Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

High thermoelectric figure of merit in well optimized and non-nanostructured $Yb_yCo_4Sb_{12}$

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SUPPLEMENTARY MATERIAL

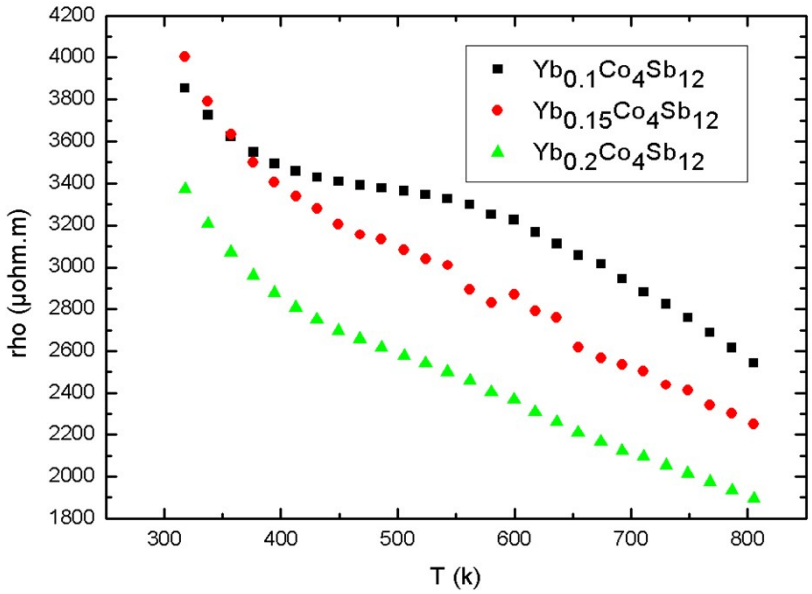

Fig. S1. Electrical resistivity in the first set $Yb_yCo_4Sb_{12}$ samples (y = 0.10, 0.15, 0.20) as a function of temperature. These measurements were carried out after spark-plasma sintering the powders used in the XANES experiment, which had been ground in air.

