Supplementary Information file for

Developing Nanotube Junctions with Arbitrary Specifications

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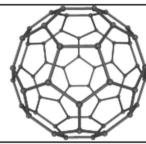
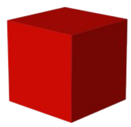
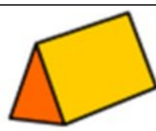
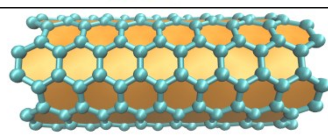
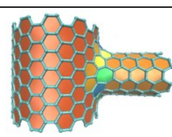
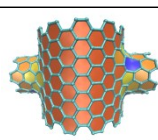
Supplementary Information S1:
Several examples showcasing the validation of Euler's law of polyhedra

Structure	Schematic	Vertices (V)	Faces (F)	Edges (E)	V+F-E	Bond Surplus -6 X (V+F-E)
C20		**20**	**12**	**30**	**2**	**-12**
C60		**60**	**32**	**90**	**2**	**-12**
Cube		**8**	**6**	**12**	**2**	**-12**
Triangular Prism		**6**	**5**	**9**	**2**	**-12**
hexagonal prism		**12**	**8**	**16**	**2**	**-12**
Triangular pyramid		**4**	**4**	**6**	**2**	**-12**
rectangular pyramid		**6**	**5**	**9**	**2**	**-12**
Open-ended CNT		**192**	**84**	**276**	**0**	**0**
T-junction (one of the nanotubes is fusing partially)		**430**	**184**	**615**	**-1**	**6**
Plus-junction (2 nanotubes fusing all the way across)		**412**	**168**	**582**	**-2**	**12**

Supplementary Information S2:

Schematics of different types of CNT junctions created by the discussed framework showcasing junctions with different degrees of CNT overlap, different orientations of CNTs, and junctions between different diameter CNTs

