Supplementary Information (SI) for Nanoscale.

Supporting Information for

Mechanics on granular distribution of aligned carbon nanotube bundles

Byeonghwa Goh,[1] Joonmyung Choi[1*]

[1]Department of Mechanical Design and Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea

[*]Corresponding author. Email: joonchoi@skku.edu

This file includes:

- **Note S1**
- **Figs. S1–S20** and their captions
- **Table S1** and its caption

Note S1. Manual demonstration of granular distribution in twisted bundle unit

The granular distribution of the diameters of aligned tubes was first demonstrated using a twisted rice straw rope. We prepared the unit section of a twisted bundle of traditional Korean rice straw rope, "Saekki." Twisting rice straw bundles into Saekki ropes is akin to the dry spinning of MWCNT yarns from MWCNT forests[1] (Fig. S2). A bundle of 25-cm-long rice straws, with diameter and thickness variations of 4–6 mm and 0.2–0.5 mm, respectively (Fig. S3), effectively demonstrated granular distribution. The bundle was tied in the middle and subjected to repeated twisting and untwisting by tightening the string in the hoop direction to induce hydrostatic pressure stimuli. After 100 twist cycles, the initial uniform distribution of the rice straw was separated into core and sheath domains, with smaller diameters inside and larger diameters outside (Fig. S4a). The rearrangement process of the small- and large-diameter straws under repeated stimuli is shown in Fig. S4b. Hydrostatic pressure elastically deformed the straws in the radial direction. Owing to structural deformation, the smaller straw gets stuck in the gap between the imperfect packing of the larger straws. Eventually, the dissipated stored elastic energy moved and rearranged the straw when the external pressure was relieved.

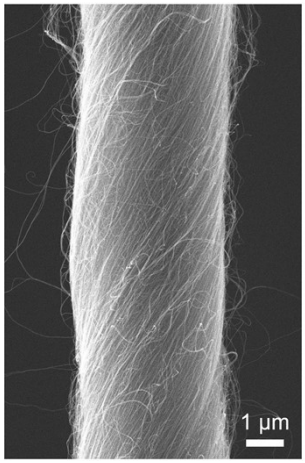

Fig. S1. Full image of the twisted MWCNT yarn. Reproduced with permission [13]. Copyright 2022, Elsevier.

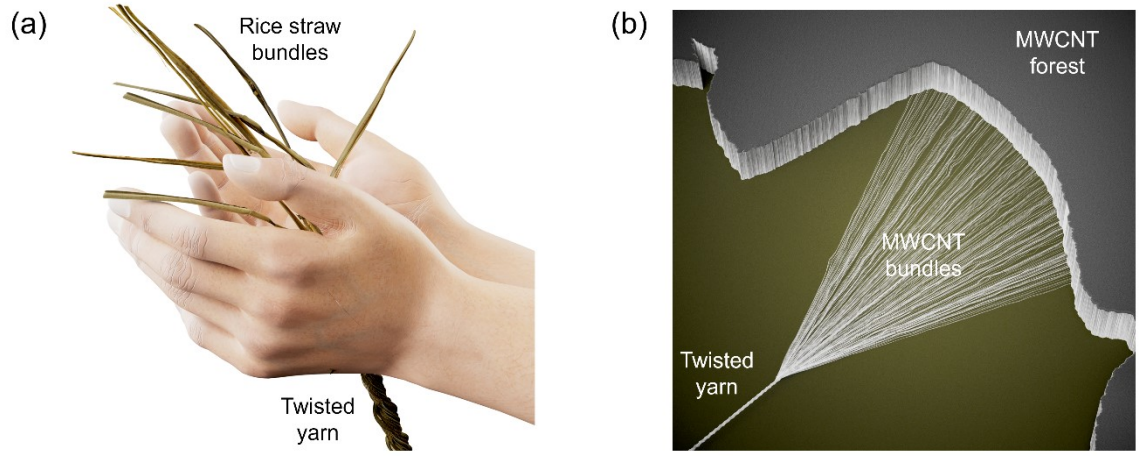

Fig. S2. Schematic showing the fabrication methods of **(a)** hand-made Saekki rope and **(b)** dry-spun MWCNT yarn twist.

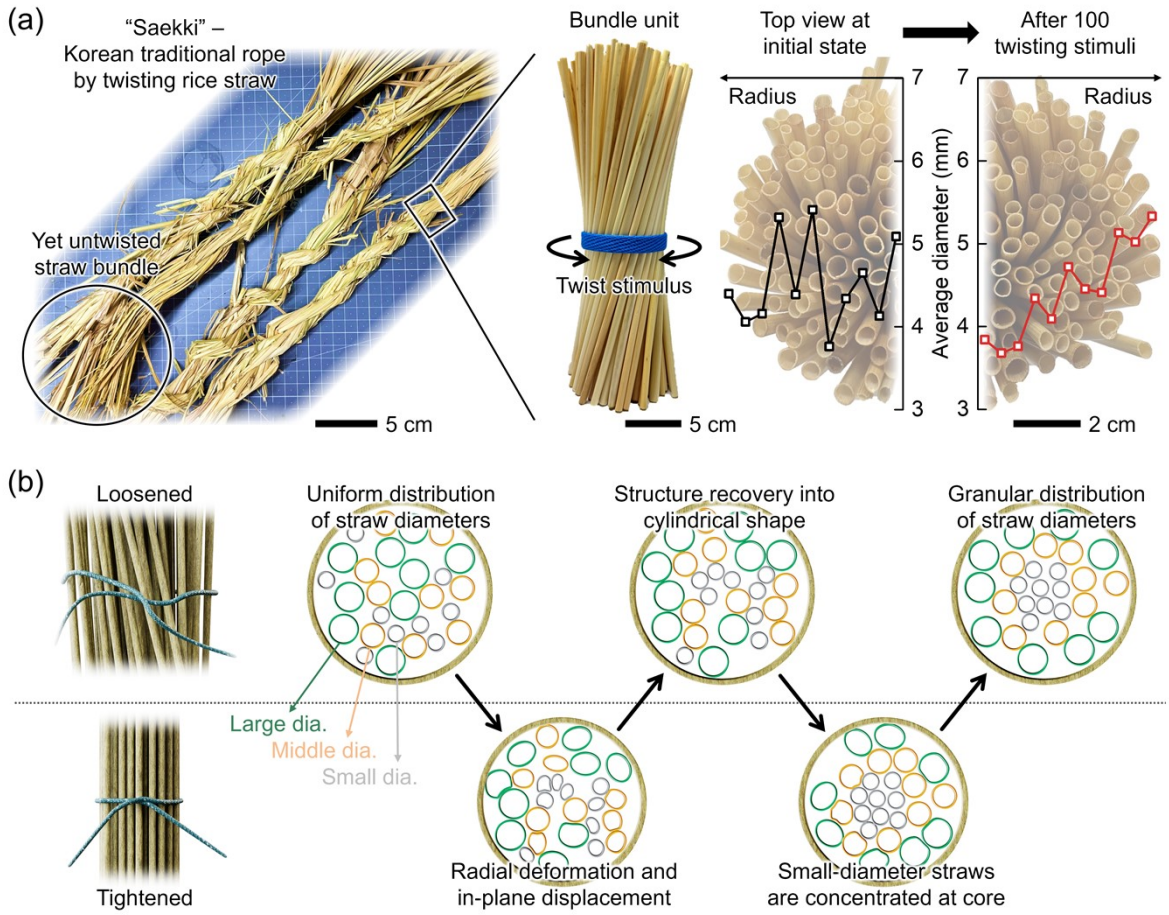

Fig. S3. Granular distribution of diameter inside the twisted bundle. **(a)** Twisted bundle unit of "Saekki" (the rice straw rope) for analysing radial distribution of individual diameters. Uniform distribution of straw diameter became granular after one hundred twisting stimuli are applied. The small- and large-diameter straws are distributed in the inside and outside, respectively, of the bundle. **(b)** Schematic showing the dynamics of straws. Twisting pressure elastically deforms the straws in the radial direction. Removed pressure relieves the elastic strain energy to shift the straws in the lateral direction. Continued deformation and shift of the straws eventually collect the small-diameter straws inside the bundle.

