Supplementary Information

Purification Method Dependent Fluorescence from Nitrogen-vacancy (NV) Centers of Nano-diamonds

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Estimation of Inter-planar lattice spacing (d) from selected area electron diffraction (SAED) pattern

For ND0, diffraction spots and rings are observed at inter-planar spacing (plane) of 0.113 nm (220), 0.063 nm (400) and 0.052 (unassigned plane) of diamond. The observed inter-planar spacing is lower than the standard inter-planar spacing of bulk diamond (PCPDF No. # 060675: 0.206 nm (111), 0.126 nm (220) and 0.890 nm (400)). This indicates presence of compressive strain in the initial sample grown by HPHT. After purification sample ND35-R8HCL shows diffraction rings at 0.231 (111), 0.143 (220) and at 0.123 (220) lattice planes respectively, see inset of Fig. 1(c). For ND35-OXDHCL the SAED patterns are observed at 0.22 nm (111) plane and 0.138 nm (220).

Scanning Electron Microscopy (SEM)

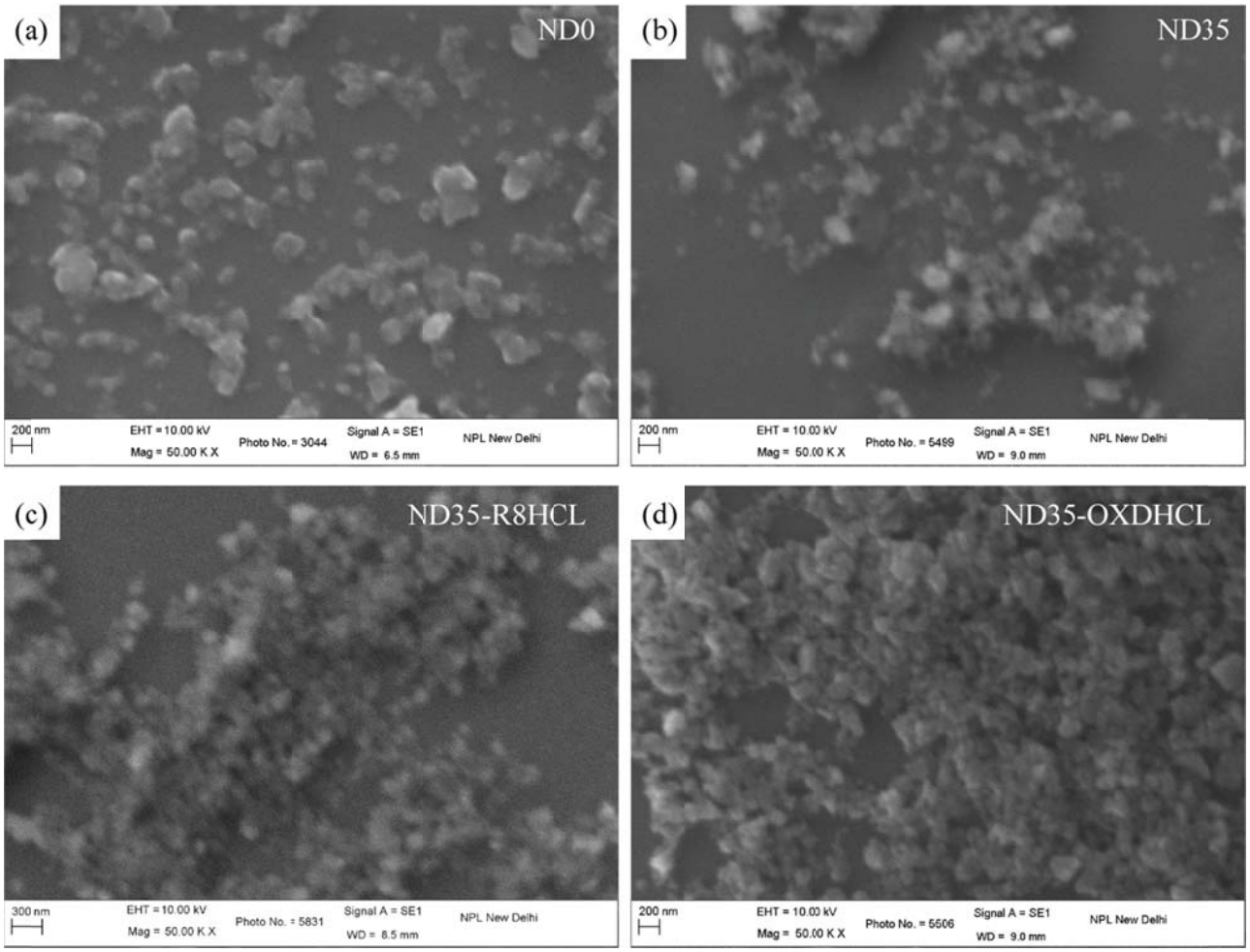

Fig. S1 SEM micrographs of initial sample (ND0), milled sample (ND35), sample purified through acid reflux (ND35-R8HCL) and sample purified through air oxidation (ND35-OXDHCL). Sample purified through wet chemical route shows smaller size than the sample purified through air oxidation.

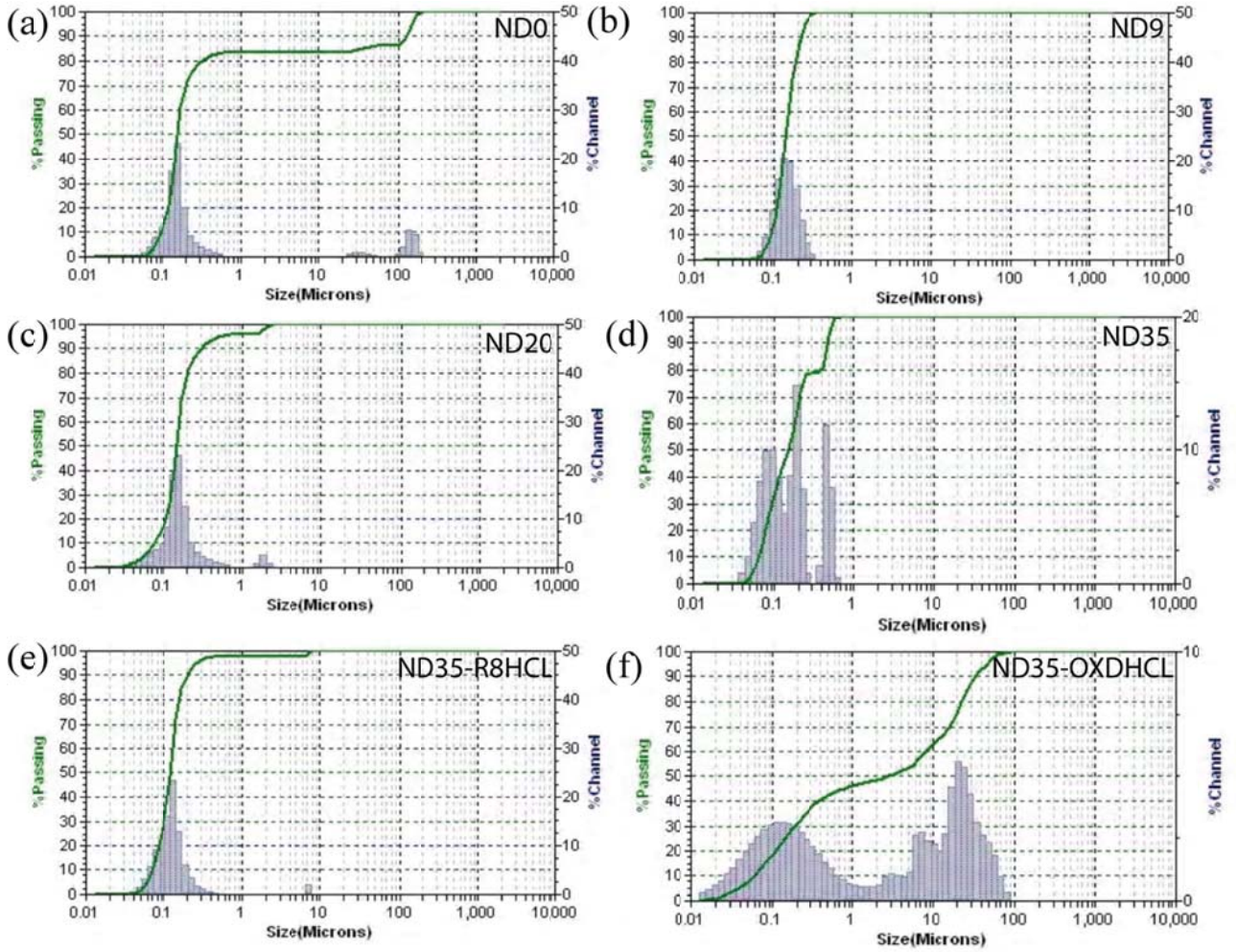

Fig. S2 Particle size distribution histogram obtained from Dynamic light scattering measurements for (a) Initial sample: ND0; Milled samples (b) ND9, (c) ND-20, (d) ND-35, Purified samples (e) ND35-R8HCl and (f) ND35-OXDHCl.