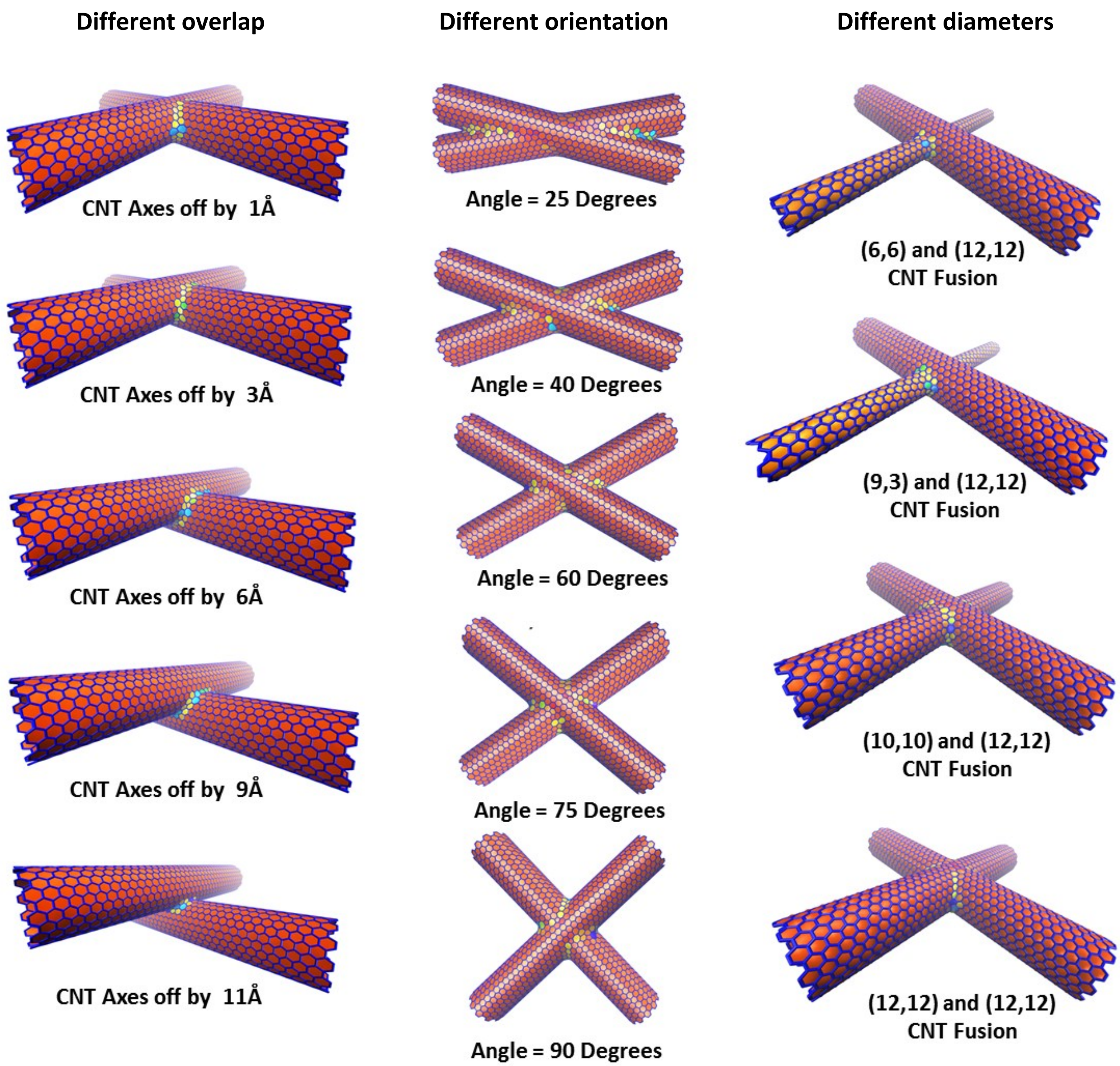


Figure S1: Schematics of different types of CNT junctions created by the discussed framework.

Supplementary Information S3:
Discussion on loop stitching

As discussed in the main text, there can be several possible realizations of open loop which is created during step 1e (see Figure 1e in the main text). Below, we discuss all possible independent cases and how such loops are closed to create completely triangulated junction structure. We discuss these examples using two independent rings of blue and purple color (for each case). These rings consist of surface nodes and surface edges (see main text discussion Steps 1(d) and 1(e)) which originate from the two nanotubes that are to be fused. This process is schematically shown in Figure S2.