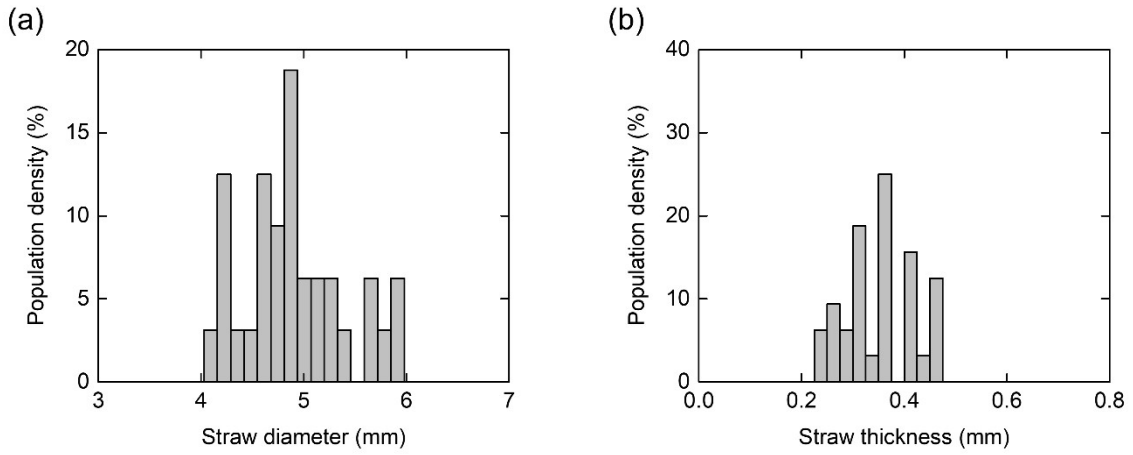

Fig. S4. Geometrical characterization of rice straw. Population densities of **(a)** diameter and **(b)** thickness of cylindrical straws.

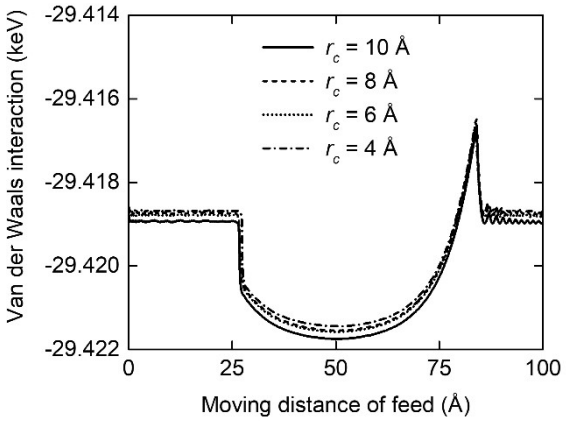

Fig. S5. van der Waals interactions of the (3,3,1.0) model with different cutoff radii for the non-bonded interaction.

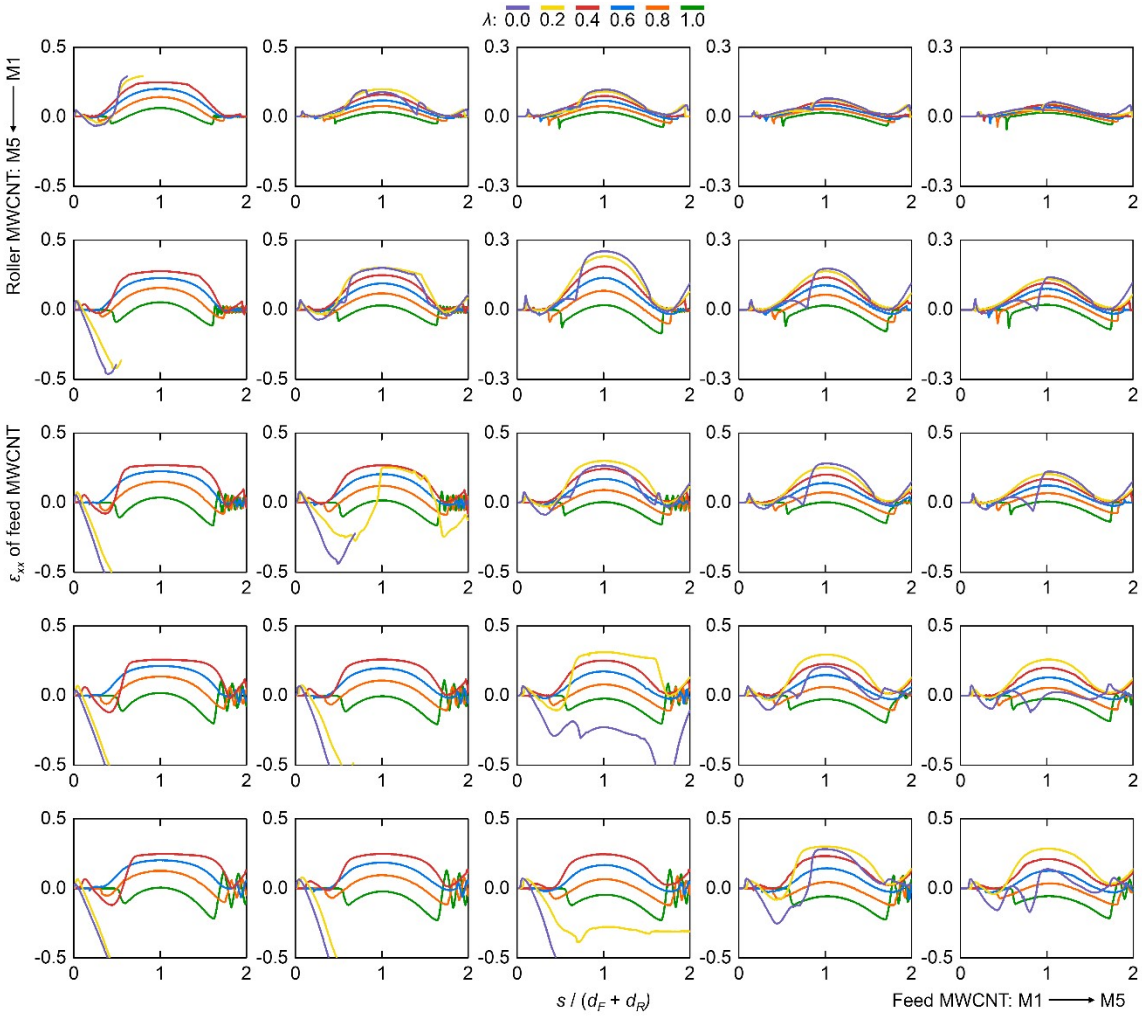

Fig. S6. In-plane strain of feed MWCNT in x-direction.

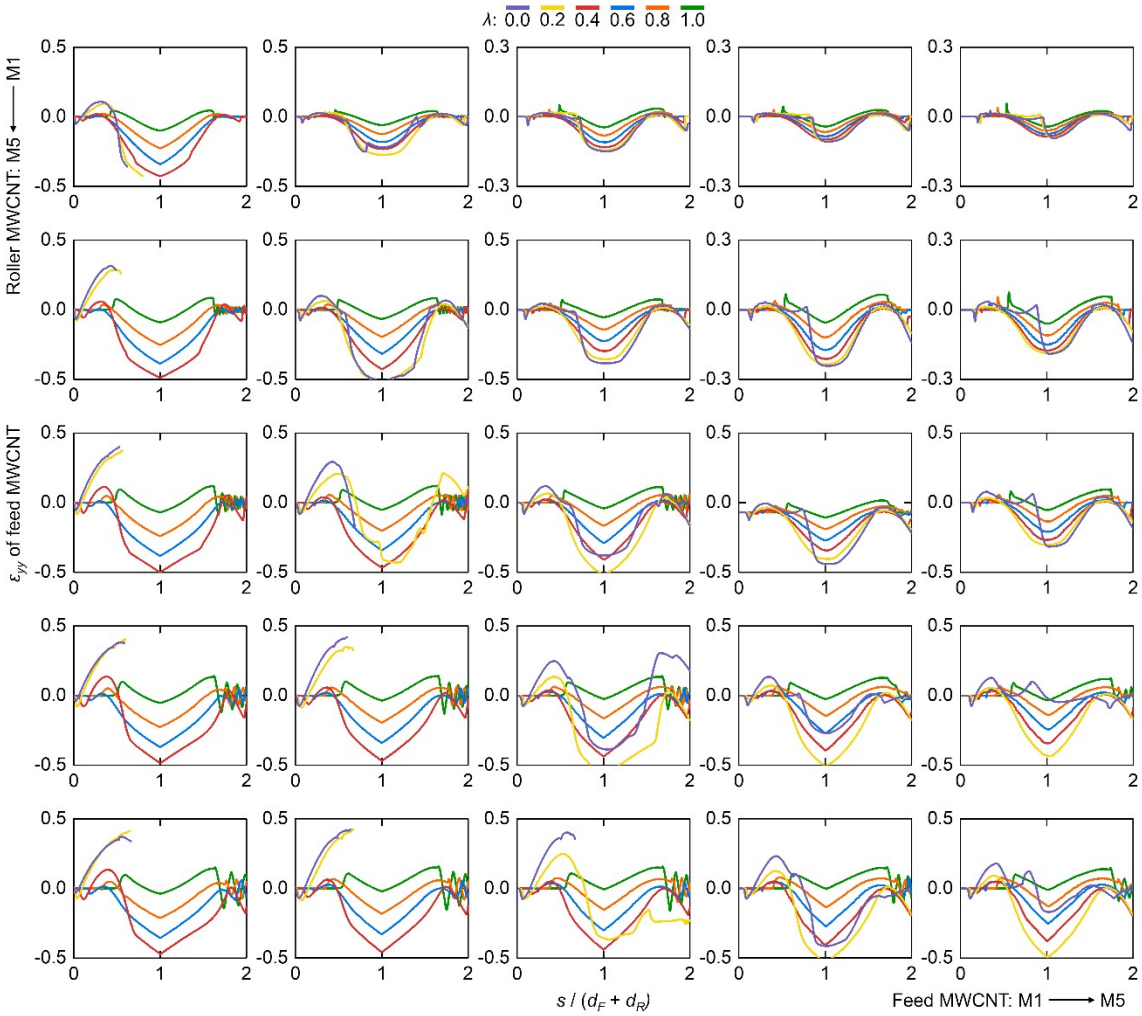

Fig. S7. In-plane strain of feed MWCNT in y-direction.