Table-SI: XRD line-profile of milled, purified and irradiated nano-diamond samples.

Peak Profile	Peak1	Peak2	Crystallite size (Peak1)	Crystallite (Peak 2)
ND0				
Center (deg)	43.703	43.888	16.3	25.5
FWHM(deg)	0.524	0.335		
Ampl.	636.7	1657.9		
ND3				
Center (deg)	43.667	43.766	13.0	29.9
FWHM(deg)	0.659	0.286		
Ampl.	660.9	1185.9		
ND6				
Center (deg)	43.541	43.791	16.5	25.3
FWHM(deg)	0.517	0.338		
Ampl.	152.7	757.8		
ND9				
Center (rad)	43.555	43.811	15.3	25.1
FWHM(rad)	0.557	0.340		
Ampl.	213.1	1011.5		
ND15				
Center (deg)	43.654	-	-	27.7
FWHM(deg)	0.309			
Ampl.	73.4			
ND20				
Center (deg)	43.775	-	-	25.3
FWHM(deg)	0.338			
Ampl.	88.4			
ND35				
Center (deg)	43.932	-	-	23.2
FWHM(deg)	0.370			
Ampl.	65.6			
ND35-R8HCl				
Center (deg)	43.364	43.724	18.7	25.8
FWHM(deg)	0.457	0.332		
Ampl.	38.1	352.4		
ND35-OXDHCl				
Center (deg)	43.383	43.710	18.8	27.4
FWHM(deg)	0.455	0.310		
Ampl.	44.1	349.4		

Table-SII: Raman peak summary

Sample		Peak1	Peak2	Peak3	Peak4	Peak5	Peak6	Peak7
ND0	*Center (cm^{-1})*	1331.8	1351.5	1482.3	1578.7	1615.7	1688.2	
	FWHM (cm^{-1})	5.9	62.3	15.6	83.5	22.6	20.7	
	Ampl.	7154.0	1545.4	213.1	3155.3	466.9	267.3	
	Area	63770	108338	4077	340281	11229	7423	
	% Content	12.4	21.2		66.4			
ND9	*Center (cm^{-1})*	1330.8	1350.8		1583.4	1614.9	1704.4	1798.1
	FWHM (cm^{-1})	6.17	53.0		80.2	21.8	59.0	66.7
	Ampl.	17940.9	3431.4		4921.6	795.0	968.3	2125.1
	Area	168099	221103		519696	26083	77703	109067
	% Content	18.5	24.3		57.2			
ND20	*Center (cm^{-1})*	1332.5	1354.1	1472.9	1584.4	1619.9		
	FWHM (cm^{-1})	5.1	62.3	40.0	75.5	24.9		
	Ampl.	8381.8	1455.4	283.7	1886.5	389.9		
	Area	65322	101288	13900	180071	14005		
	% Content	18.8	29.2		51.9			
ND35	*Center (cm^{-1})*	1331.5	1354.9		1586.8	1618.2	1737.8	
	FWHM (cm^{-1})	5.4	41.0		78.7	18.2	188.6	
	Ampl.	5438.7	1208.4		1662.7	385.2	2074.8	
	Area	44902	65447		167108	10518	271365	
	% Content	16.2	23.6		60.2			
ND35-R8HCL	*Center (cm^{-1})*	1332.7	1340.9	1461.6	1569.5			1227.1
	FWHM (cm^{-1})	5.1	74.0	107.7	80.9			54.7
	Ampl.	34947.3	2972.9	2747.4	3937.9			1779.4
	Area	277912	310740	401831	427637			129470
	% Content	27.3	30.6		42.1			
ND35-OXDHCL	*Center (cm^{-1})*	1332.3	1344.3		1576.1			
	FWHM (cm^{-1})	4.7	65.0		70.2			
	Ampl.	73315.1	2374.8		1853.9			
	Area	542284	218668		182997			
	% Content	57.4	23.2		19.4			
ND0-IRR	*Center (cm^{-1})*	1331.9	1338.6	1368.9	1579.6	1608.6		
	FWHM (cm^{-1})	3.5	86.5	4.04	63.5	41.3		
	Ampl.	1371.1	10310.0	3155.5	6313.9	7291.9		
	Area	7466	1237256	199958.8	426512.7	320858.5		
	% Content	0.4	66.1	10.7	22.8			
ND35- R8 HCLIRR	*Center(cm^{-1})*	1332.5	1340.5	1494.1	1595.9	1690.9		
	FWHM (cm^{-1})	5.8	73.7	87.2	32.9	23.2		
	Ampl.	3423.6	15939.1	7378.5	14838.05	3223.7		
	Area	21256	3342994	1761906	1225006	217282		
	% Content	0.5	72.8		26.7			
ND35-OXD HCLIRR	*Center (cm^{-1})*	1331.6	1345.7	1479.6	1580.3			
	FWHM (cm^{-1})	4.64	55.6	71.4	70.46			
	Ampl.	19380.05	5159.2	2451.9	7719.2			
	Area	139895	402604	247595	578979			
	% Content	12.5	35.9		51.6			
Assignment		*D-band*		*Poly-acetylene*	*G-band*	*D´-band*	M^-	M^+