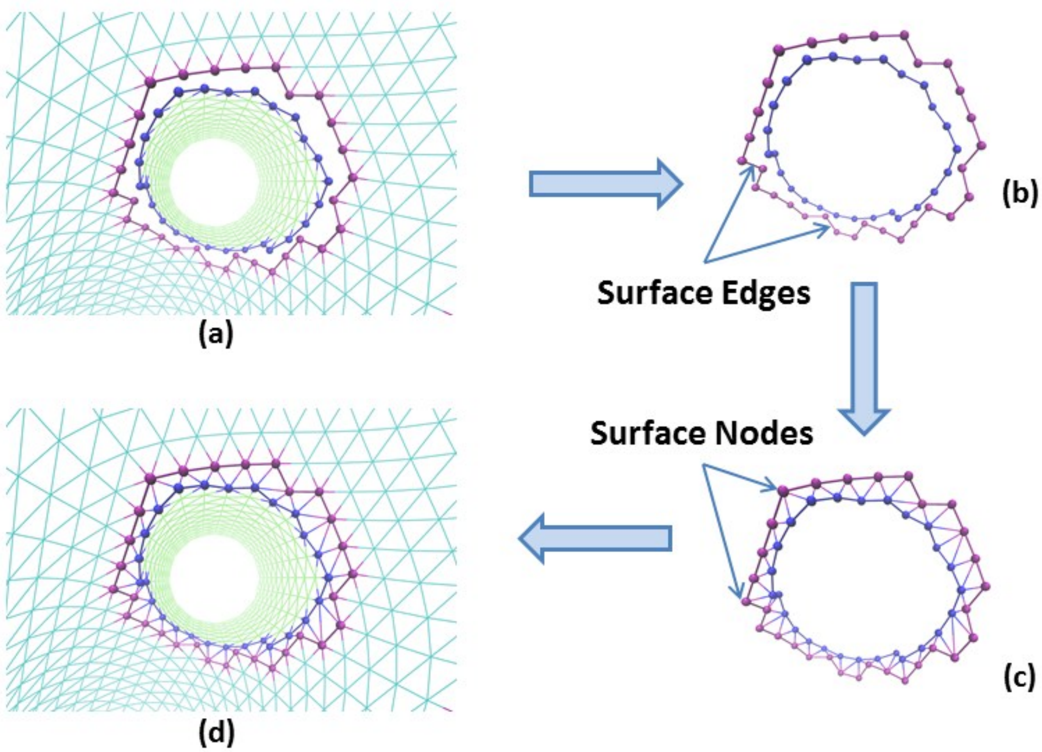


Figure S2: A generic process of loop stitching is shown. (a) two nanotubes (green and cyan color) create two loops (b) which are triangulated as seen in (c). (d) depicts the final structure. Surface edges and nodes are also shown for the clarity of discussion below.

No overlapping nodes:

When these surface rings (of surface nodes and surface edges) are formed, there is a possibility that after step 1d (in the main text), there are no nodes which are too close (within 1Å). In such a case, two neighboring rings are generated in which no nodes are overlapping as shown in Figure S3a. In order to stitch such a loop,

a) First, the nearest distance pair (one node from each ring) is found and is connected via an edge (Figure S3b). Once a connection is formed, both loops are connected via a bond (a new edge is created) so, it can be treated as one long loop consisting of two chains passing through the newly formed edge twice.

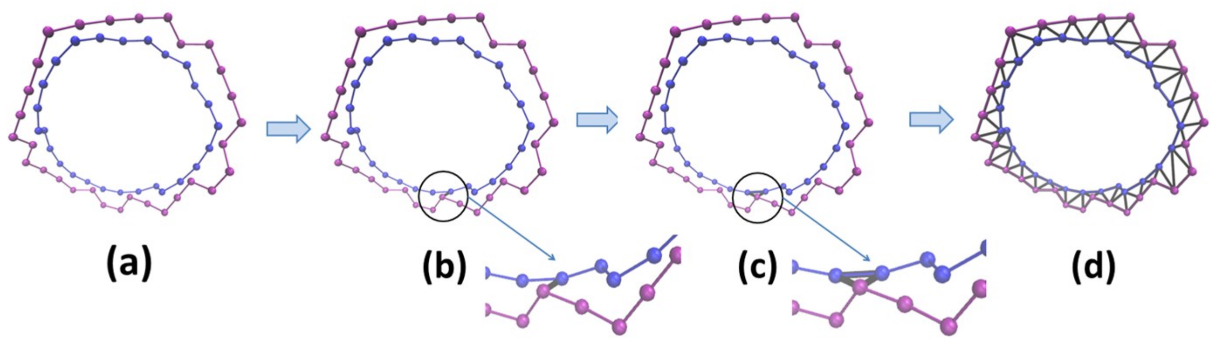


Figure S3: (a) Schematic of two rings where no nodes are fused. (b) Schematic showing connection of 1st edge (see zoom in sub-figure). (c) Schematic showing formation of 1st triangle (see zoom in sub-figure). (d) Final triangulated structure.

b) Then, all 1-3 pairs are searched (*1-3 pair is a pair where there is connected surface edge between 1st and 2nd node and an surface edge between 2nd and 3rd node, but no surface edge between 1st and 3rd node*) to identify all the possible triangles by connecting all possible pairs of 1st and 3rd node (*1-2 and 2-3 are already connected*) where all angles are acute angles. If more than one such possible triangle is found, a determination is made to identify the set of 1-3 pair such that the formed triangle is as close to equilateral triangle as possible. A new edge is thus created between the determined pair of 1st and 3rd node (See Figure S3c).

c) The open loop becomes smaller after for formation of the triangle. The list of open loop 1-3 pairs is then revised and step b) and step c) are iteratively followed to identify new edges to create best possible acute angle triangle until the whole loop is closed, entirely consisting entirely of triangles (see Figure S3d).

