

Supplementary Information

Deep learning classification of Li-ion battery materials targeting accurate composition classification from laser-induced breakdown spectroscopy high-speed analyses

Marie-Chloé Michaud Paradis¹, François R. Doucet^{2,*}, Steeve Rousselot¹, Alex Hernández-García³, Kheireddine Rifai², Ouardia Touag¹, Lütfü Ç. Özcan², Nawfal Azami⁴ and Mickaël Dollé^{1,*}

¹ Laboratory of chemistry and electrochemistry of solids, Department of Chemistry, Université de Montréal, 1375 Ave. Thérèse-Lavoie-Roux, Montréal, QC, Canada, H2V 0B3

² ELEMISSION Inc., 3410 Thimens Blvd, Montréal, QC, Canada, H4R 1V6

³ Mila - Institut Québécois d'Intelligence Artificielle, Université de Montréal, 6666 Saint-Urbain Street, Montréal, QC, Canada, H2S 3H1

⁴ Optics Lab, Institut National des Postes et Télécommunications, Avenue Allal Al Fassi, Rabat, Maroc

* Correspondence:

FD: fdoucet@elemission.ca; Tel.: 1-514-998-3713

MD: mickael.dolle@umontreal.ca; Tel.: 1- 514-343-7054

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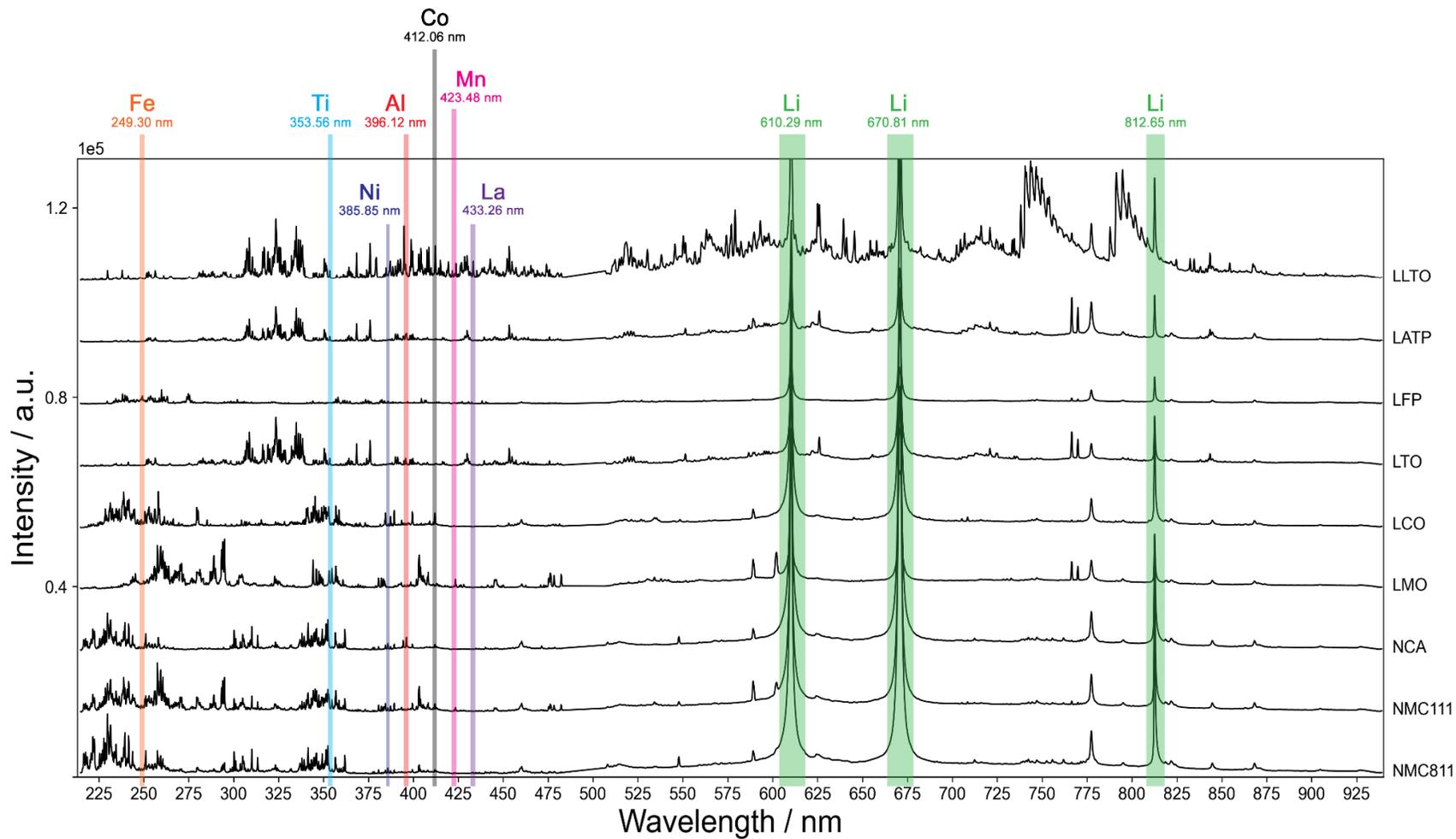


