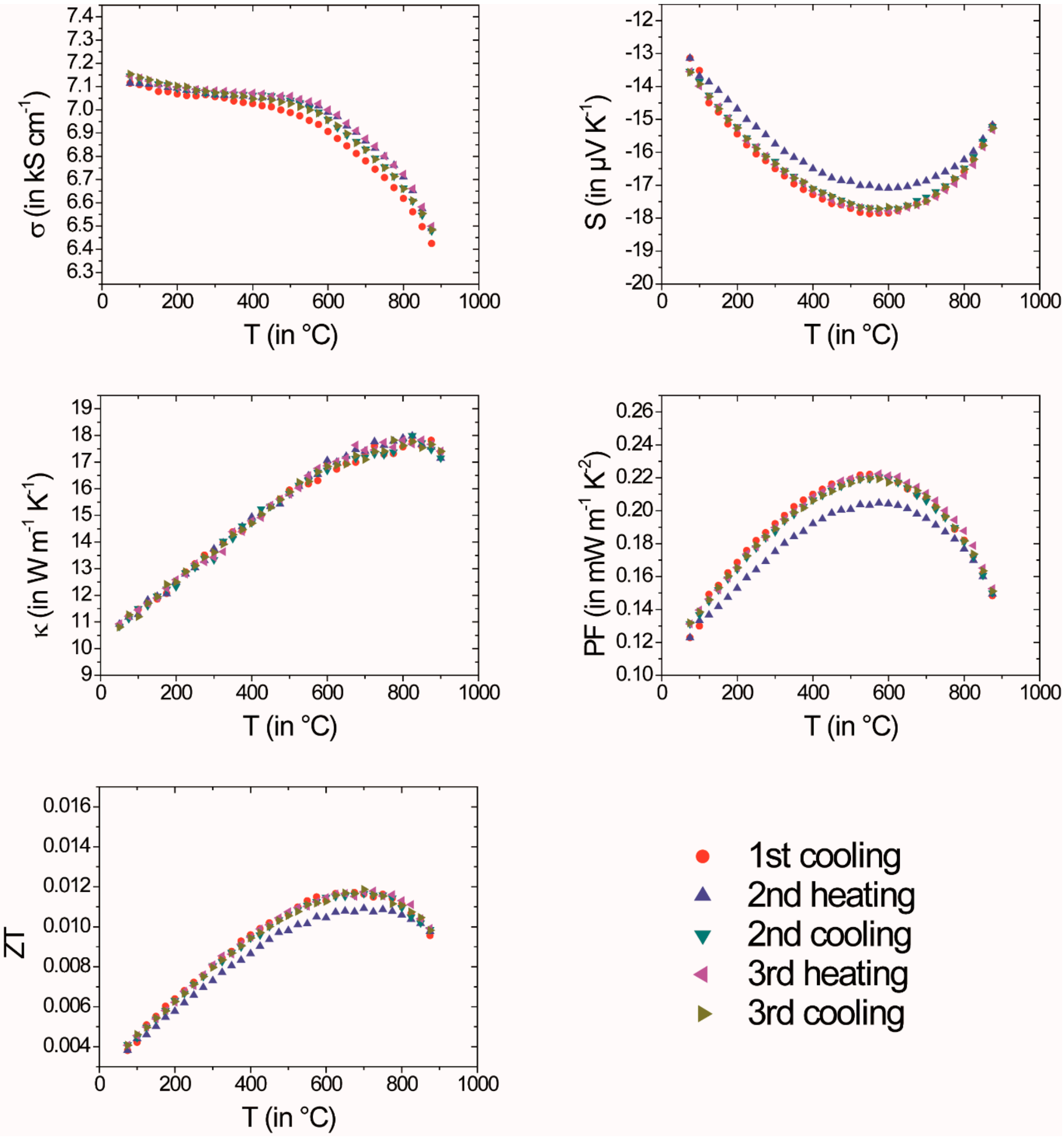

Figure S13. Thermoelectric properties of $Al_2CrCoFeNi$ + 5 w% Sc for three consecutive cycles (without first heating) up to 875 °C: electrical conductivity σ (top, left), Seebeck coefficient S (top, right), thermal conductivity κ (middle, left), power factor PF (middle, right) and thermoelectric figure of merit ZT (bottom, left).