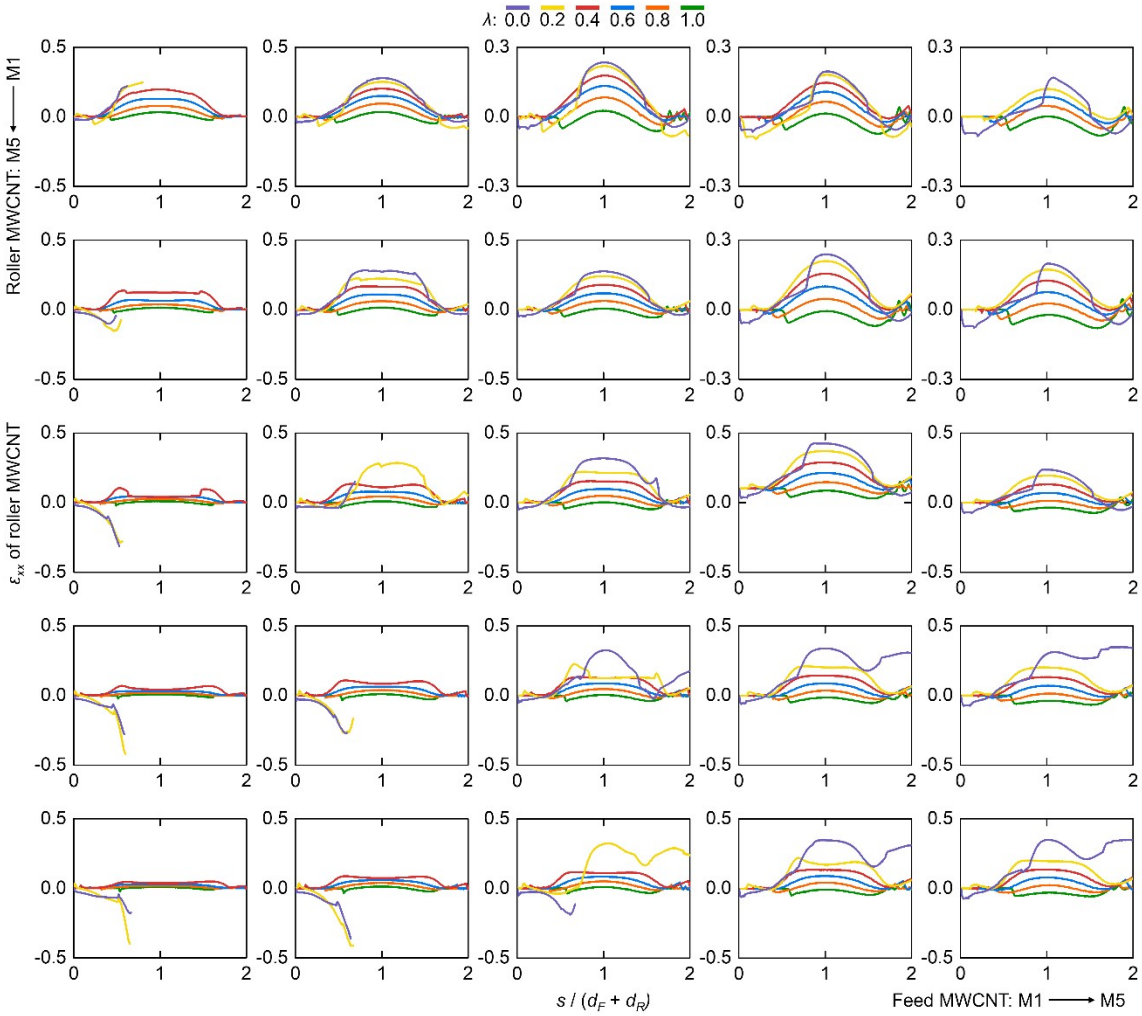

Fig. S8. In-plane strain of roller MWCNT in x-direction.

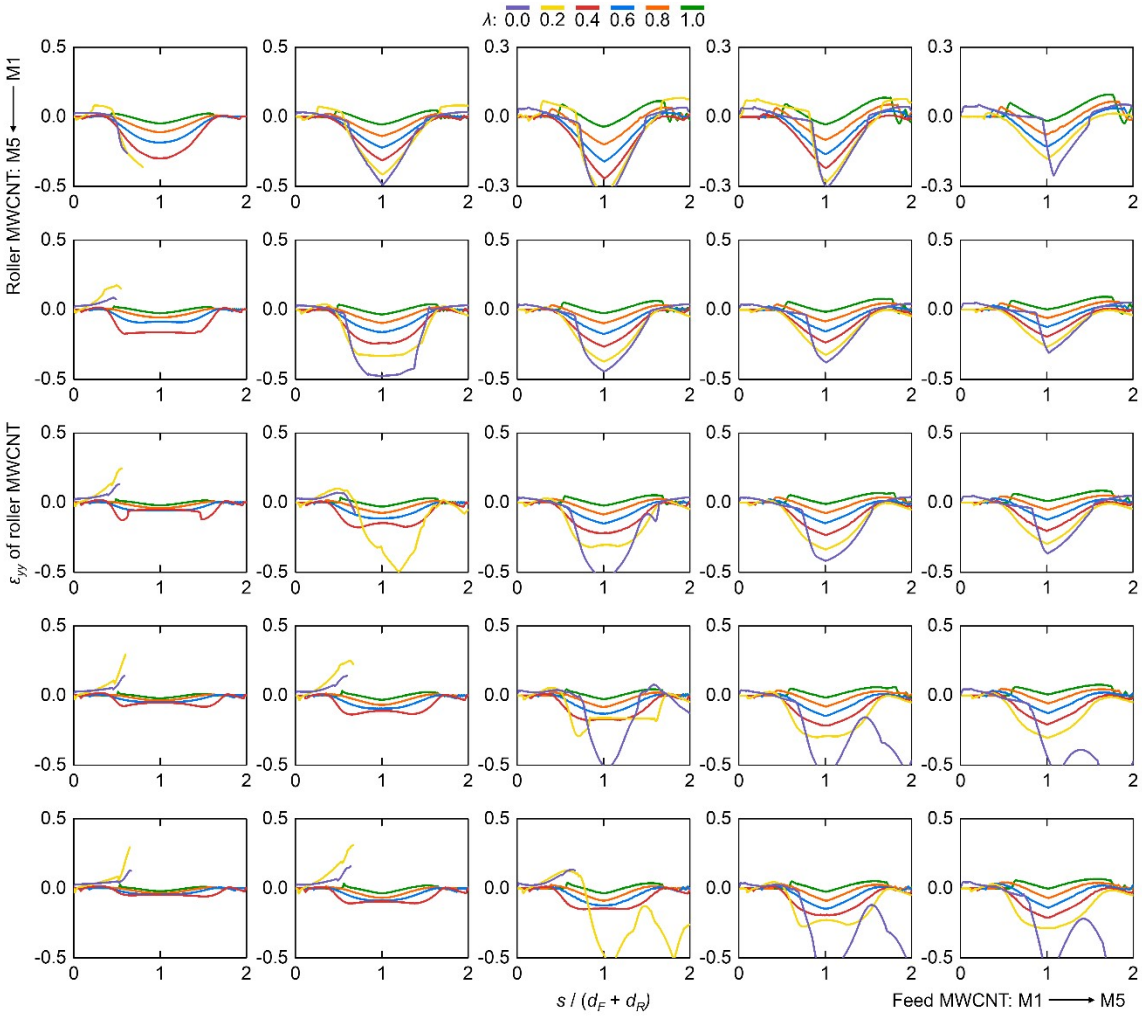

Fig. S9. In-plane strain of roller MWCNT in y-direction.