Table-SIII Shows the fitted XPS C 1*s* core peaks for the different Nano diamond samples.

Sample Name	Peak1	Peak2	Peak3	Peak4	Peak5	Peak6
ND0						
Center(eV)	286.2	286.9				
Area	5673.6	1419.1				
FWHM (eV)	11.5	3.1				
% Content	79.9	20.0				
ND35						
Center(eV)	285.2	285.7				
Area	3970.3	975.7				
FWHM (eV)	1.2	2.7				
% Content	80.2	19.8				
ND35-R8HCL						
Center(eV)	285.0	286.2	288.8	288.8	294.4	297.0
Area	688.3	456.3	695.5	695.5	73.8	30.9
FWHM (eV)	1.5	1.8	6.9	6.9	1.7	1.5
% Content	26.1	17.3	26.3	26.3	2.8	1.2
ND35-OXYHCL						
Center(eV)	284.9	286.5	291.1	293.7		
Area	1583.3	289.8	140.6	35.5		
FWHM (eV)	1.6	2.3	9.3	1.8		
% Content	77.3	14.2	6.9	1.7		
ND0-IRR						
Center(eV)	285.3	286.5				
Area	4441.4	2462.5				
FWHM (eV)	1.3	2.6				
% Content	64.3	35.7				
ND35-R8HCLIRR						
Center(eV)	285.4	285.9		293.7		
Area	4729.6	1505.9		288.4		
FWHM (eV)	1.2	2.4		12.8		
% Content	72.4	23.1		4.4		
ND35-OXYHCLIRR						
Center(eV)	288.7	288.9		290.4		
Area	1015.9	344.1		4718		
FWHM (eV)	4.7	1.3		1.4		
% Content	16.7	5.6		77.6		
Assignment	sp^2	sp^3	*R-OH, RC(=O)R', R-COOH groups*			

Table-SIV Shows the fitted XPS C1*s* core plasmon peaks for the different Nano diamond samples.

Sample Name	Peak1	Peak2	Peak3	Peak4	Peak5	Peak6	Peak7
ND0							
Center(eV)					313.0	321.0	340.1
Area					2643.4	1096.8	3289.4
FWHM(eV)					20.3	9.0	48.6
ND35							
Center(eV)					309.0	319.5	348.1
Area					1175.1	1716.5	919.3
FWHM(eV)					11.1	11.0	19.6
ND35-R8HCL							
Center(eV)	306.3		347.6	351.1	356.1	368.5	374.4
Area	277.5		10364	943.0	588.2	91.3	21.1
FWHM(eV)	2.7		1.7	1.9	10.5	1.8	0.7
ND35-OXYHCL							
Center(eV)	307.6	318.9	347.5	351.0		368.3	374.2
Area	334.2	530.5	89.1	109.5		44.1	18.8
FWHM(eV)	9.4	12.4	1.7	2.6		1.0	0.8
ND0-IRR							
Center(eV)		315.9	320.5				
Area		4248.4	590.3				
FWHM(eV)		23.9	7.3				
ND35-R8HCLIRR							
Center(eV)	309.6		320.3	348.0			
Area	1675.3		2054.6	2821.1			
FWHM(eV)	13.5		10.7	36.4			
ND35-OXYHCLIRR							
Center(eV)	314.0	324.8	351.7				
Area	1339.5	1849.6	1081				
FWHM(eV)	13.7	11.1	22.3				

Table-SV Shows the XPS fitted peaks for the Nitrogen 1*s* and Oxygen 1*s* core of different Nano-diamond samples.

