Electronic supplementary information

Image Learning to Accurately Identify Complex Mixture Components

Qiannan Duan, Jianchao Lee*, Jiayuan Chen, Yunjin Feng, Run Luo, Can Wang, Sifan Bi, Fenli Liu, Wenjing Wang, Yicai Huang, Zhaoyi Xu

Table of Contents

Figures and tables:

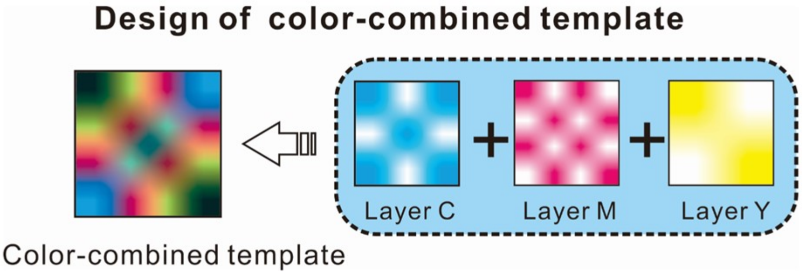


Figure S1 A combined template (in size of 1200×1200 dpi) was designed for preparing the multi-color wave filtering chip. The template consisted of three overlapping C/M/Y color layers (referring to inks: cyan, magenta and yellow), which were combined by presetting the CMY values in various directions.

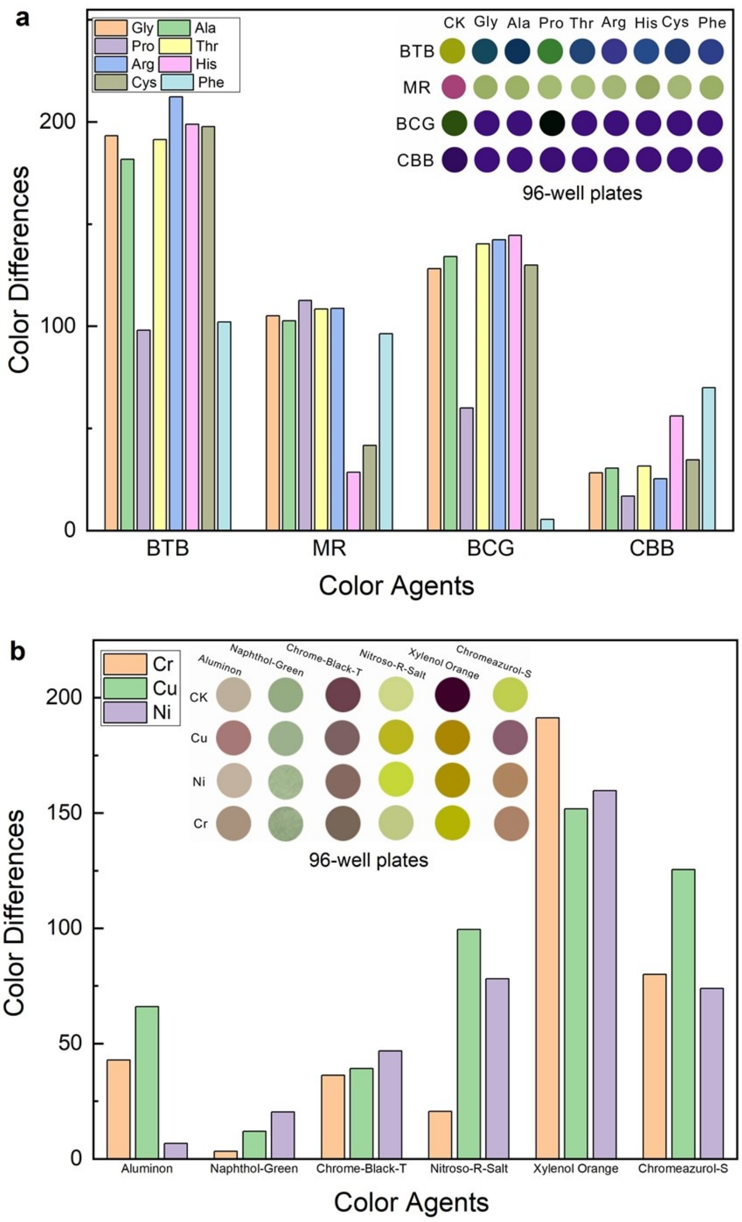


Figure S2 Color differences between the reaction groups and CK groups. The color differences of (a) the selected AA chromogenic agents and (b) the selected heavy metal chromogenic agents.