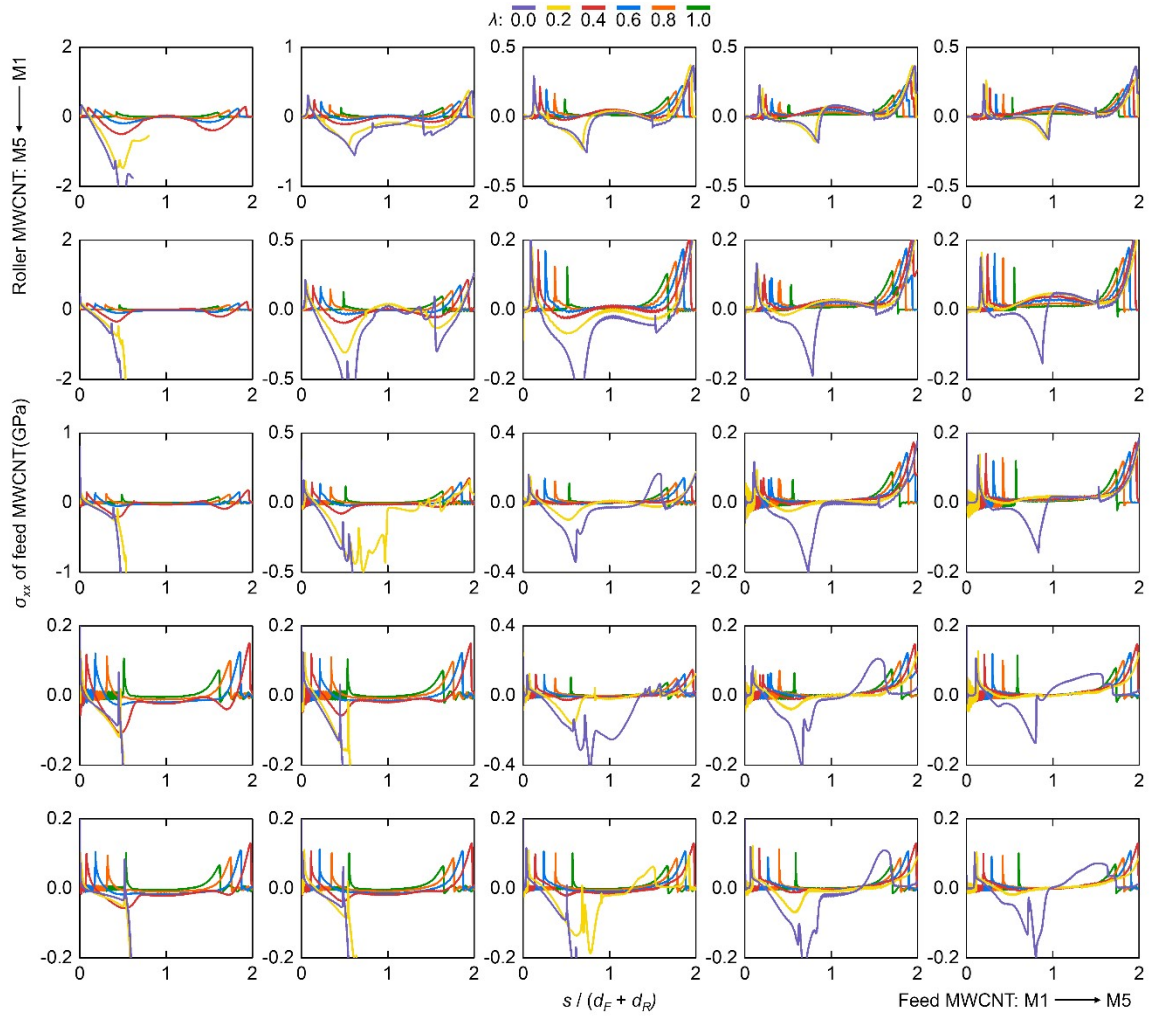

Fig. S10. Stress profiles of feed MWCNTs in x-direction.

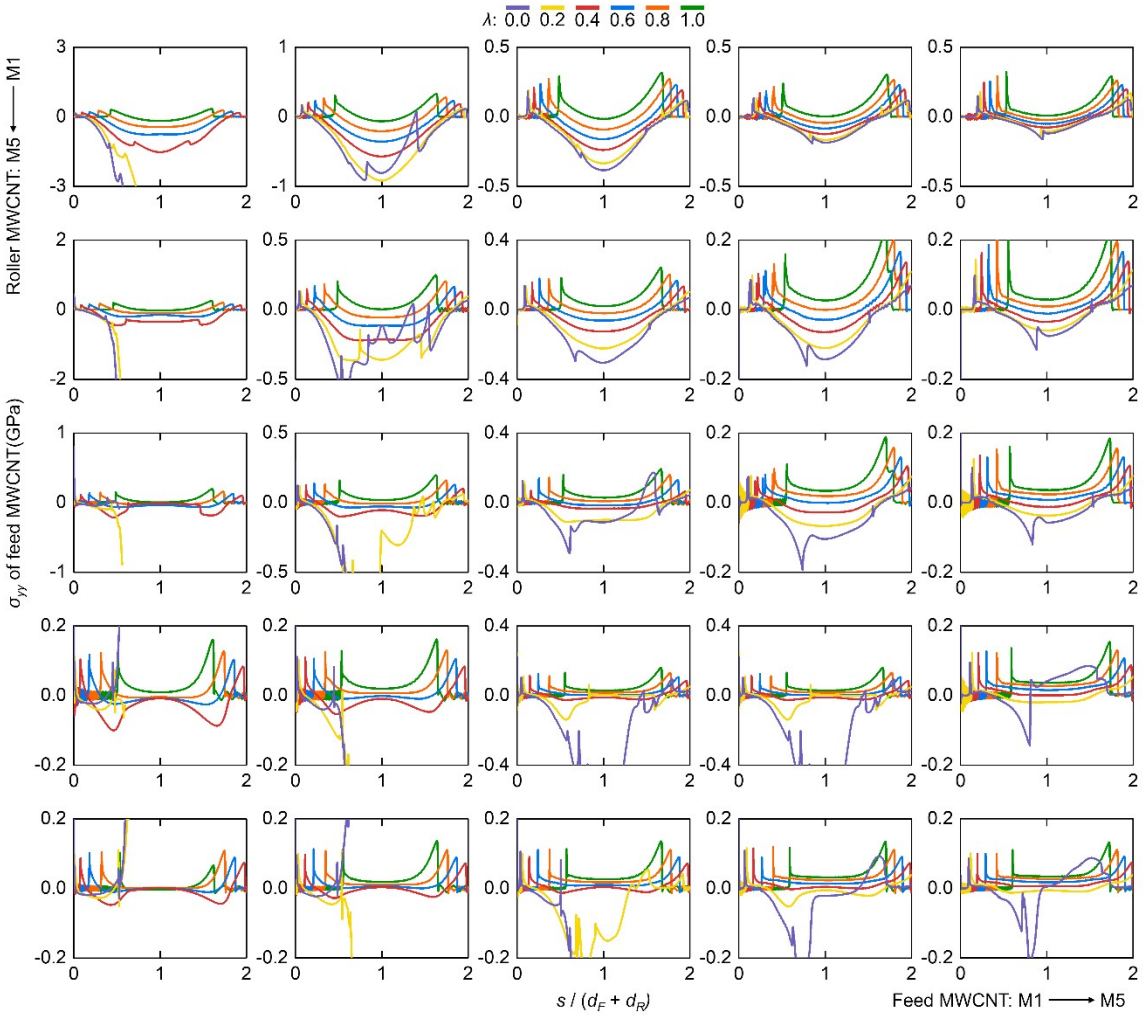

Fig. S11. Stress profiles of feed MWCNTs in y-direction.