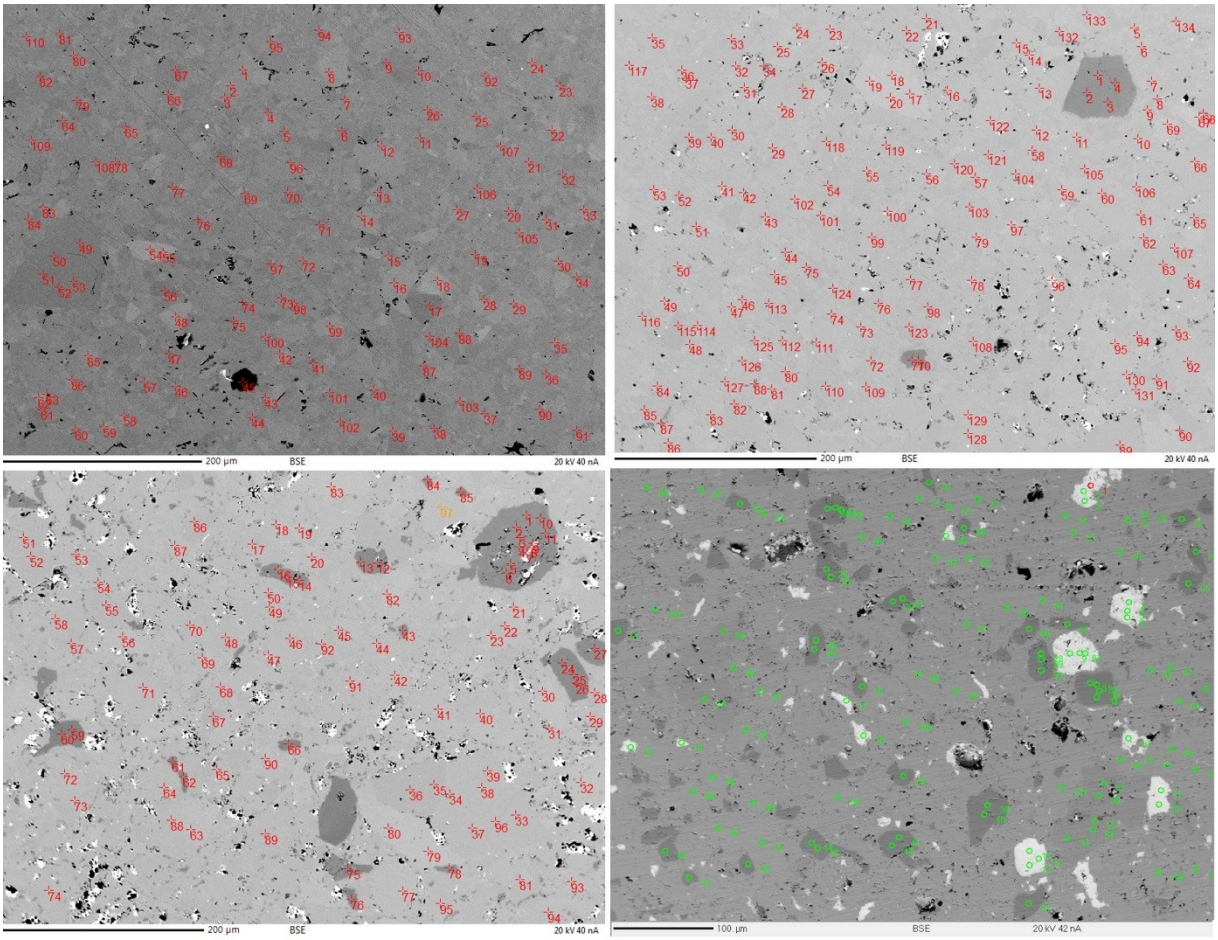

Fig. S2. Back-scattered electron image obtained with the electron micro-probe on some of the samples from set2. $Yb_{0.2}Co_4Sb_{12}$ (top-left corner), $Yb_{0.3}Co_4Sb_{12}$ (top-right corner), $Yb_{0.4}Co_4Sb_{12}$ (bottom-left corner) and $Yb_{0.5}Co_4Sb_{12}$ (bottom-right corner). The medium-grey phase is the skutterudite. The dark-grey phase is $CoSb_2$. The white phase is $YbSb_2$. The red or green dots and numbers represents the positions where the Electron Probe Micro-Analysis was performed.