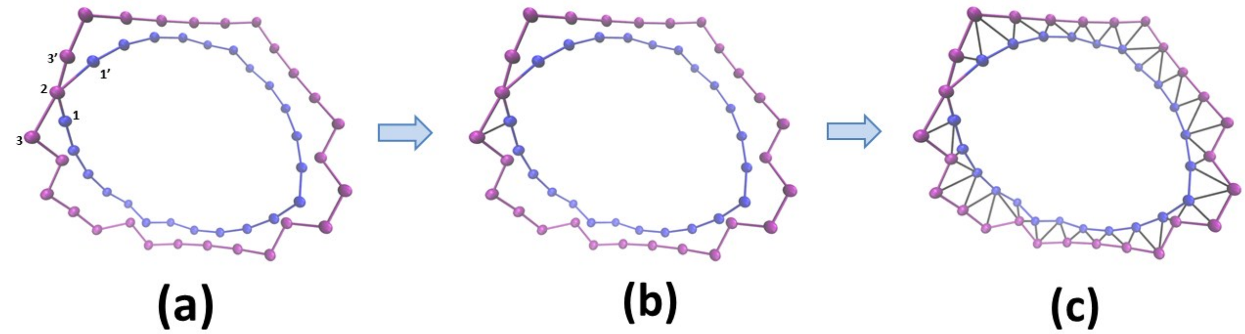


Figure S4: Schematic showing 1 overlapping case (a), creation of first edge and first triangle (b), and fully triangulated structure (c).

<u>1 overlapping node</u>

Figure S4a shows the case where only 1 node pair was observed to be close (within 1Å) and was fuzed into a single node. Let us call this node as node 2. In addition, let us assume that the nodes connected to 2 on blue chain are 1 and 1’ while the node connected to 2 on the purple chain are 3 and 3’. As node 2 has 4 surface edges, there are 4 possible ways a new edge can be formed to create a triangle.

a) Link between 1-3 creating a triangle between nodes 1-2-3
b) Link between 1-3’ creating a triangle between nodes 1-2-3’
c) Link between 1’-3 creating a triangle between nodes 1’-2-3
d) Link between 1’-3’ creating a triangle between nodes 1’-2-3’

Here, we analyze all 4 triangles and discard the ones which are obtuse in nature (2 of them). Then, one of the two remaining acute triangle (which most resembles the equilateral triangle) is created by connected a new edge as shown in the Figure S4b. Once these edges are formed, all the 1-3 pairs are identified and the subsequent stitching of the loop is carried out in the similar manner as discussed in step b) and step c) of ‘no overlapping nodes’ section above.

