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Probing the doping mechanisms and electrical properties of Al, Ga and In doped ZnO prepared by spray pyrolysis

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Supplementary Figures

Figure S1 - UV-Vis spectra of ZnO films doped with 0 **-** 3 at%, showing a) AZO, b) GZO and c) IZO. The measurements confirm high optical transparency across the visible and excellent thickness uniformity/homogeneity.

Figure S2 - a) shows the calculated bandgap values of doped ZnO films, doping with 0 - 3 at% Al (yellow), Ga (orange) and In (red). Band gaps were obtained form Tauc analysis using direct bandgap method, typical Tauc plot shown in b).

Figure S3 – XRD analysis of 2 at% AZO sample as deposited (blue) and following annealing (red). Spectra are focussed upon the dominant (002) orientation to highlight the lack of change in intensity or shift in position

Table S1 Calculated texture coefficients, obtained using formula at foot of table showing the calculated preferred orientation for all films deposited in the current study.

Table S2 Shows measured electrical characteristics for a range of AZO films reported in the literature – **note** all film thicknesses < 300 nm.

Table S3 Showing measured electrical characteristics for a range of AZO films reported in the literature – **note** all film thicknesses > 300 nm.

Table S4 Shows a selection of AZO films from existing literature subjected to post-deposition annealing treatments.

Table S1

Dopant concentration	**Species**	**Tc_{hkl}**			
		(002)	**(101)**	**(102)**	**(103)**
0	ZnO	4.29	0.18	0.97	1.56
0.1 at%	AZO	4.54	0.39	0.76	1.31
	GZO	4.10	0.14	1.07	1.69
	IZO	3.74	0.20	1.09	1.97
0.2 at%	AZO	3.51	0.38	1.24	1.87
	GZO	3.62	0.38	1.18	1.82
	IZO	3.34	0.16	1.24	2.26
0.3 at%	AZO	3.48	0.42	1.33	1.77
	GZO	3.34	0.18	1.66	1.82
	IZO	3.51	0.19	1.24	2.06
0.4 at%	AZO	3.85	0.29	1.01	1.84
	GZO	3.03	0.14	1.35	2.49
	IZO	4.56	0.18	0.78	1.48
0.5 at%	AZO	4.27	0.30	0.76	1.68
	GZO	3.55	0.13	1.43	1.89
	IZO	4.85	0.20	0.67	1.29
1 at%	AZO	5.15	0.14	0.53	1.19
	GZO	2.57	0.14	1.71	2.59
	IZO	5.18	0.09	0.51	1.21
2 at%	AZO	4.96	0.23	0.57	1.24
	GZO	4.37	0.21	0.94	1.48
	IZO	5.71	0.14	0.31	0.84
3 at%	AZO	3.96	0.57	1.22	1.25
	GZO	4.38	0.19	1.62	0.81
	IZO	5.09	0.12	0.74	1.05

$$Tc_{hkl} = \frac{I_{(hkl)} /\ I_{x(hkl)}}{(1/n_d)\sum I_{(hkl)} /\ I_{x(hkl)}}$$

Table S2.

Al at%	**Thickness (nm)**	**Resistivity (Ω.cm)**	**Hall Mobility (cm^2/V.s)**	**Carrier concentration (cm^{-3})**	**Year and reference**
1	270	1.3	1.0	2.4×10^{19}	2012 [1]
1	200	0.8	-	-	2006 [2]
2	275	1.75	-	-	2014 [3]
3	~200	0.6			2004 [4]
0.3	200	3	-	-	2006 [5]
3	200	0.9	-	-	2007 [6]
1.5	~285	0.1	-	-	2011 [7]
3	45	3	-	-	2014 [8]

Table S3

Al at%	Thickness (nm)	Resistivity (Ω.cm)	Hall Mobility ($cm^2/V.s$)	Carrier concentration (cm^{-3})	Year and reference
3	~1000	3×10^{-3}	-	-	2007 [9]
3	2400	8×10^{-2}	-	-	2007 [6]
2	~450	9×10^{-1}	-	-	1995 [10]
2	1000	4×10^{-3}	12	9×10^{19}	2000 [11]
2	1800	2.1×10^{-2}	3	9×10^{19}	2014 [3]
2.5	600	3×10^{-2}	-	-	2007 [12]
3	600	2×10^{-2}	1	3×10^{20}	2010 [13]
3	600	1×10^{-2}	~5	~7×10^{19}	2010 [14]
3	603	2×10^{-3}	8	8×10^{20}	2013 [15]
~1.75	~500	5×10^{-2}	-	-	2014 [16]
1	400	3.3	20	1×10^{17}	2015 [17]
1	1260	2×10^{-2}	0.6	9×10^{20}	2015 [17]

Table S4

Al at %	Thickness (nm)	Resistivity (Ω.cm)	Hall Mobility (cm^2/V.s)	Carrier concentration (cm^{-3})	Anneal temp (° C)	Anneal environment	Year and reference
3	200	3.6×10^{-3}	7	2.5×10^{20}	600	N_2 + 5% H_2	2012 [18]
3	2400	2×10^{-3}	-	-	400	Vacuum	2007 [6]
0.3	1000	0.9			700	O_2	2004 [19]
3	~200	1×10^{-1}	-	-	500	N_2	2004 [4]
3	~200	1×10^{-2}	-	-	500	N_2 + 5% H_2	2004 [4]
1	~450	1.4×10^{-3}	-	-	350	H_2	1995 [10]
2	2200	9.1×10^{-3}	4.4	1.6×10^{20}	400	Ar + H_2	2014 [3]
1.2	6600	2×10^{-3}			400	H_2	1991 [20]
1.75	~500	7×10^{-3}	-	-	350	N_2	2014 [16]
3	600	4×10^{-3}	~1	~10^{20}	400	Vacuum	2007[12]

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