Figure S1. LIBS mean spectra of certain the battery materials and chosen transition lines analyzed using the described method between 215 and 940 nm.

Table S1. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using selected lines as inputs and using the AM algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	9.09	9.09	9.09	2.0E-04	1.5E-01
	RSD	<0.01	<0.01	<0.01	2.1E+00	7.4E-02
0.02	Mean	9.09	9.10	9.10	3.0E-04	1.5E-01
	RSD	<0.01	<0.01	<0.01	1.6E+00	6.1E-02
0.05	Mean	9.16	9.09	9.09	2.0E-04	1.5E-01
	RSD	0.02	<0.01	<0.01	2.1E+00	8.3E-02
0.1	Mean	9.09	9.09	9.09	2.9E-04	1.5E-01
	RSD	<0.01	<0.01	<0.01	1.6E+00	6.0E-02
0.2	Mean	9.09	9.09	9.09	3.0E-04	1.6E-01
	RSD	<0.01	<0.01	<0.01	1.6E+00	5.9E-02
0.5	Mean	9.09	9.09	9.09	4.0E-04	1.6E-01
	RSD	<0.01	<0.01	<0.01	1.3E+00	4.2E-02

Table S2. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using selected lines as inputs and using the SVM algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	75.82	66.57	66.87	2.9E-03	7.2E-02
	RSD	0.06	0.10	0.11	3.0E-01	4.9E-02
0.02	Mean	68.55	64.94	64.73	3.6E-03	9.4E-02
	RSD	0.08	0.07	0.07	3.0E-01	5.4E-02
0.05	Mean	61.75	61.13	61.25	7.3E-03	1.7E-01
	RSD	0.09	0.08	0.08	2.7E-01	7.2E-02
0.1	Mean	60.09	58.14	58.14	1.3E-02	2.9E-01
	RSD	0.08	0.11	0.11	2.1E-01	7.1E-02
0.2	Mean	59.69	58.72	58.65	3.8E-02	5.1E-01
	RSD	0.03	0.06	0.06	1.3E-01	4.7E-02
0.5	Mean	59.45	59.07	58.44	1.7E-01	1.1E+00
	RSD	0.04	0.05	0.05	9.1E-02	2.6E-02

Table S3. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using selected lines as inputs and using the RF algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	100.00	87.94	87.77	1.2E-01	8.1E-01
	RSD	<0.01	0.02	0.02	4.5E-02	2.8E-02
0.02	Mean	100.00	92.19	92.26	1.3E-01	8.1E-01
	RSD	<0.01	0.01	0.01	3.2E-02	1.4E-02
0.05	Mean	100.00	94.66	94.57	1.5E-01	8.3E-01
	RSD	<0.01	0.01	0.01	2.9E-02	3.8E-02
0.1	Mean	100.00	95.47	95.54	1.8E-01	8.2E-01
	RSD	<0.01	<0.01	<0.01	3.7E-02	1.4E-02
0.2	Mean	100.00	96.43	96.26	2.6E-01	8.4E-01
	RSD	<0.01	<0.01	<0.01	3.1E-02	3.0E-02
0.5	Mean	100.00	96.65	96.81	5.1E-01	9.0E-01
	RSD	<0.01	<0.01	<0.01	2.1E-02	2.9E-02