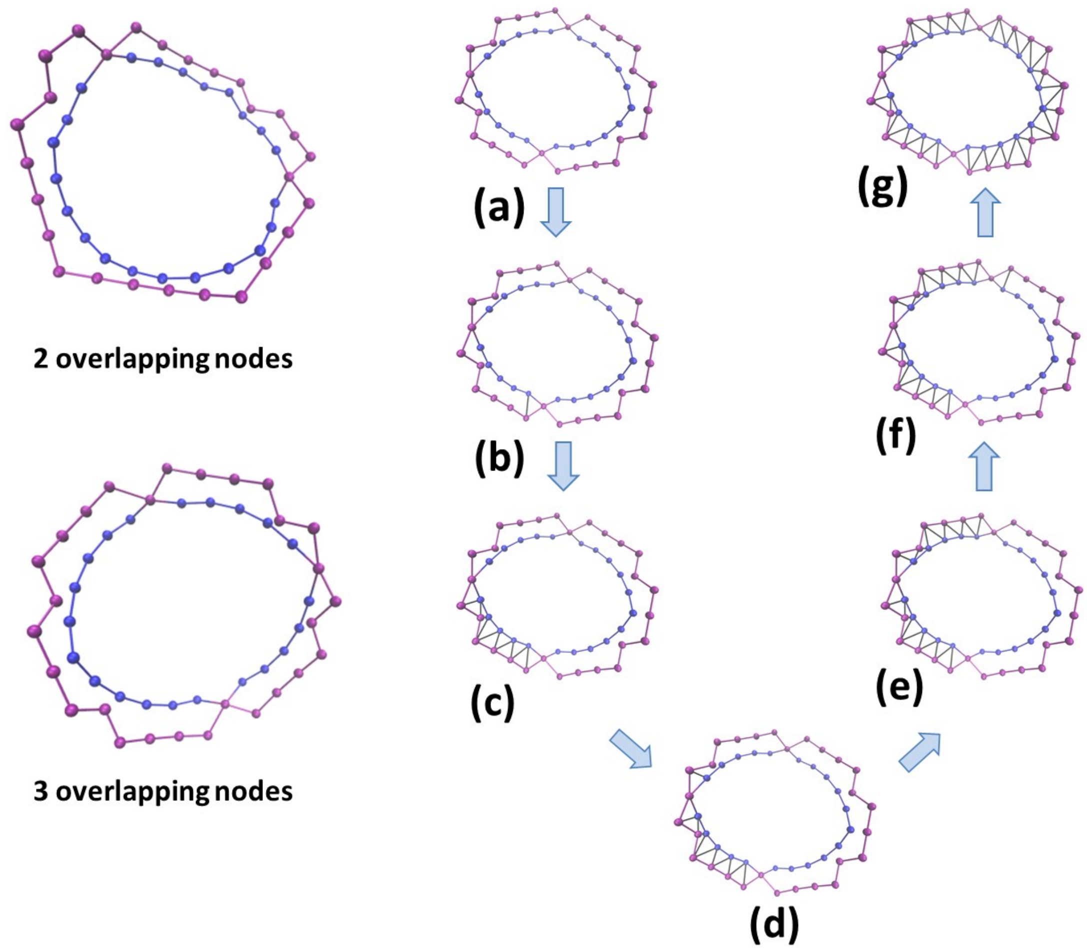


Figure S5: (Left) Schematics of 2 and 3 fused loop structures with no overlapping surfaces. (Right) Schematic process of how such structures are triangulated (see discussion) using the example of 3 overlapping node structure.

2 or more overlapping nodes with no overlapping surface edges

In Figure S5 (left), blue and purple rings from the two CNTs are shown having 2 and 3 fuzed nodes. This suggests that we have to respectively stitch 2 and 3 loops. If there were *n* such fused nodes, it will lead to stitching of *n* loops. First, one fused node (assume node A) among *n* (in general) is identified (Figure S5a). As this node has 4 surface edges, a similar methodology as discussed in "1 overlapping node" is used to identify possible 1-3 edges which will lead to acute triangle formation. The subsequent stitching of the loop which will terminate on the some other node (assume node B) as shown in see Figure S5c.

The node B, which originally had 4 surface edges, now only has 2 surface edges as the other two were used in the formation of the triangle using previous loop stitching. In such a case, only one possible edge can be formed to create a new triangle as shown in Figure S5d. We form that edge and follow previously discussed process of scanning all possible 1-3 pairs of that loop to determine next best acute triangle and stitch that loop (see Figure S5e). For *n* fused node cases, we repeat this process n-1 times to fully stitch blue and purple rings to create fully triangulated mesh (see Figure S5e-g).