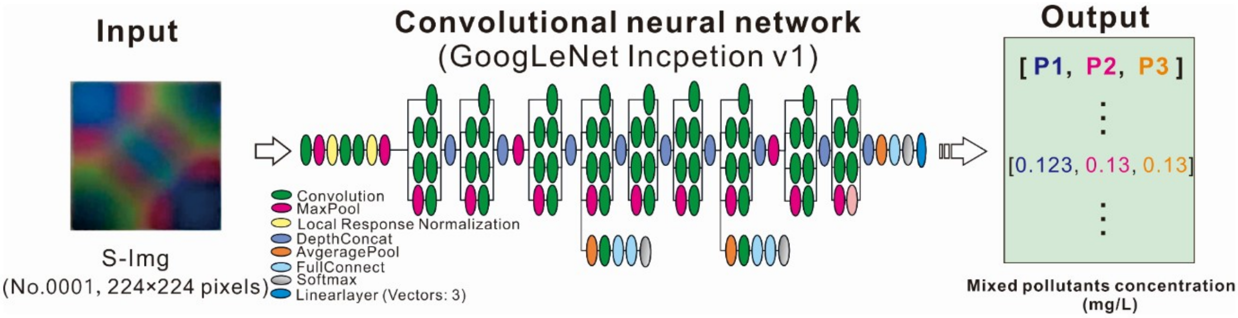


Figure S3 A modified Inception v1 framework for predicting mixture concentrations by inputting the S-Img. Detailed architectures were shown in Table S5, and reprinted from https://www.cv-foundation.org/openaccess/content_cvpr_2015/html/Szegedy_Going_Deeper_With_2015_CVPR_paper.html.