Table S4. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using selected lines as inputs and using the DNN1 algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	97.09	94.76	94.96	1.1E+02	3.4E-01
	RSD	0.02	0.01	<0.01	2.5E-02	1.6E-01
0.02	Mean	96.64	95.05	95.05	8.4E+01	3.2E-01
	RSD	0.02	0.01	<0.01	2.0E-01	3.5E-02
0.05	Mean	96.07	95.68	95.46	7.3E+01	3.3E-01
	RSD	0.01	<0.01	<0.01	1.5E-01	2.7E-02
0.1	Mean	96.02	95.58	95.55	5.9E+01	3.4E-01
	RSD	0.01	<0.01	<0.01	3.3E-01	3.3E-02
0.2	Mean	96.38	95.91	95.81	5.5E+01	3.3E-01
	RSD	0.01	<0.01	<0.01	3.3E-01	7.2E-02

0.5	Mean	96.16	95.97	96.03	4.9E+01	3.3E-01
	RSD	0.01	<0.01	<0.01	4.3E-01	1.8E-02

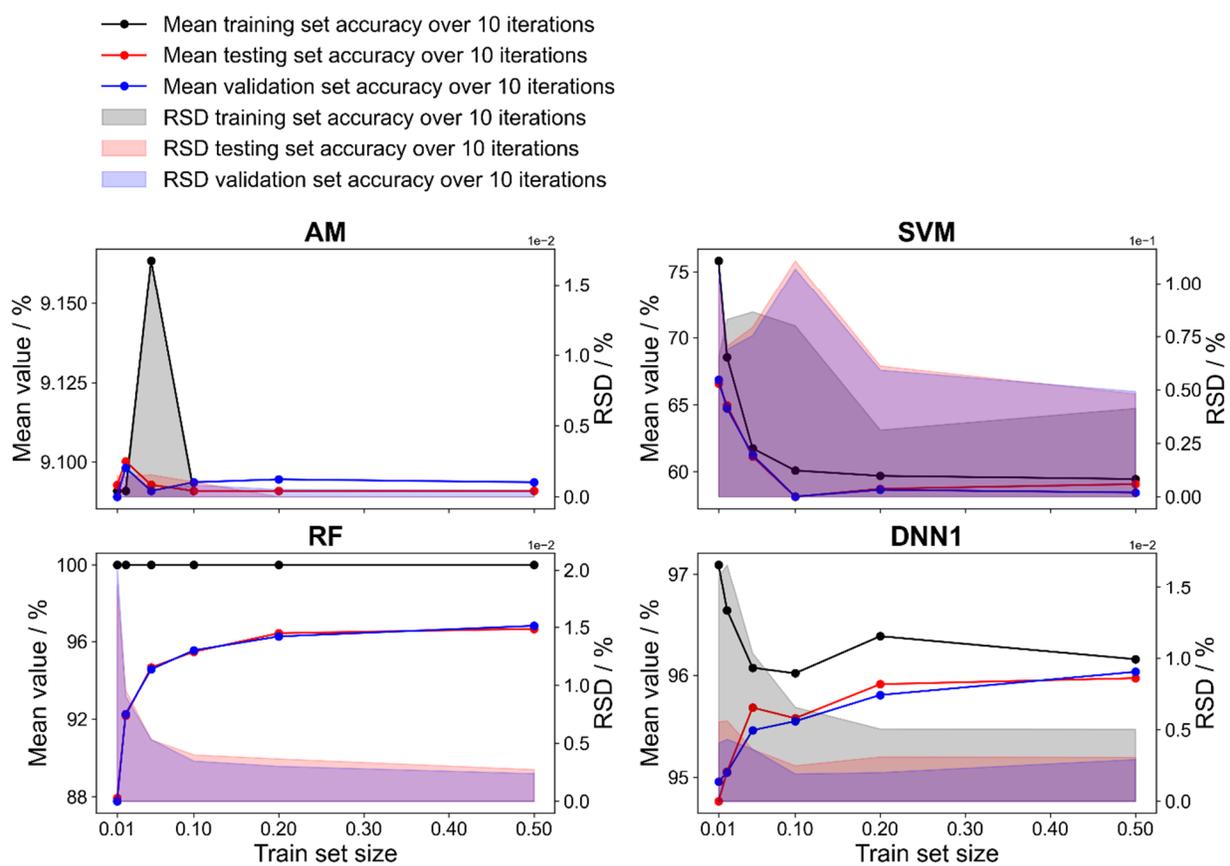