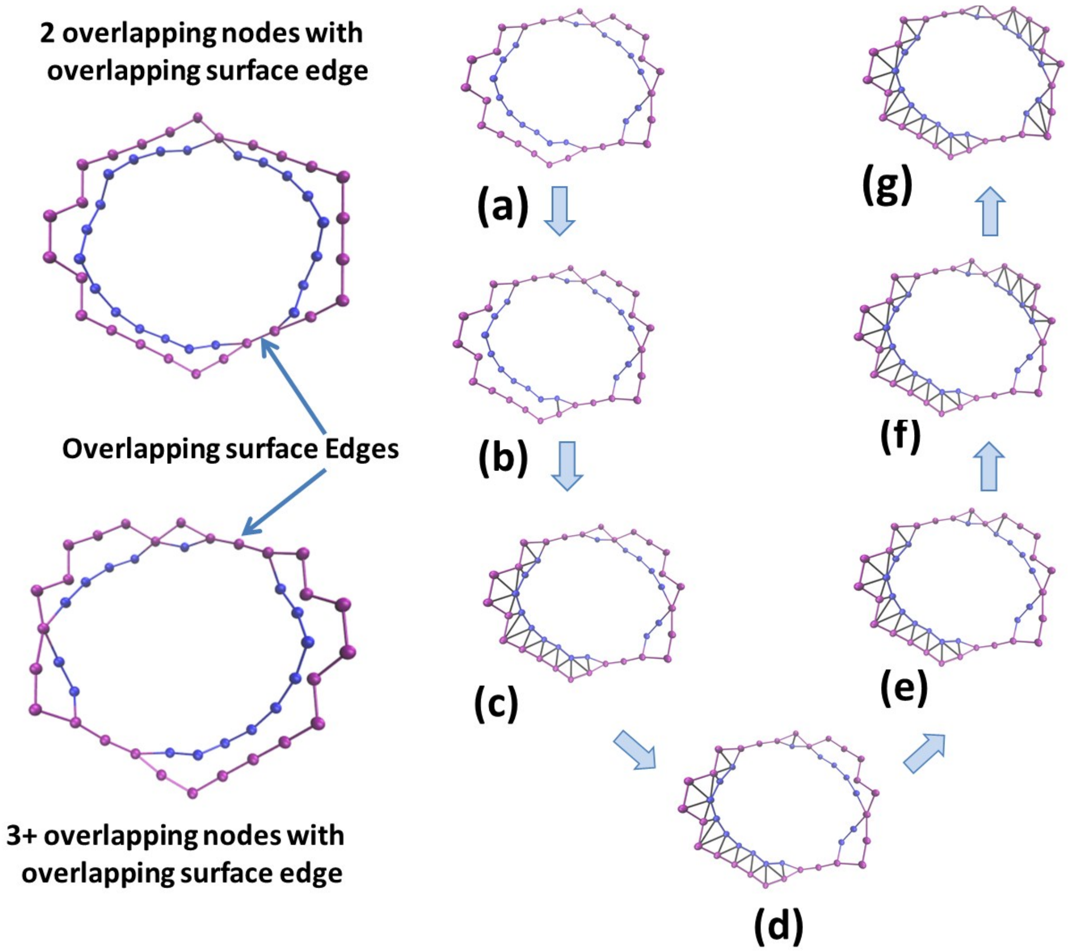


Figure S6: (Left) Schematics of 2 and 3+ fused loop structures with overlapping surface edges. (Right) Schematic process of how such structures are triangulated (see discussion) using the example of 3 overlapping node structure.

2 or more overlapping nodes with overlapping surfaces edges

Figure S6 show cases where two or more consecutive nodes are fused leading to creation of overlapping surface edges. Stitching these cases is relatively easier than previously discussed cases. Here, unlike previous discussed cases, there is no ambiguity to choose the direction with respect to starting edge node (only one triangle is possible from the starting edge node).

In order to fuse such loops, one of the nodes corresponding to overlapping surface edges is chosen to be the starting node (Figure S6b). As it has only 2 surface edges (one each on blue and purple ring), a triangle is formed by connecting the nearby 1-3 nodes. Thereafter, searching of 1-3 pairs of that particular loop (half loop is blue and half loop is purple) to identify possible edges which will result in best acute triangles as discussed in previous section to stitch that loop (Figure S6c). On completing first loop, stitching of remaining loops is done as discussed in previous sections (Figure S6d-g).