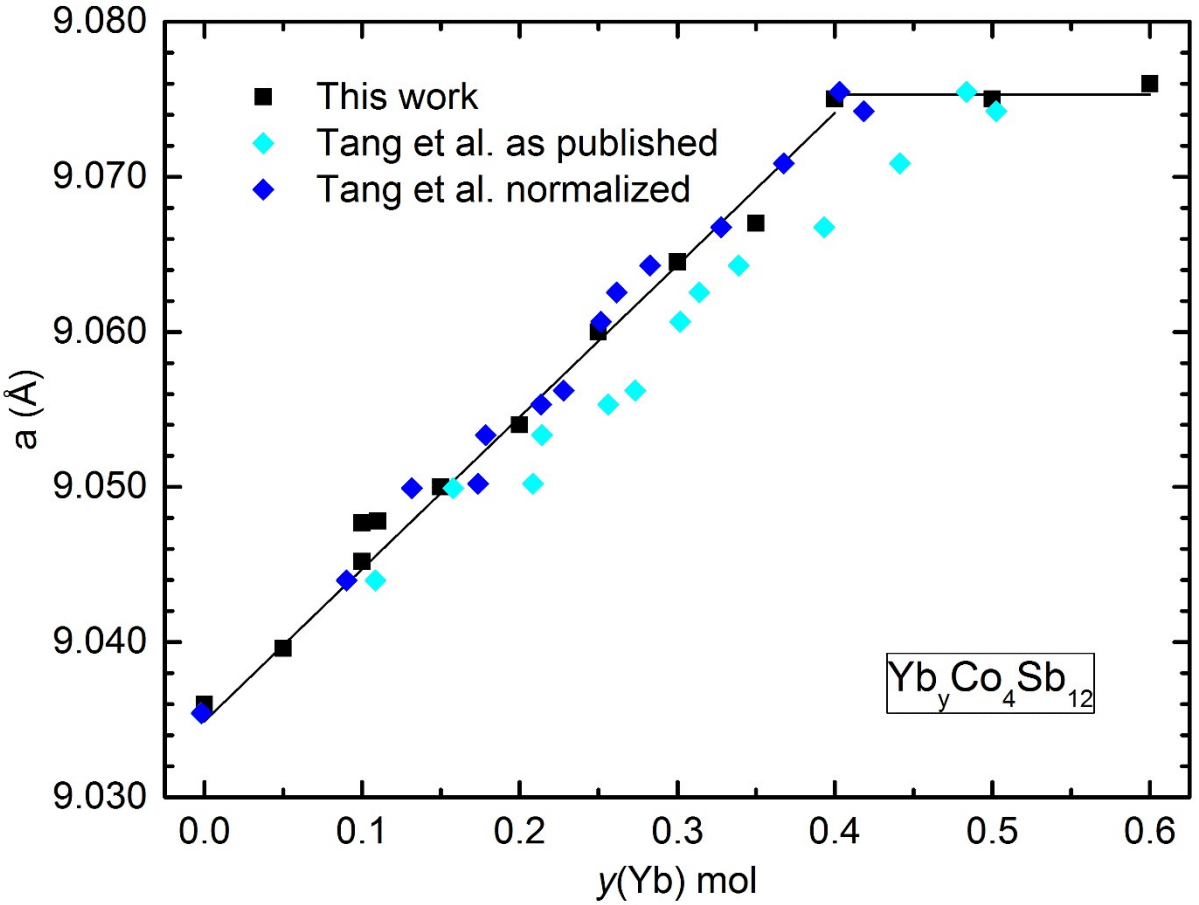

Fig. S3. Lattice parameter as a function of the ytterbium concentration in $Yb_yCo_4Sb_{12}$ synthesized at 1073 K. Comparison of the data of this work with the data of Tang et al. [1]. For this work y is both the nominal and effective ytterbium concentration until the solubility limit is reached at y_{lim} = 0.41. In Tang et al., y is the effective concentration measured by EPMA. Their data as published are shown along with their data where y has been normalized by 1.2. This 20% correction is required to make their sample with its composition measured by EPMA (y = 0.30) match its nominal composition (y = 0.25). After this correction, Tang et al. data match very well the present one.

T(K) \ y	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40
300	1.55	1.66	1.80	1.85	1.93	2.01	2.11	2.22
400	1.53	1.60	1.71	1.75	1.82	1.89	1.98	2.11
500	1.52	1.57	1.66	1.69	1.74	1.80	1.89	2.01
600	1.51	1.55	1.62	1.64	1.69	1.74	1.82	1.93
700	1.51	1.54	1.60	1.62	1.65	1.7	1.76	1.86
800	1.50	1.53	1.58	1.59	1.63	1.67	1.72	1.81

Table SI. Lorenz number calculated as a function of the ytterbium concentration (y) and temperature (T). The unit is 10^{-8} V^2 K^{-2}. The values for the y = 0.50 and 0.60 samples were taken equal to those of the y = 0.40 sample.

The model used here is a single parabolic band model with an effective density of states mass m^* = $3.2m_e$ (m_e is the bare electron mass) and an electronic relaxation time $\tau = \tau_0 \varepsilon^{-1/2}$. The ytterbium concentration is related to the measured Hall electron concentration n at 300 K through the expressions: $n(y,300K) = n_{Hall}(y,300K)$. The reduced chemical potential η (η = μ/k_BT) is related to y through the electron concentration $n(y,T) = n(\eta,T)$ by solving for η the following equation: $n(\eta,T) = 4\pi\left(\frac{2m * k_BT}{h^2}\right)^{\frac{3}{2}} F_{\frac{1}{2}}(\eta)$ with h and k_B the Planck and the Boltzmann constant respectively and with F_q a Fermi integral defined by the expression: $F_q = \int_0^\infty \frac{x^q}{[1+e^{(x-\eta)}]} dx$ where $x = \frac{\varepsilon}{k_BT}$ is the reduced energy. Finally, the Lorenz number is related to η by the expression:

$$L(\eta(n,T)) = \left(\frac{k_B}{e}\right)^2 \frac{3F_0(\eta)F_2(\eta) - 4F_1(\eta)^2}{F_0(\eta)^2}$$

These *L(y, T)* values were used to calculate the lattice thermal conductivity using the expression: $\lambda_L(y,T) = \lambda(y,T) - \frac{L(y,T)T}{\rho(y,T)}$.

1. Tang, Y.; Chen, S.-W.; Snyder, G. J., Temperature dependent solubility of Yb in Yb-CoSb3 skutterudite and its effect on preparation, optimization and lifetime of thermoelectrics. *Journal of Materiomics* **2015,** *1* (1), 75-84.