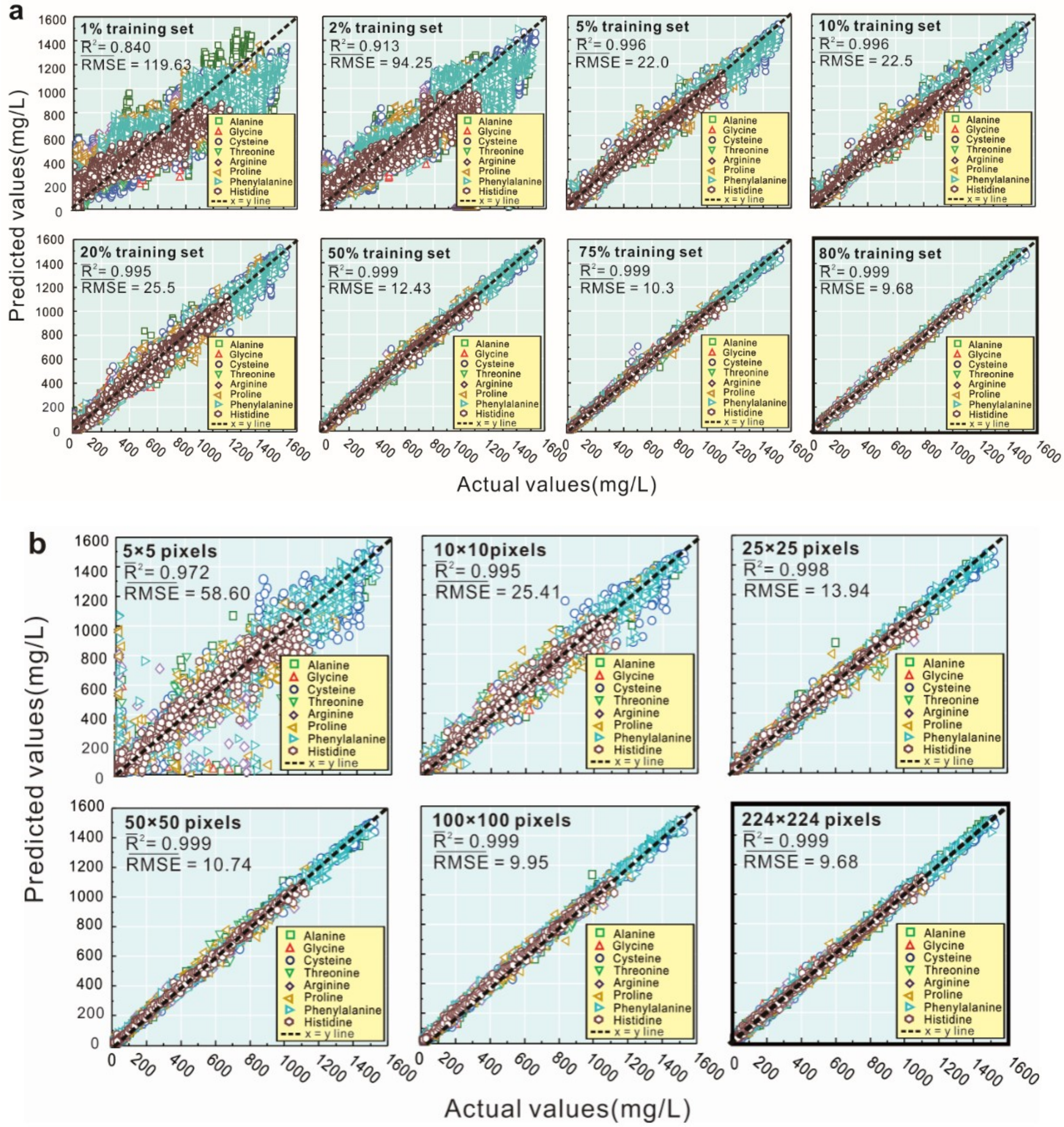


Figure S4 Test set performances of the Inception V1 model. Scatter plots of (**a**) variable proportions of the training data, and (**b**) different image sizes. Evaluation parameters: R^2 and RMSE; dashed line, y = x line. Mean R^2 and RMSE of 8 AAs were remarked.