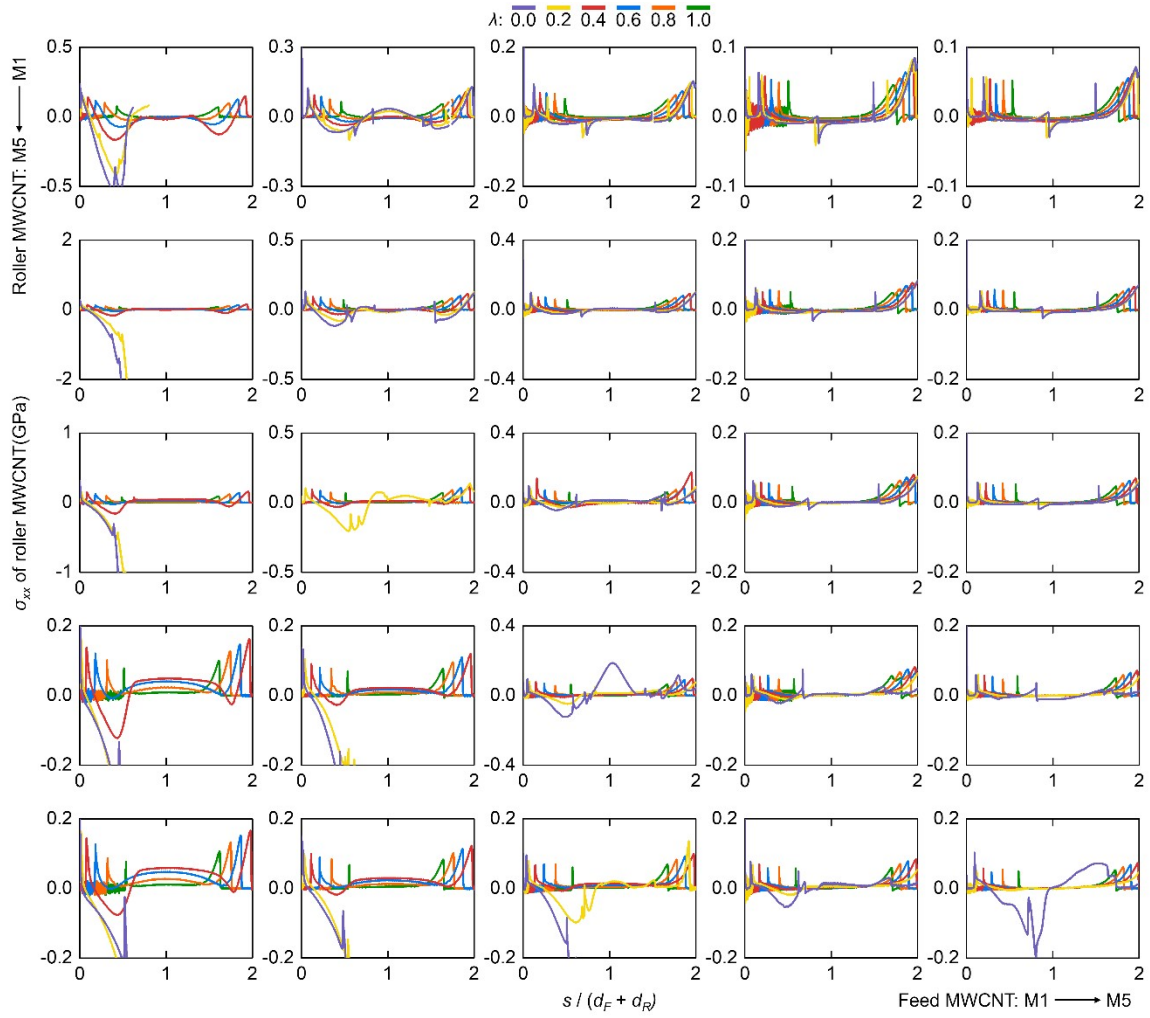

Fig. S12. Stress profiles of roller MWCNTs in x-direction.

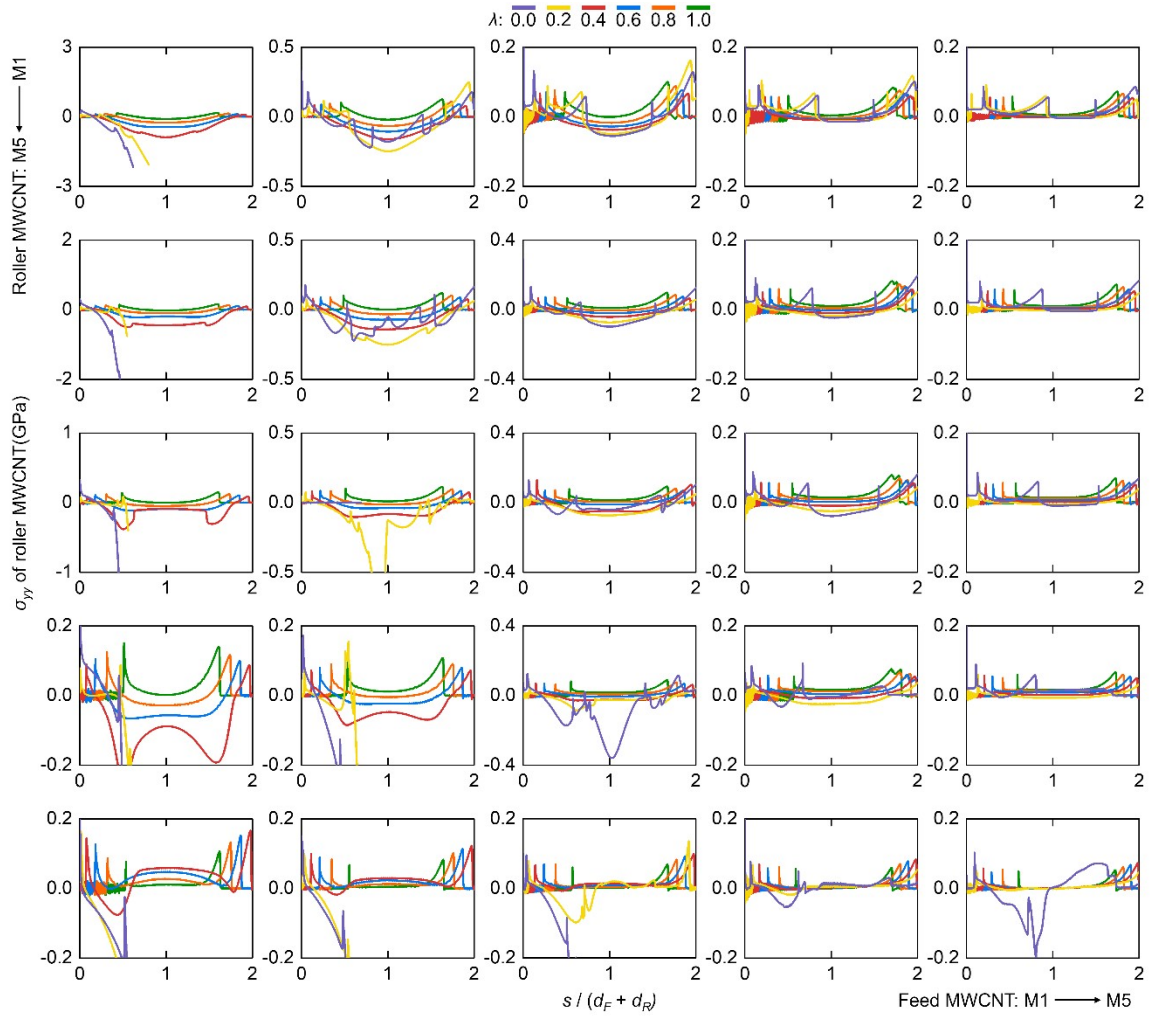

Fig. S13. Stress profiles of roller MWCNTs in y-direction.

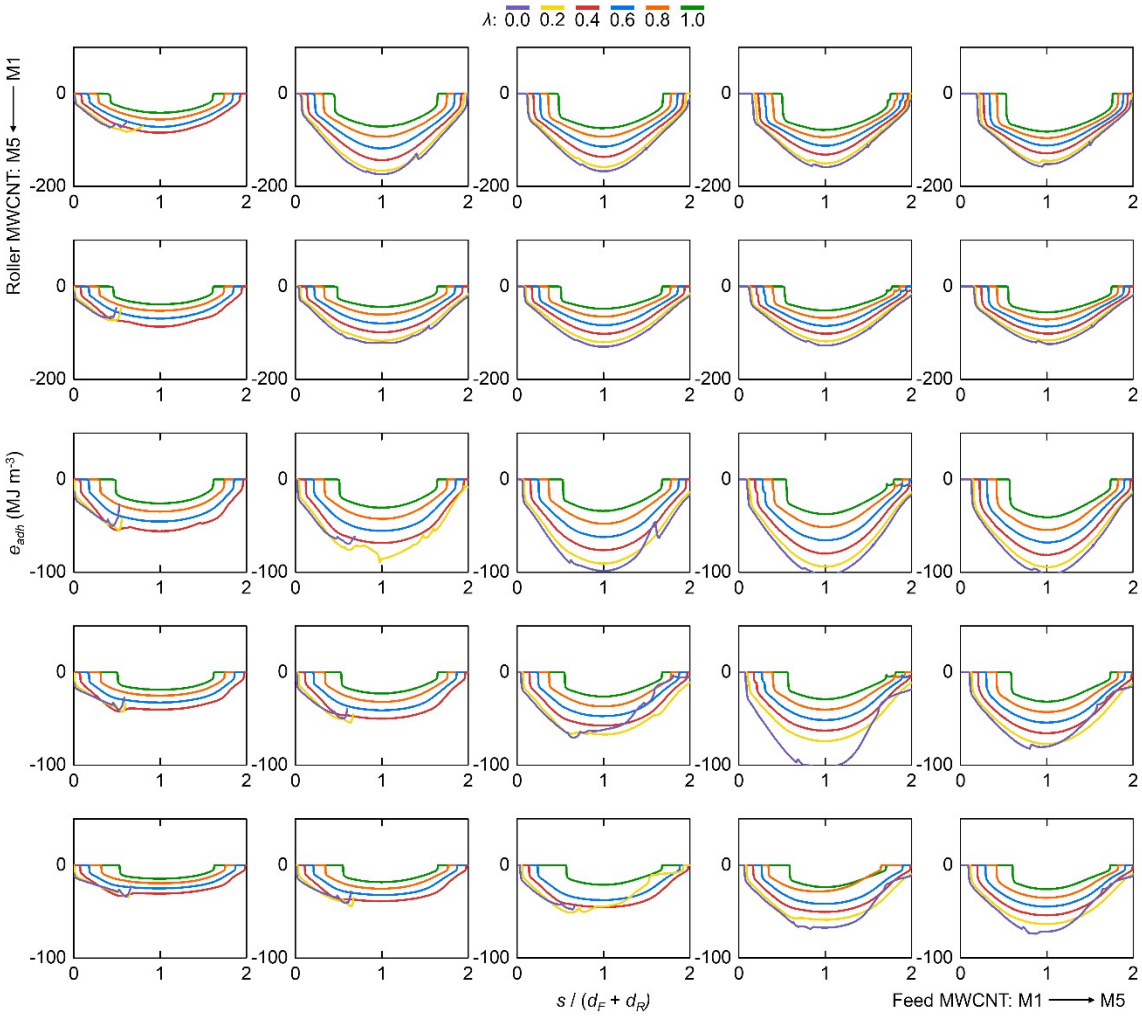

Fig. S14. AED profiles between feed and roller MWCNTs.