Figure S2. Accuracy and RSD as functions of the size of the training set for different machine learning classification algorithms over 10 iterations using selected transition lines.

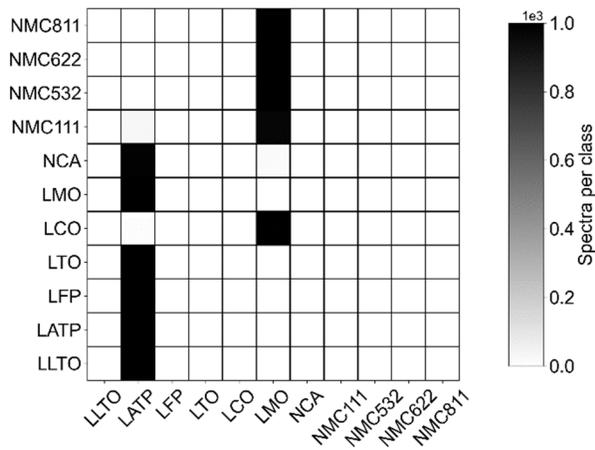


Figure S3. Confusion matrix using selected lines as inputs and using the AM algorithm with a 0.01 train set size.

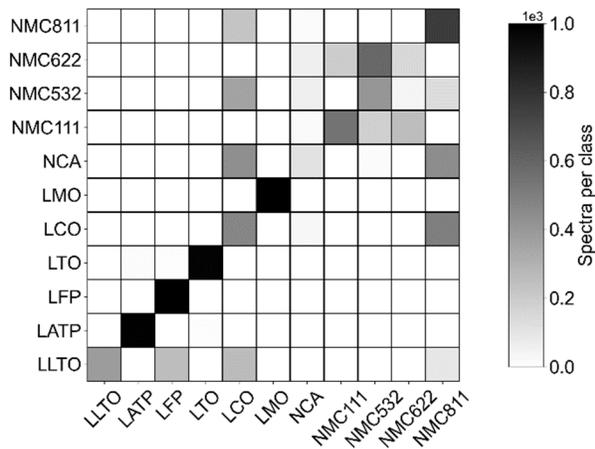


Figure S4. Confusion matrix using selected lines as inputs and using the SVM algorithm with a 0.01 train set size.

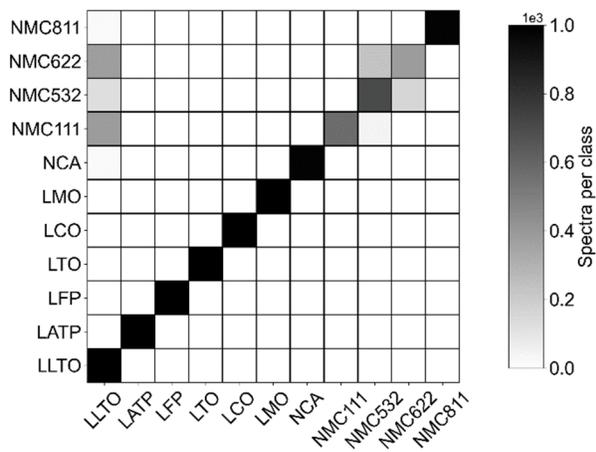


Figure S5. Confusion matrix using selected lines as inputs and using the RF algorithm with a 0.01 train set size.