Sample Name	Peak1	Peak2	Peak3	Peak4	Peak5	Peak6	Peak7
ND0							
Center(eV)	400.1	531.4	533.6		536.3		
Area	22.3	116.5	713.2		98.7		
FWHM(eV)	2.3	1.7	2.7		5.0		
ND35							
Center(eV)		530.8	532.2	530.2			
Area		788.5	1045.7	296.1			
FWHM(eV)		1.6	2.1	0.9			
ND35-R8HCL							
Center(eV)		531.0	532.5				
Area		2358.7	1270.0				
FWHM(eV)		1.8	1.9				
ND35-OXYHCL							
Center(eV)		531.4	533.8	530.7			
Area		1219.0	134.5	1908.8			
FWHM(eV)		2.5	1.6	1.3			
ND0-IRR							
Center(eV)		531.6	533.1				555.3
Area		62.5	818.2				179.1
FWHM(eV)		1.8	2.0				14.7
ND35-R8HCLIRR							
Center(eV)		531.8	533.5				556.2
Area		138.5	1247.9				245.1
FWHM(eV)		2.1	2.0				11.5
ND35-OXYHCLIRR							
Center(eV)	405.1	531.5	533.5		536.3	538.0	
Area	14.5	102.6	323.7		152.3	463.1	
FWHM(eV)	3.4	1.6	1.9		2.1	2.1	
Assignment	*N 1s*	*O 1s core*					

Table-SVI shows the XPS core area and the atomic percentage for the different elements present at surface (within XPS probing depth).

Sample name	Area			At. Conc. (%)		
	C 1*s* core	O 1*s* core	N 1*s* core	Carbon	Oxygen	Nitrogen
ND0	7092.7	928.7	22.3	95.7	4.1	0.2
ND35	4946.0	2130.3	-	87.5	12.5	-
ND35-R8HCL	1944.8	3628.7	-	61.9	38.1	-
ND35-OXYHCL	2049.2	3262.3	-	65.5	34.5	-
ND0-IRR	6903.9	1059.8	-	95.2	4.8	-
ND35-R8HCLIRR	6523.9	1631.5	-	92.4	7.6	-
ND35-OXYHCLIRR	6078.0	1041.7	14.5	94.5	5.3	0.1

Table-SVII: Photoluminescence spectra fitted with Gaussian line-shapes.

Sample	Peak1	Peak2	Peak3	Peak4	Peak5	Peak6	Peak7	Peak8
ND0								
Center (nm)	552	559		627			666	700
FWHM(nm)	9.2	4.7	-	122.4		-	40.0	63.1
Ampl.	201	199		3647			516	1623
ND35								
Center (nm)	552	558	559	609			667	707
FWHM(nm)	2.6	40.7	4.4	73.2	-	-	63.8	61.2
Ampl.	74	157	59.2	921			884	520
ND35-R8HCL								
Center (nm)		569		608		662		
FWHM(nm)	-	63.4	-	53.2	-	96.2	-	-
Ampl.		1566		1583		3650		
ND35-OXDHCL								
Center (nm)	551		575	602	638	659		683
FWHM(nm)	0.2	-	4.0	74.8	6.5	18.3	-	82.9
Ampl.	509		51.3	418	126	134		1444
ND0-IRR								
Center (nm)	552	559	576	605		655		695
FWHM(nm)	6.4	2.9	7.9	68.1		58.7	-	62.8
Ampl.	742	776	1099	13118		17395-		19432
ND35-R8HCLIRR								
Center (nm)	552	559	575	605	638	658	660	698
FWHM(nm)	2.5	1.3	6.4	64.6	5.4	8.6	57.6	61.4
Ampl.	408	285	4818	17468	3235	1838	22019	20699
ND35-OXDHCLIRR								
Center (nm)			575.1	600.8	637.8	658.6	675.6	715.8
FWHM(nm)			5.02	63.8	4.95	9.17	80.7	32.8
Ampl.			3186.6	12267.8	3920.8	2766.1	38595.8	2370.1
Assignment	*D-band*	*G-band*	*NV^0*	*NDs*	*NV^-*	*Fluorescence from NDs*		