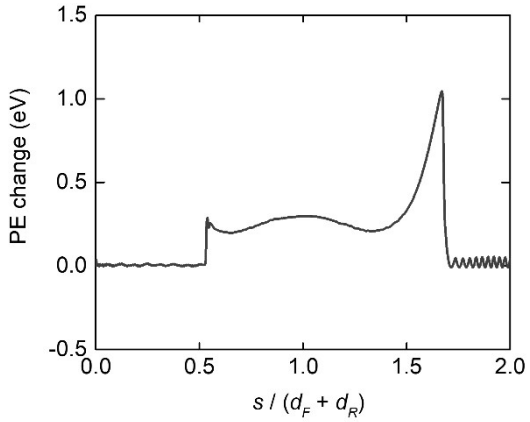

Fig. S15. Change in internal potential energy of the sole M3 MWCNT feed while passing through M3 MWCNT rollers.

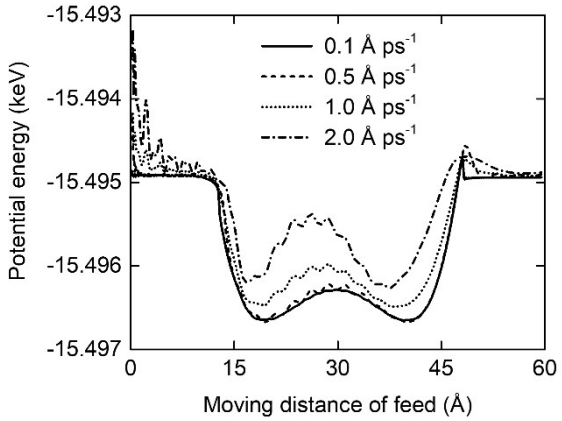

Fig. S16. Potential energy profiles of the (1,1,1.0) model with different feed moving speeds. The statistics behavior of the MWCNTs is saturated under 0.1 Å ps^{-1} of the moving speed.

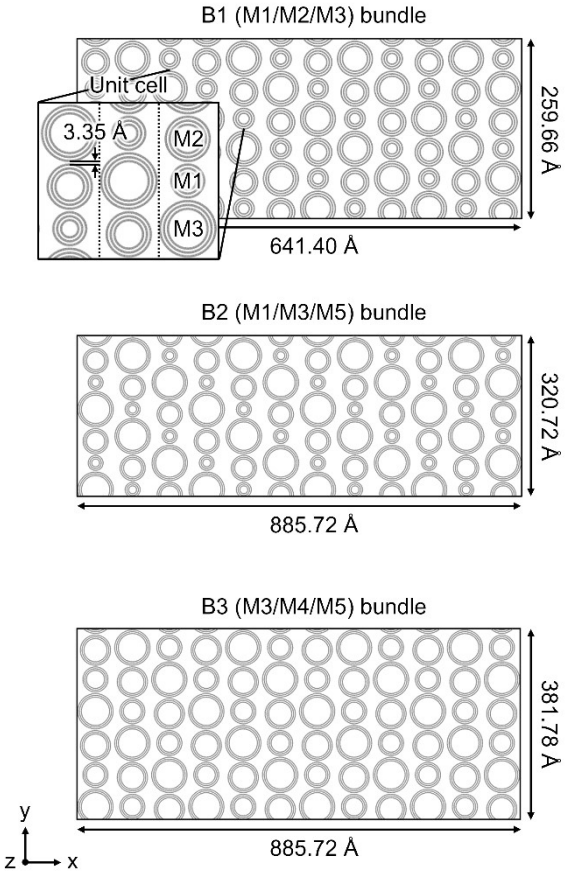

Fig. S17. Initial configuration of MWCNT bundle model.

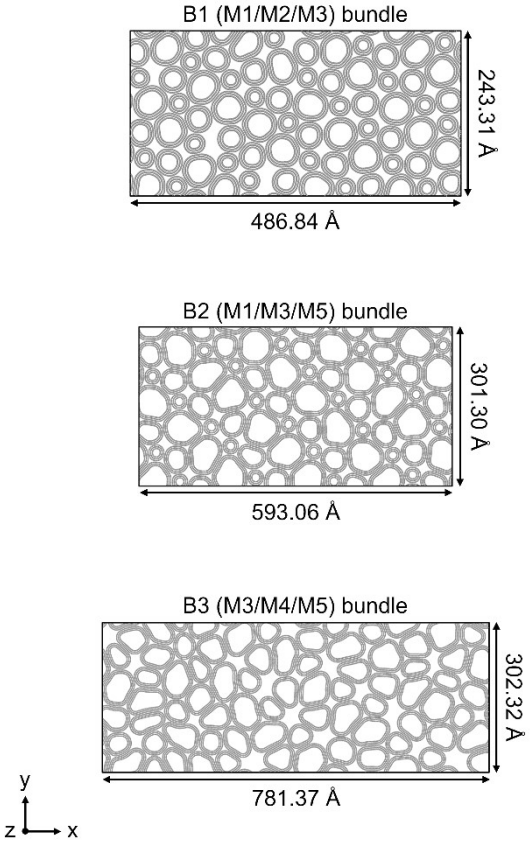

Fig. S18. Configuration after thermal equilibrium of MWCNT bundle model.

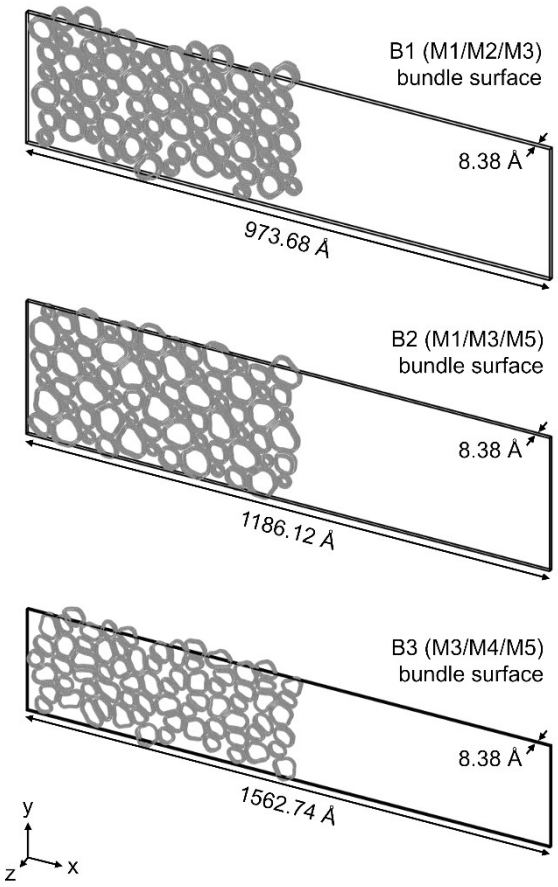

Fig. S19. Isometric view of the surface of the thermally equilibrated MWCNT bundle. The leftmost slab layer is fixed, while the remainder of the system is equilibrated.