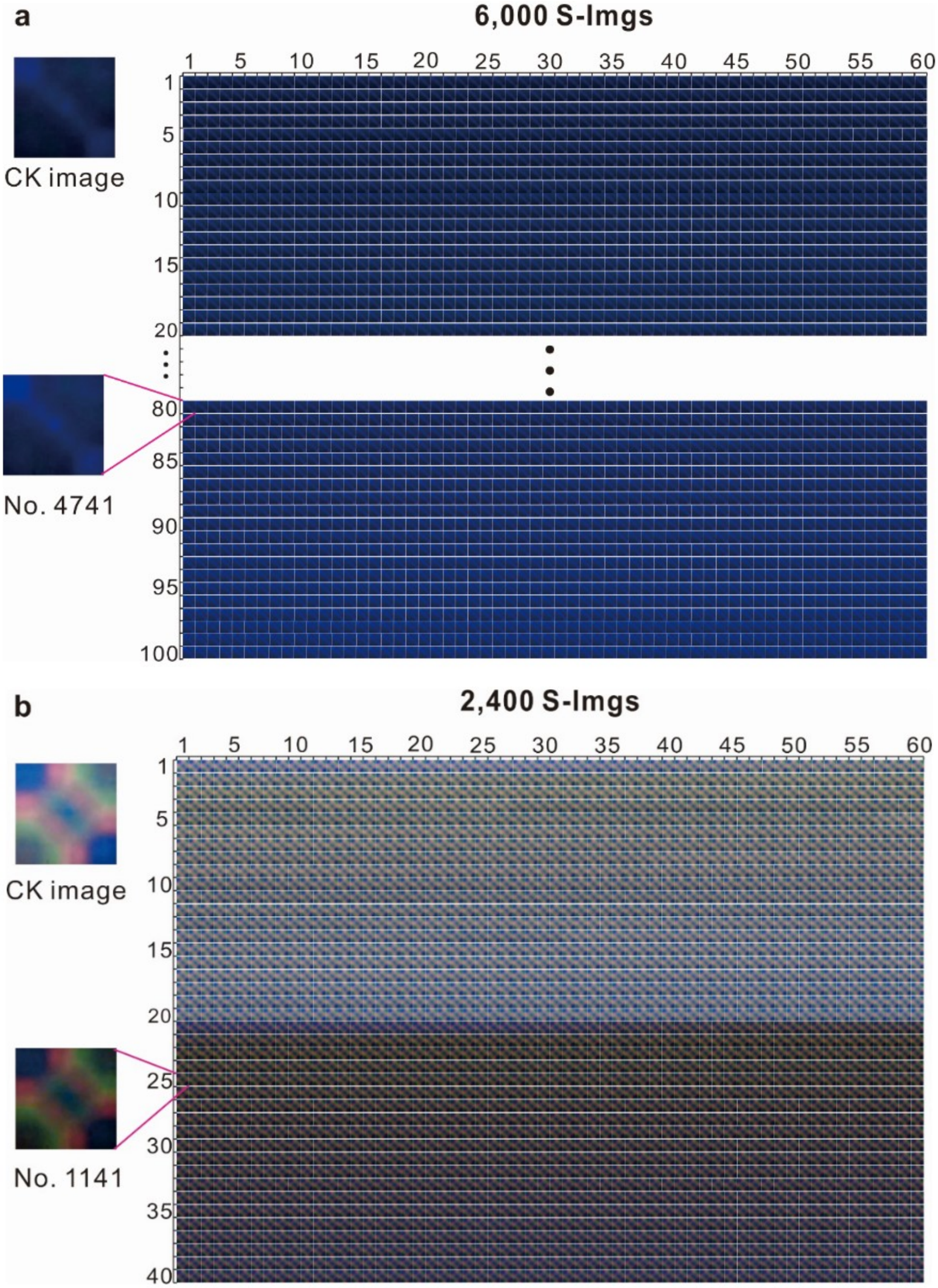


Figure S5 (**a**) S-Img set collected from 6,000 urine samples (containing 5 AAs: threonine, proline, histidine, glycine and phenylalanine). (**b**) S-Img set collected from 2,400 soil solution samples (containing 3 heavy mental ions: Ni^{2+}, Cu^{2+} and Cr^{3+}).