Supplementary Information S4:

Comparison of relative stability of 16 representative junctions after minimization using different carbon based force-fields using minimized energy/atom criterion

	Force Fields		
Case	**PCFF (kcal/mol)**	**Tersoff (eV)**	**AIREBO (eV)**
Ni00Nj00	46.54684	-7.14504	-7.19971
Ni00Nj01	46.33044	-7.15645	-7.20935
Ni00Nj10	46.78737	-7.12555	-7.18717
Ni00Nj11	46.22752	-7.17105	-7.21973
Ni01Nj00	46.36971	-7.16056	-7.20831
Ni01Nj01	46.57441	-7.14395	-7.19895
Ni01Nj10	46.80898	-7.12627	-7.18728
Ni01Nj11	46.23563	-7.17145	-7.21971
Ni10Nj00	46.4172	-7.16013	-7.20934
Ni10Nj01	46.40496	-7.16012	-7.20999
Ni10Nj10	46.04675	-7.18414	-7.22636
Ni10Nj11	46.4798	-7.15182	-7.20352
Ni11Nj00	46.3767	-7.15816	-7.20791
Ni11Nj01	46.36998	-7.15805	-7.20833
Ni11Nj10	46.54321	-7.14445	-7.19803
Ni11Nj11	46.43152	-7.147	-7.20255
Average	**46.43444**	**-7.15401**	**-7.20602**

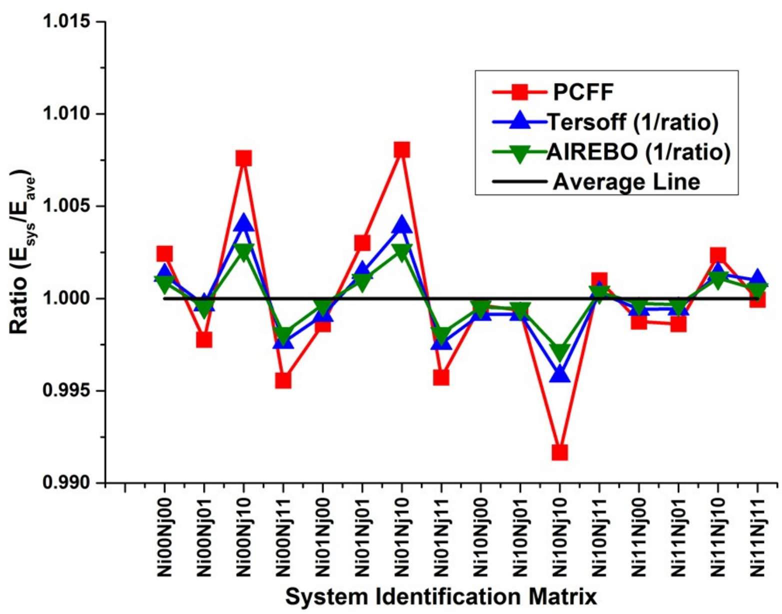


Figure S7: Plot showing relative stability of studied 16 junctions with respect to different force-fields.

In the table above, the differences in the absolute values in the force-fields are reflective of the force-field parameters used to model carbon. The –ve values in bond-order potentials (tersoff and AIREBO) are due to the fact that all interactions in bond-order potentials are non-bonded (no physical bond) interactions, where zero energy correspond to the case when the atoms are infinitely far apart.

In the above graph (Figure S7), we plot the ratio of each of the system minimized energy to its corresponding average for all studied force-fields. For PCFF force field, the lower the ratio, the higher the stability of the structure. For –ve values force-fields however, the case is vice-versa, i.e., the larger ratio will be more stable. Larger ratio value means that the numerator is more –ve then denominator (average value). To account for that, for Tersoff and AIREBO force-fields, we have plotted 1/ratio in order to observe the same consistency in the trends.

It is very clear from the plot that each of the force-fields shows similar behavior with respect to the relative stability of different generated cases. This further confirms that discussed results are independent for PCFF force-field as employed for analysis in the main text.