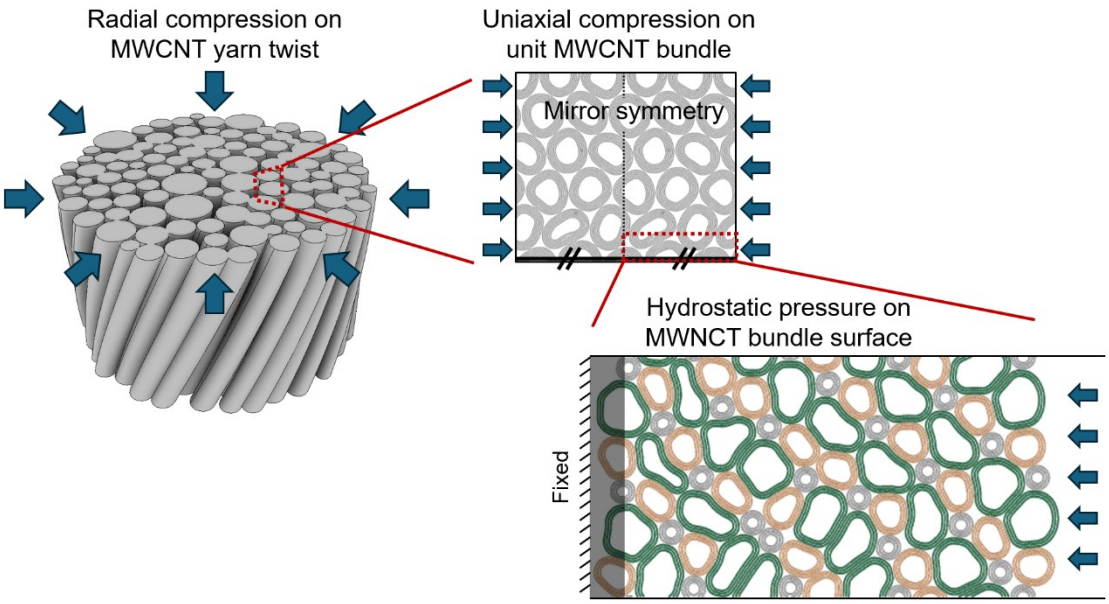

Fig. S20. Illustration of characterizing the deformation mode of the cross-sectional MWCNT bundles inside the twisted MWCNT yarn structure.

Table S1. All the energy components are calculated during the entry and exit of each object. All units, except for the model name, are MJ m^{-3}. The u_x and u_y components are expressed as absolute values to represent the sum of the energy levels required to escape. The energy levels of the fully clogged cases are denoted as n/a (not available).

Model	e_{adh}		Entry		Exit		Sum,	Sum,	Total
	Entry	Exit	u_x	u_y	u_x	u_y	Entry	Exit	
(1,1,1.0)	-21.66	26.84	11.54	23.45	10.83	27.03	13.33	-11.02	2.31
(1,1,0.8)	-35.54	42.67	18.7	34.03	15.83	34.82	17.19	-7.98	9.21
(1,1,0.6)	-51.85	57.83	43.79	58.94	37.08	60.79	50.88	-40.04	10.84
(1,1,0.4)	-63.21	69.07	134.74	299.29	114.73	308.8	370.82	-354.46	16.36
(1,1,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(1,1,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(1,2,1.0)	-33.36	56.93	5.63	17.19	4.8	12.37	-10.54	39.76	29.22
(1,2,0.8)	-56.44	66.9	5.62	24.85	4.75	21.58	-25.97	40.56	14.59
(1,2,0.6)	-80.23	91.77	5.81	49.68	4.68	50.4	-24.74	36.69	11.95
(1,2,0.4)	-104.56	117.68	14.1	98.05	10.76	95.35	7.59	11.57	19.16
(1,2,0.2)	-127.95	138.24	59.38	188.33	41.59	160.44	119.76	-63.79	55.97
(1,2,0.0)	-134.07	152.07	88.95	265.84	59.84	135.13	220.72	-42.91	177.81
(1,3,1.0)	-25.19	53.72	5.59	16.93	4.26	8.74	-2.67	40.72	38.05
(1,3,0.8)	-40.62	69.08	5.95	12.82	4.83	9.02	-21.86	55.23	33.37
(1,3,0.6)	-65.13	87.58	4.37	16.44	3.97	17.07	-44.32	66.54	22.21
(1,3,0.4)	-81.13	107.76	4.69	25.76	4.42	27.64	-50.69	75.7	25.01
(1,3,0.2)	-111.05	131.51	15.54	44.35	29.32	45.34	-51.16	56.86	5.7
(1,3,0.0)	-107.18	139.48	20.72	50	17.93	47.11	-36.46	74.44	37.98
(1,4,1.0)	-20.08	51.61	3.53	13.39	5.25	7.87	-3.16	38.49	35.33
(1,4,0.8)	-32.66	65.93	4.4	10.06	5.09	6.76	-18.19	54.08	35.88
(1,4,0.6)	-50.91	85.71	4.24	9.67	4.41	10.54	-37	70.75	33.75
(1,4,0.4)	-74.87	104.04	4.06	10.81	5.27	15.33	-60	83.44	23.45
(1,4,0.2)	-86.65	121.48	11.44	21.03	19.27	19.94	-54.18	82.27	28.09
(1,4,0.0)	-96.74	126.33	11.67	22.69	17.1	20.55	-62.39	88.67	26.28
(1,5,1.0)	-15.41	56.67	5.71	20.28	5.99	6.81	10.58	43.87	54.45
(1,5,0.8)	-33.18	69.59	6.91	12.96	5.47	6	-13.32	58.12	44.8
(1,5,0.6)	-42.91	83.83	6.2	9.99	4.59	8.33	-26.72	70.91	44.19
(1,5,0.4)	-60.54	102.37	5.52	8.35	5.14	11.09	-46.68	86.14	39.46
(1,5,0.2)	-82.85	123.14	8.8	14.71	8.15	14.76	-59.33	100.23	40.89
(1,5,0.0)	-92.95	128.53	8.32	13.98	18.21	17.65	-70.64	92.67	22.03
(2,1,1.0)	-18.23	24.67	6.21	16.78	12.3	18.28	4.76	-5.9	-1.14
(2,1,0.8)	-32.6	37.17	3.9	15.49	44.19	79.46	-13.2	-86.49	-99.69
(2,1,0.6)	-50.18	56.32	15.01	43.49	53.93	85.15	8.32	-82.75	-74.44
(2,1,0.4)	-68.66	75.47	35.78	91.97	134.8	193.02	59.08	-252.36	-193.28
(2,1,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(2,1,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(2,2,1.0)	-17.03	30.48	12.61	13.9	26.02	35.88	9.48	-31.42	-21.94
(2,2,0.8)	-33.82	46.54	12.17	8.76	17.82	16.84	-12.89	11.89	-1
(2,2,0.6)	-55.99	64.25	24.46	9.81	17.05	11.13	-21.72	36.07	14.35
(2,2,0.4)	-73.9	82.91	44.39	20.64	55.5	31.51	-8.87	-4.09	-12.96
(2,2,0.2)	-90.62	100.38	130.32	218.9	85.28	170.36	258.6	-155.26	103.34
(2,2,0.0)	-91.28	101.03	248.82	330.82	115.77	357.05	488.36	-371.79	116.58
(2,3,1.0)	-17.22	33.07	5.12	15.26	5.86	11.05	3.15	16.16	19.31
(2,3,0.8)	-33.92	49.98	1.17	5.03	15.17	21.68	-27.72	13.13	-14.59
(2,3,0.6)	-50.85	67.27	1.11	6.75	19.56	24.96	-42.99	22.75	-20.23
(2,3,0.4)	-67.96	86.57	3.6	16.65	23.67	28.33	-47.71	34.57	-13.14
(2,3,0.2)	-86.54	103.98	10.52	29.8	25.33	33.64	-46.22	45.01	-1.21
(2,3,0.0)	-95.41	110.15	25.56	40.16	30.52	37.37	-29.69	42.26	12.57
(2,4,1.0)	-14.04	35.4	3.52	12.6	6.89	12.22	2.08	16.29	18.37
(2,4,0.8)	-30	52.02	1.73	5.06	13.13	15.38	-23.22	23.51	0.3
(2,4,0.6)	-49.22	69.03	1.92	3.14	11.02	11	-44.16	47.01	2.85