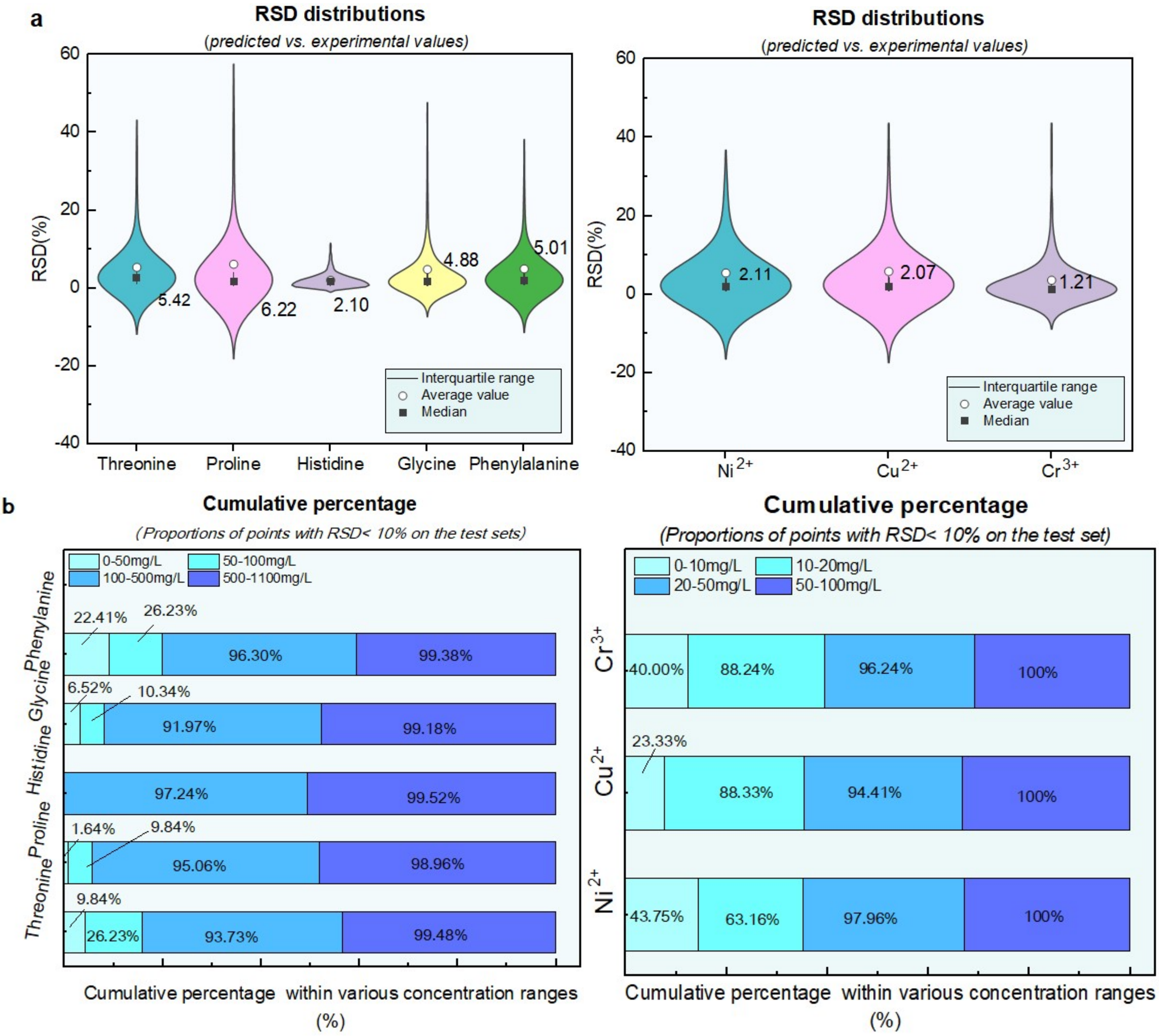


Figure S6 Distribution of relative standard deviation (RSD) between predicted vs. experimental values. (**a**) Violin plot of the RSD on five amino acids and ternary heavy mental ions. (**b**) Cumulative RSD on various concentration ranges of five amino acids and ternary heavy mental ions.

Table S1 List of chromogenic reagents

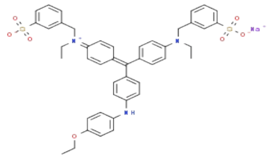
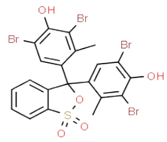
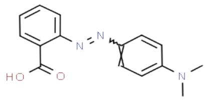
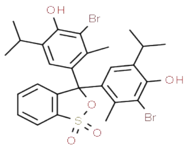
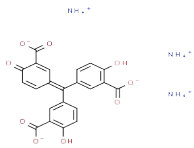
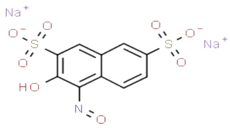
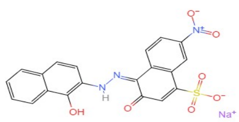
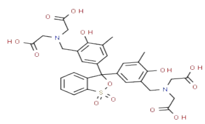
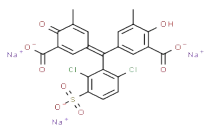
Entry	Description	Formula
1	Coomassie Brilliant Blue (CBB-G250)	
2	Bromocresol Green (BCG)	
3	Methyl Red (MR)	
4	Bromothymol Blue (BTB)	
5	Aluminon	
6	Naphthol Green	
7	Eriochrome Black T	
8	Nitroso-R Salt	
9	Xylenol Orange	
10	Chrome Azurol-S (CAS)	