Supplementary Information S5:

Plot of energy/atom against the energy parameter D_{RSQ} for topological defect annihilation of 16 different studied cases.

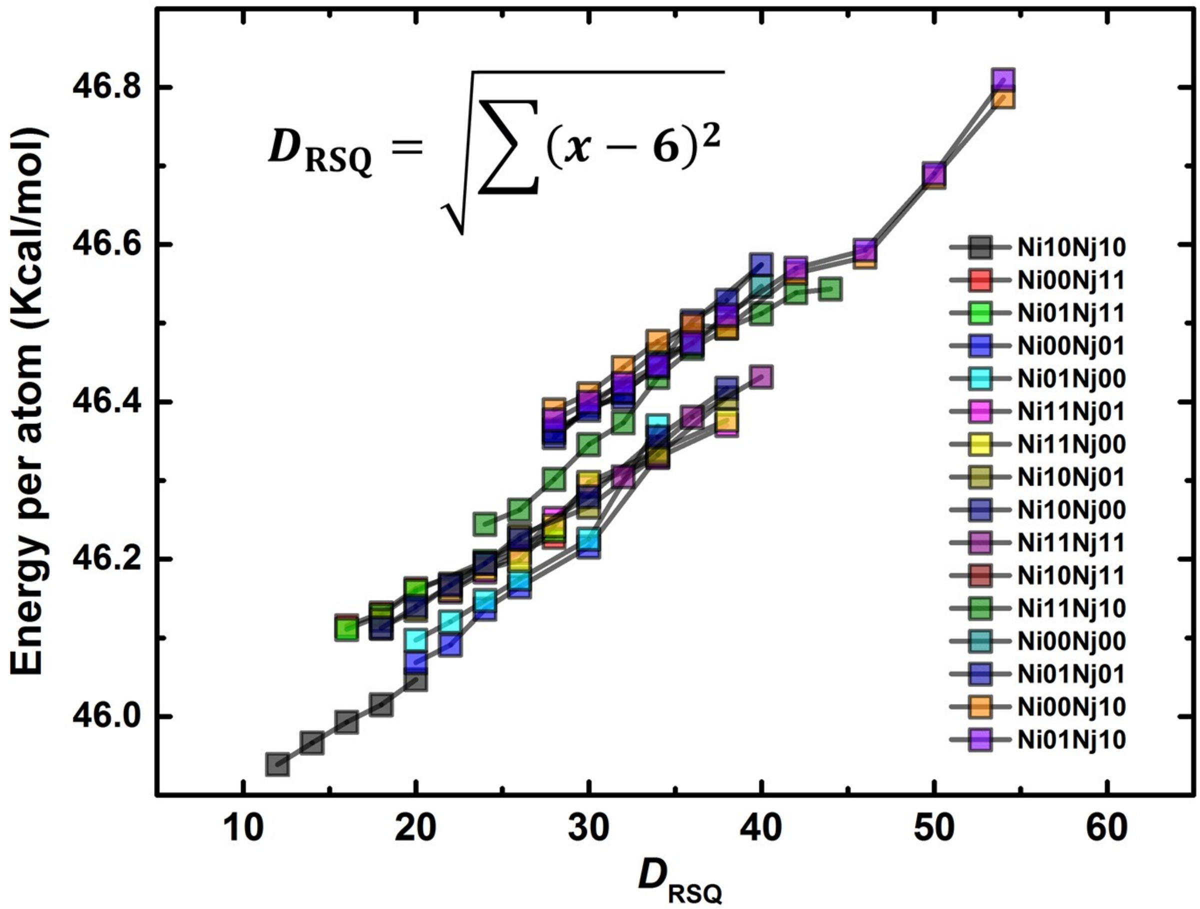


Figure S8: Plot showing energy/atom as a function of defect parameter D_{RSQ} for 16 different created systems (shown in different colors). The decrease in energy for each color with decreasing D_{RSQ} symbolizes the correlation between degree of junction defects (non-hexagonal rings) and stability of the structure in terms of energy/atom.