

Electronic supplementary information ESI-I:

Table1. The various molar ratios of hydrated zinc nitrate, ferric nitrate, chromium nitrate and their corresponding weights and the weight of the PVA used for the synthesis of the different $\text{ZnFe}_{2-x}\text{Cr}_x\text{O}_4$ samples.

Sample description	Molar ratios of nitrates of Zn, Fe and Cr	Weight of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (gm)	Weight of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (gm)	Weight of $\text{Cr}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (gm)	Weight of PVA (gm)
ZnFe_2O_4	0.1:0.2:0	2.970	8.080	0	1.771
$\text{ZnFe}_{1.8}\text{Cr}_{0.2}\text{O}_4$	0.1:0.18:0.02	2.970	7.272	0.800	1.763
$\text{ZnFe}_{1.6}\text{Cr}_{0.4}\text{O}_4$	0.1:0.16:0.04	2.970	6.464	1.601	1.756
$\text{ZnFe}_{1.4}\text{Cr}_{0.6}\text{O}_4$	0.1:0.14:0.06	2.970	5.656	2.401	1.748
$\text{ZnFe}_{1.2}\text{Cr}_{0.8}\text{O}_4$	0.1:0.12:0.08	2.970	4.848	3.201	1.740
ZnFeCrO_4	0.1:0.1:0.1	2.970	4.040	4.001	1.733
$\text{ZnFe}_{0.8}\text{Cr}_{1.2}\text{O}_4$	0.1:0.08:0.12	2.970	3.232	4.802	1.725
$\text{ZnFe}_{0.6}\text{Cr}_{1.4}\text{O}_4$	0.1:0.06:0.14	2.970	2.424	5.602	1.717
$\text{ZnFe}_{0.4}\text{Cr}_{1.6}\text{O}_4$	0.1:0.04:0.16	2.970	1.616	6.402	1.709
$\text{ZnFe}_{0.2}\text{Cr}_{1.8}\text{O}_4$	0.1:0.02:0.18	2.970	0.808	7.203	1.702
ZnCr_2O_4	0.1:0:0.2	2.970	0	8.003	1.694

Electronic supplementary information ESI-II:

The analysis of the positron lifetime spectra using the PALSfit program necessarily involves the subtraction of the background, de-convolution of the instrumental resolution and iteration for obtaining the best-fitted lifetime components and their intensities. It is however also important to delete from the spectra the contributions arising from the positron source itself by virtue of its configuration and geometry. A fraction of positrons while penetrating through the Ni foil may get annihilated in it and a minute number of positrons may get backscattered while entering into the sample. In addition to these, there is also the probability for a fraction of positrons to encounter annihilations within the source material ($^{22}\text{NaHCO}_3$) itself due to the presence of orbital electrons of the atoms of the elements composing it. The “source correction parameters” were estimated by first performing the positron lifetime measurements on high pure Al single crystals (99.999% purity), which were initially annealed at 873K for 2 hours in high vacuum ($\sim 10^{-6}$ mbar). The analyzed spectrum of Al gave three positron lifetimes (107 ps, 420 ps and 1.67 ns) besides the positron lifetime of 170 ps in it and they respectively originated from positron annihilations in Ni foil and the source material and scattering of positrons at the source-sample interfaces. The fraction of positrons annihilating within the Ni foil is calculated using the equation (M. Bertolaccini and L. Zappa, *Nuovo Cimento B*, 1967, 52, 487 – 494)

$$I_{\text{foil}}(\%) = 0.324Z^{0.93}d^{3.45/Z^{0.41}} \quad (16)$$

where Z is the atomic number of the sample and d is the thickness of the foil in mgcm^{-2} . This fraction is obtained as 11.9% while that from the source and the one due to back scattering are obtained directly from the analysis of the spectrum of Al as 8.9% and 0.08% respectively.

Electronic supplementary information ESI-III:

In the figure given here, the peak-normalized raw positron lifetime spectra of all the samples are illustrated. Although the differences can be brought out more prominently only when the positron lifetimes and their relative intensities are resolved during the PALSfit analysis, the nature of the spectra are indicative of very long positron lifetimes with appreciable intensities and indirectly hints at the presence of strong positron trapping centers of significant concentration in the samples. Further, the spectra also depict the statistics of the data collected. More than 10^6 coincidence events were accumulated under each spectrum with a peak-to-background ratio better than 40,000:1.

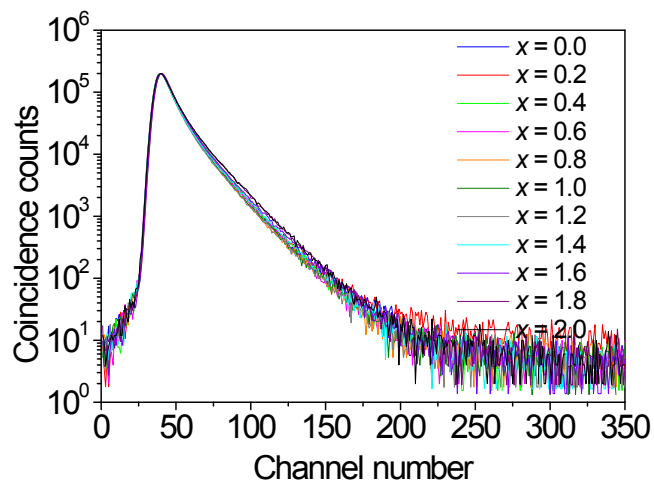


Figure 17. Peak-normalized positron lifetime spectra of all the samples. (1 channel = 50 ps).