Table S2 List of four mixed systems

Entry	Description	pH	Working Conc. (g/L)	Temperature (°C)
System I: 3-dimensional pollutant samples (bulk solution: water)				
1	$NiSO_4 \cdot 6H_2O$	3	1	25
2	$Cu(NO_3)_2 \cdot 3H_2O$	3	1	25
3	$CrCl_3 \cdot 6H_2O$	3	1	25
System II: 8-dimensional amino acid samples (bulk solution: water)				
1	Alanine	8	20	25
2	Glycine	8	20	25
3	Cysteine	8	20	25
4	Threonine	8	20	25
5	Arginine	8	20	25
6	Proline	8	20	25
7	Phenylalanine	8	20	25
8	Histidine	8	20	25
System III: 5-dimensional amino acid samples (bulk solution: human urine)				
1	Threonine	8	20	25
2	Proline	8	20	25
3	Histidine	8	20	25
4	Glycine	8	20	25
5	Phenylalanine	8	20	25
System IV: 3-dimensional pollutant samples (bulk solution: soil solution)				
1	$NiSO_4 \cdot 6H_2O$	3	1	25
2	$Cu(NO_3)_2 \cdot 3H_2O$	3	1	25
3	$CrCl_3 \cdot 6H_2O$	3	1	25

All the working concentrations of heavy metal solutions refers to the metal atom concentrations.

Table S3 Shooting parameters

Image parameters				
Image resolution	Bit depth	Horizontal resolution	Vertical resolution	Color model
1280×720	24	600 dpi	600 dpi	RGB
Camera parameters				
Recording time	Private int width	Private int height	Video bit rate	Refocus
4 min	1280	720	1080 kbps	30 fps

Table S4 Background values of the target free amino acids and heavy metals in real samples

5 amino acids in urine solution (mg L^{-1})				
Thr	Pro	His	Gly	L-Phe
-	-	215.65	13.87	3.42
3 heavy metals in soil solution (mg L^{-1})				
	Ni	Cu	Cr	
	0.03	0.14	0.02	

Table S5 Detailed architecture of CNNs

Google Inception v1 CNN		
Layer	**Layer(type)**	**Output Shape**
0	Input Layer	(None, 224,224,3)
1	Convolution Layer	(None, 112,112,64)
2	Relu function	(None, 112,112,64)
3	Padding Layer	(None, 113, 113, 64)
4	Max Pooling Layer	(None, 56, 56, 64)
5	Local Response Normalization Layer	(None, 56, 56, 64)
6	Convolution Layer	(None, 56, 56, 64)
7	Relu function	(None, 56, 56, 64)
8	Convolution Layer	(None, 56, 56, 192)
9	Relu function	(None, 56, 56, 192)
10	Local Response Normalization Layer	(None, 56, 56, 192)
11	Max Padding Layer	(None, 57, 57, 192)
12	Pooling Layer	(None, 28, 28, 192)
13	Inception_Net Graph	(None, 28, 28, 256)
14	Inception_Net Graph	(None, 28, 28, 480)
15	Padding Layer	(None, 29, 29, 480)
16	Max Pooling Layer	(None, 14, 14, 480)
17	Inception_Net Graph	(None, 14, 14, 512)
18	Inception_Net Graph	(None, 14, 14, 512)
19	Inception_Net Graph	(None, 14, 14, 512)
20	Inception_Net Graph	(None, 14, 14, 528)
21	Inception_Net Graph	(None, 14, 14, 832)
22	Padding Layer	(None, 15, 15, 832)
23	Max Pooling Layer	(None, 7, 7, 832)
24	Inception_Net Graph	(None, 7, 7, 832)
25	Inception_Net Graph	(None, 7, 7, 1024)
26	Max Pooling Layer	(None, 1, 1, 1024)
27	Dropout Layer	(None, 1, 1, 1024)
28	Flatten Layer	(None, 1024)
29	Linear Layer	(None, 1000)
30	Ramp	(None, 1000)
31	Linear Layer	(None, X)
32	Output	(None, X)