(2,4,0.4)	-62.87	85.22	1.58	7.22	9.76	10.78	-54.07	64.68	10.6
(2,4,0.2)	-79.98	101.01	2.47	13.22	8.04	12.29	-64.29	80.68	16.39
(2,4,0.0)	-88.54	107.76	10.17	16.14	7.32	13.92	-62.24	86.52	24.28
(2,5,1.0)	-22.43	41.66	4.95	16.84	5.16	9.57	-0.64	26.92	26.29
(2,5,0.8)	-38.89	55.32	2.21	5.87	12.2	14.81	-30.81	28.31	-2.5
(2,5,0.6)	-55.53	60.22	2.91	2.83	9.73	8.43	-49.79	42.06	-7.73
(2,5,0.4)	-70.48	85.34	3.74	3.1	7.96	6.48	-63.63	70.9	7.28
(2,5,0.2)	-82.7	98.94	3.69	6.73	6.98	7.05	-72.27	84.92	12.64
(2,5,0.0)	-83.76	105.62	4.38	8.4	6.81	7.86	-70.99	90.94	19.95
(3,1,1.0)	-11.89	18.27	6.62	13.86	13.48	17.29	8.6	-12.5	-3.9
(3,1,0.8)	-21.35	27.39	6.68	12.56	8.34	11.25	-2.11	7.8	5.7
(3,1,0.6)	-33	36.85	11.29	25.68	7.99	24.85	3.97	4.01	7.98
(3,1,0.4)	-42.37	46.38	59.96	93.12	31.59	71.87	110.71	-57.08	53.63
(3,1,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(3,1,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(3,2,1.0)	-62.28	22.01	5.24	13.37	13.63	18.26	-43.68	-9.88	-53.55
(3,2,0.8)	-24.75	32.43	5.34	9.09	8.91	10	-10.32	13.52	3.2
(3,2,0.6)	-38.43	45.13	6.47	14.43	6.35	16.17	-17.53	22.61	5.07
(3,2,0.4)	-49.12	57.42	20.29	47.38	16.68	46.66	18.55	-5.92	12.63
(3,2,0.2)	-69.89	76.1	223.96	489.64	32.09	79.13	643.72	-35.13	608.59
(3,2,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(3,3,1.0)	-10.88	25.17	12.02	12.57	33.56	40.43	13.7	-48.82	-35.12
(3,3,0.8)	-27.64	40.13	8.9	5.24	20.75	17.14	-13.5	2.24	-11.26
(3,3,0.6)	-42.38	50.44	8.7	4.41	7.17	6.79	-29.26	36.48	7.22
(3,3,0.4)	-58.18	65.07	10.25	8.93	19.88	16.62	-39.01	28.56	-10.45
(3,3,0.2)	-67.55	76.88	63.49	20.28	76.08	20.57	16.22	-19.77	-3.55
(3,3,0.0)	-74.93	81.33	76.89	47.96	57.05	42.52	49.92	-18.24	31.68
(3,4,1.0)	-11.03	32.38	3.93	9.22	8.28	11.35	2.13	12.75	14.87
(3,4,0.8)	-25.16	47	3.49	8.16	8.25	8.57	-13.51	30.18	16.67
(3,4,0.6)	-38.87	60.92	3.43	4.28	8.3	7.67	-31.16	44.95	13.79
(3,4,0.4)	-53.98	69.75	3.35	9.14	11.98	14.16	-41.49	43.62	2.13
(3,4,0.2)	-64.22	80.23	8.15	21.8	10.43	22.08	-34.26	47.71	13.45
(3,4,0.0)	-75.12	86.65	45.17	51.88	15.67	30.98	21.93	40	61.93
(3,5,1.0)	-12.42	30.36	5.03	12.88	5.55	10.08	5.48	14.73	20.21
(3,5,0.8)	-24.64	43.71	5.07	11.21	4.58	9.77	-8.36	29.36	21
(3,5,0.6)	-44.2	57.52	5.38	8.35	6.62	10.34	-30.47	40.56	10.09
(3,5,0.4)	-57.13	70.69	6.15	7.56	12.58	12.5	-43.42	45.62	2.2
(3,5,0.2)	-68.86	80.35	7.98	12.11	13.37	14.94	-48.77	52.04	3.27
(3,5,0.0)	-73.39	86.99	31.57	30.67	18.19	17.48	-11.15	51.32	40.17
(4,1,1.0)	-4.93	11.67	5.71	10.59	12.06	14.36	11.36	-14.76	-3.39
(4,1,0.8)	-14.41	18.14	5.67	7.44	6.9	5.85	-1.29	5.39	4.09
(4,1,0.6)	-22.33	26.49	8.47	11.22	5.6	11.63	-2.65	9.25	6.61
(4,1,0.4)	-30.77	32.75	39.71	43.34	18.79	38.1	52.29	-24.14	28.14
(4,1,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(4,1,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(4,2,1.0)	-8.31	16.69	4.95	10.79	11.99	15.21	7.43	-10.51	-3.08
(4,2,0.8)	-19.55	25.27	4.82	8.04	6.44	7.64	-6.7	11.19	4.5
(4,2,0.6)	-27.82	33.32	5.52	6.62	5.27	7.83	-15.69	20.22	4.53
(4,2,0.4)	-38.71	41.11	16.66	24.14	13.01	25.09	2.1	3.02	5.11
(4,2,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(4,2,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(4,3,1.0)	-8.48	19.62	3.85	10.12	9.57	13.05	5.48	-3	2.48
(4,3,0.8)	-20.81	29.11	4.02	9.31	5.47	9.14	-7.48	14.51	7.02
(4,3,0.6)	-30.53	39.18	4.41	5.81	5.11	7.92	-20.31	26.14	5.83
(4,3,0.4)	-41.11	50.76	9.06	13.27	10.47	16.93	-18.79	23.36	4.58
(4,3,0.2)	-49.52	55.74	54.24	62.73	10.27	23.32	67.45	22.15	89.59
(4,3,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(4,4,1.0)	-10.61	22.41	15.41	13.9	24.98	31.91	18.71	-34.49	-15.78
(4,4,0.8)	-22.93	33.39	11.21	8.21	15.98	11.6	-3.5	5.81	2.31

(4,4,0.6)	-39.67	44.98	10.04	6.48	6.94	6.57	-23.15	31.47	8.32
(4,4,0.4)	-51.09	55.56	16.43	8.03	22.62	18.25	-26.62	14.68	-11.94
(4,4,0.2)	-63.06	62.93	46.91	15.79	55.61	11.92	-0.36	-4.6	-4.95
(4,4,0.0)	-74.7	84.36	39.18	30.23	12.97	13.67	-5.29	57.72	52.43
(4,5,1.0)	-11.01	24.75	4.53	11.44	6.29	9.97	4.96	8.49	13.45
(4,5,0.8)	-25.8	35.22	4.73	12.1	3.87	9.77	-8.97	21.57	12.61
(4,5,0.6)	-38.68	42.76	4.47	10.73	5.61	11.78	-23.48	25.37	1.89
(4,5,0.4)	-49.92	48.76	4.26	7.34	11.95	11.99	-38.32	24.83	-13.49
(4,5,0.2)	-57.81	53.3	6.52	12.33	9.84	12.22	-38.95	31.23	-7.72
(4,5,0.0)	-64.94	61.88	29.87	21.05	5.5	7.58	-14.02	48.8	34.78
(5,1,1.0)	-5.04	9.31	2.65	2.64	15.65	11.08	0.25	-17.42	-17.16
(5,1,0.8)	-10.81	14.39	2.35	4.22	11.78	10.99	-4.24	-8.38	-12.62
(5,1,0.6)	-17.29	19.72	5.18	4.41	13.96	12.59	-7.69	-6.83	-14.52
(5,1,0.4)	-23.65	25.03	25.03	17.99	11	19.23	19.37	-5.2	14.17
(5,1,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,1,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,2,1.0)	-6.78	13.1	2.3	6.01	17.12	21.06	1.53	-25.08	-23.54
(5,2,0.8)	-17.87	24.24	2.03	6.52	13.16	14.85	-9.33	-3.77	-13.1
(5,2,0.6)	-22.13	26.54	2.93	3.47	13.07	12.5	-15.73	0.98	-14.75
(5,2,0.4)	-27.83	33.64	10.97	10.51	11.77	15.57	-6.35	6.3	-0.05
(5,2,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,2,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,3,1.0)	-7.66	16.3	2.46	6.71	12.4	14.51	1.51	-10.61	-9.1
(5,3,0.8)	-15.2	22.89	0.72	5.21	33.42	48.02	-9.27	-58.55	-67.82
(5,3,0.6)	-26.54	31.73	2.69	6.29	7.83	11.17	-17.56	12.73	-4.83
(5,3,0.4)	-33.22	39.7	6.71	6.49	11.66	12.96	-20.02	15.08	-4.93
(5,3,0.2)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,3,0.0)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
(5,4,1.0)	-6.9	18.9	2.47	7.1	11.1	13.73	2.67	-5.93	-3.26
(5,4,0.8)	-19.47	26.99	2.05	9.54	8.05	12.63	-7.89	6.32	-1.57
(5,4,0.6)	-29.62	35.8	2.57	8.26	8.63	13.47	-18.79	13.7	-5.09
(5,4,0.4)	-37.68	44.86	4.79	4.54	11.15	11.32	-28.35	22.39	-5.95
(5,4,0.2)	-43.98	49.26	23.32	23.71	6.66	12.11	3.05	30.5	33.55
(5,4,0.0)	-48.73	55.84	78.38	99.84	11.18	8.18	129.5	36.48	165.97
(5,5,1.0)	-7.26	20.62	2.7	8.84	5.62	9.06	4.27	5.93	10.21
(5,5,0.8)	-18.67	29.91	2.74	9.47	3.6	9.64	-6.46	16.67	10.2
(5,5,0.6)	-29.92	39.02	2.65	9.78	4.71	12.45	-17.5	21.86	4.36
(5,5,0.4)	-41.06	48.56	3.89	6.9	10.49	12.67	-30.27	25.39	-4.88
(5,5,0.2)	-49.91	53.85	7.84	9.34	8.71	8.41	-32.72	36.73	4.01
(5,5,0.0)	-57.01	61.83	64.02	45.04	10.38	13.3	52.06	38.15	90.21