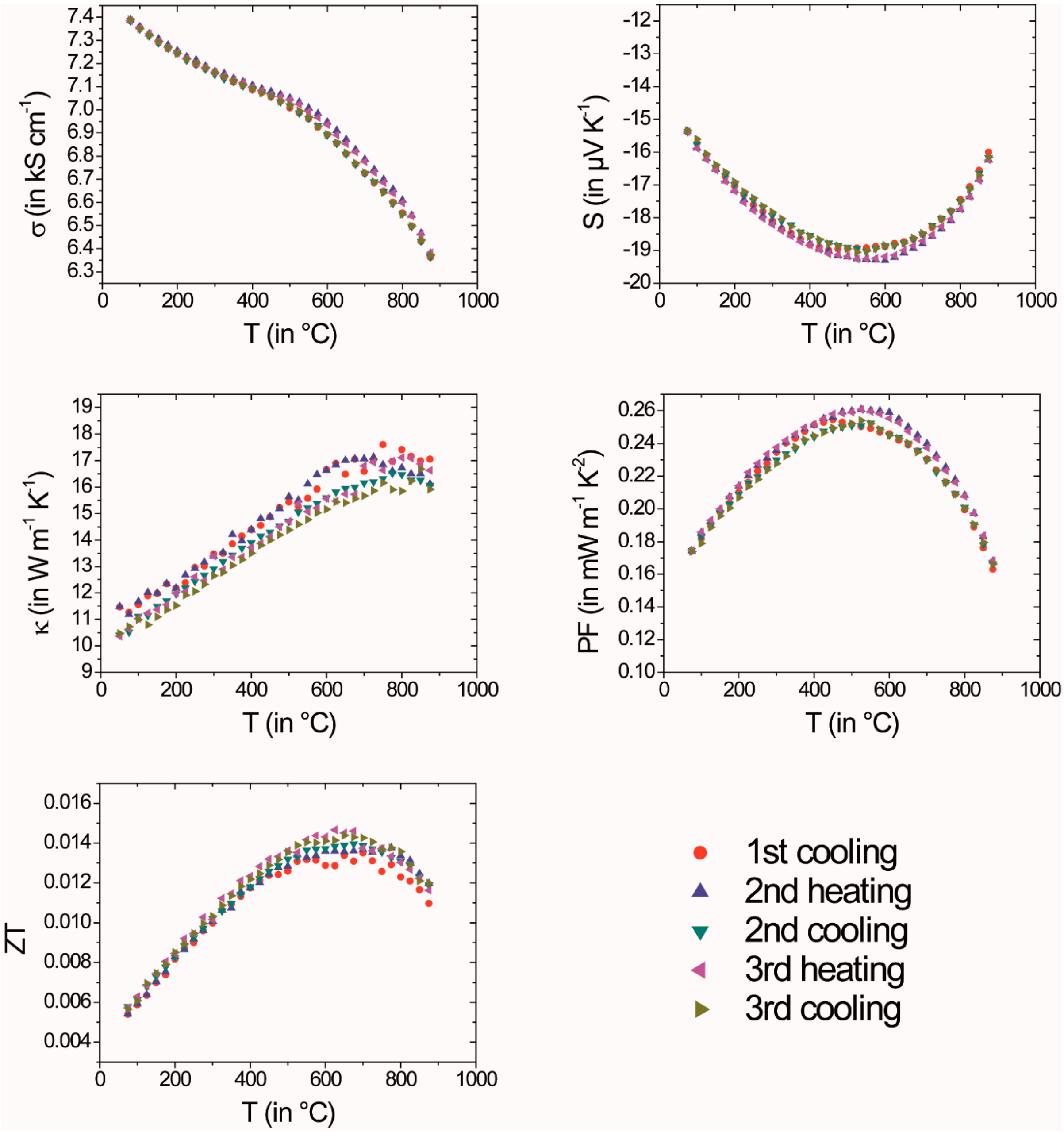

Figure S14. Thermoelectric properties of $Al_2CrCoFeNi$ + 0.3 w% Sc for three consecutive cycles (without first heating) up to 875 °C: electrical conductivity σ (top, left), Seebeck coefficient S (top, right), thermal conductivity (middle, left), power factor PF (middle, right) and thermoelectric figure of merit ZT (bottom, left).

References

1. Norris, S.D., Parker, J.D., Deformation processes during disc bend loading, *Mat. Sci. Tecnol.* **1996**, *12*, 163-170.
2. Lacalle, R., Alvarez, J.A., Gutiérrez-Solana, F., Analysis of key factors for the interpretation of small punch test results, *Fatigue Fract. of Eng. Mat Struct.* **2008**, *31*, 841-849.
3. Hurst, R., The european code of practice for small punch testing: where do we go from here?, *Metall. J.* **2010**, *63*, 5-11.
4. Džugan, J., Konopìk, P., Evaluation of fracture toughness properties for low carbon steel in the brittle state by small punch test technique, *Metall. J.* **2010**, *63*, 119-122.