RestNet-50		
Layer	**Layer(type)**	**Output Shape**
0	Input Layer	(None, 224, 224, 3)
1	Convolution Layer	(None, 112, 112, 64)
2	Batch Normalization Layer	(None, 112, 112, 64)
3	Ramp	(None, 112, 112, 64)
4	Padding Layer	(None, 113, 113, 64)
5	Pooling Layer	(None, 56, 56, 64)

6	NetGraph (12 nodes)	(None, 56, 56, 256)
7	NetGraph (10 nodes)	(None, 56, 56, 256)
8	NetGraph (10 nodes)	(None, 56, 56, 256)
9	NetGraph (12 nodes)	(None, 28, 28, 512)
10	NetGraph (10 nodes)	(None, 28, 28, 512)
11	NetGraph (10 nodes)	(None, 28, 28, 512)
12	NetGraph (10 nodes)	(None, 28, 28, 512)
13	NetGraph (12 nodes)	(None, 14, 14, 1024)
14	NetGraph (10 nodes)	(None, 14, 14, 1024)
15	NetGraph (10 nodes)	(None, 14, 14, 1024)
16	NetGraph (10 nodes)	(None, 14, 14, 1024)
17	NetGraph (10 nodes)	(None, 14, 14, 1024)
18	NetGraph (10 nodes)	(None, 14, 14, 1024)
19	NetGraph (12 nodes)	(None, 7, 7, 2048)
20	NetGraph (10 nodes)	(None, 7, 7, 2048)
21	NetGraph (10 nodes)	(None, 7, 7, 2048)
22	Pooling Layer	(None, 1, 1, 2048)
23	Flatten Layer	Vector (None, 2048)
24	Linear Layer	Vector (None, 1000)
25	Ramp	Vector (None, 1000)
26	Linear Layer	Vector (None, X)
27	Output	Vector (None, X)

SqueezeNet V1.1

Layer	Layer(type)	Output Shape
0	Input Layer	(None, 227, 227, 3)
1	Convolution Layer	(None, 113, 113, 64)
2	Ramp	(None, 113, 113, 64)
3	Pooling Layer	(None, 56, 56, 64)
4	NetGraph (7 nodes)	(None, 56, 56, 128)
5	NetGraph (7 nodes)	(None, 56, 56, 128)
6	Padding Layer	(None, 57, 57, 128)
7	Pooling Layer	(None, 28, 28, 128)
8	NetGraph (7 nodes)	(None, 28, 28, 256)
9	NetGraph (7 nodes)	(None, 28, 28, 256)
10	Padding Layer	(None, 29, 29, 256)
11	Pooling Layer	(None, 14, 14, 256)
12	NetGraph (7 nodes)	(None, 14, 14, 384)
13	NetGraph (7 nodes)	(None, 14, 14, 384)
14	NetGraph (7 nodes)	(None, 14, 14, 512)
15	NetGraph (7 nodes)	(None, 14, 14, 512)
16	Dropout Layer	(None, 14, 14, 512)
17	Convolution Layer	(None, 14, 14, 1000)
18	Ramp	(None, 14, 14, 1000)
19	Aggregation Layer	(None, 1000)
20	Flatten Layer	(None, 1000)
21	Ramp	(None, 1000)
22	Linear Layer	(None, X)

23	Output	(None, X)

LeNet-5		
Layer	**Layer(type)**	**Output Shape**
0	Input Layer	(None, 28, 28, 3)
1	Convolution Layer	(None, 40, 40, 20)
2	Ramp Layer	(None, 40, 40, 20)
3	Pooling Layer	(None, 12, 12, 20)
4	Convolution Layer	(None, 8, 8, 50)
5	Ramp Layer	(None, 8, 8, 50)
6	Pooling Layer	(None, 4, 4, 50)
7	Flatten Layer	(None, 800)
8	Linear Layer	(None, 500)
9	Ramp Layer	(None, 500)
10	Linear Layer	(None, X)
11	Output	(None, X)

X represents the mixture dimension of output vectors, and its value is as follows: 3, 5 or 8. Besides, all datasets used for training and test the models can be publicly downloaded at https://github.com/Jianchaoleesnnu/Image-Dataset-from-Complex-Chemistry without any restriction.