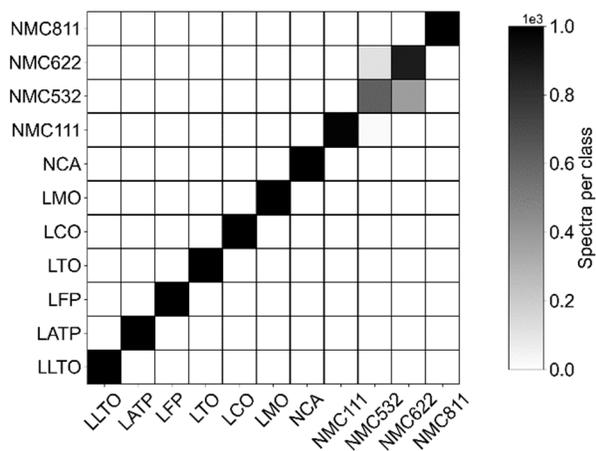


Figure S6. Confusion matrix using selected lines as inputs and using the DNN1 algorithm with a 0.01 train set size.

Table S5. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the AM algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	85.64	82.45	82.38	1.0E-03	1.8E-01
	RSD	0.03	0.02	0.02	4.2E-01	2.1E-01
0.02	Mean	85.45	82.16	82.07	1.2E-03	1.7E-01
	RSD	0.04	0.03	0.03	4.4E-01	1.9E-01
0.05	Mean	86.62	84.73	84.88	3.0E-03	2.0E-01
	RSD	0.01	<0.01	0.01	3.9E-01	1.9E-01
0.1	Mean	85.16	84.72	84.58	4.2E-03	1.7E-01
	RSD	0.01	0.01	0.01	2.9E-01	1.9E-01
0.2	Mean	85.41	84.56	84.68	8.9E-03	1.9E-01
	RSD	0.01	0.01	0.01	2.8E-01	2.1E-01
0.5	Mean	84.91	84.71	84.88	1.8E-02	1.8E-01
	RSD	0.01	0.01	<0.01	2.9E-01	1.8E-01

Table S6. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the SVM algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	80.55	74.89	75.04	1.5E-02	2.2E+00
	RSD	0.06	0.05	0.05	1.1E+00	3.6E-02
0.02	Mean	78.45	75.65	75.44	2.0E-02	2.9E+00
	RSD	0.04	0.04	0.04	9.5E-01	9.6E-02
0.05	Mean	79.85	78.64	78.86	3.3E-02	3.8E+00
	RSD	0.04	0.02	0.02	6.0E-02	2.5E-02
0.1	Mean	81.05	80.89	80.74	7.3E-02	4.2E+00
	RSD	0.02	0.02	0.01	3.6E-01	1.2E-02
0.2	Mean	83.04	82.56	82.70	1.6E-01	4.8E+00
	RSD	0.01	0.02	0.01	1.2E-01	7.7E-03
0.5	Mean	84.75	84.49	84.63	6.4E-01	6.6E+00
	RSD	<0.01	0.01	<0.01	2.8E-02	7.0E-03

Table S7. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the RF algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	100.00	78.95	78.97	2.5E-01	1.2E+00
	RSD	<0.01	0.04	0.04	6.8E-02	2.3E-02
0.02	Mean	100.00	88.93	88.73	4.0E-01	1.2E+00
	RSD	<0.01	0.01	0.01	4.3E-02	1.4E-02
0.05	Mean	100.00	94.47	94.27	9.1E-01	1.2E+00
	RSD	<0.01	0.01	0.01	1.3E-02	2.5E-02
0.1	Mean	100.00	96.49	96.44	1.8E+00	1.2E+00
	RSD	<0.01	0.01	0.01	1.0E-02	2.6E-02
0.2	Mean	100.00	97.91	97.99	3.8E+00	1.3E+00
	RSD	<0.01	0.01	0.01	1.5E-02	1.5E-02
0.5	Mean	100.00	99.16	99.14	1.0E+01	1.3E+00
	RSD	<0.01	<0.01	<0.01	9.1E-03	3.5E-02

Table S8. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the DNN1 algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	100.00	93.02	93.03	9.6E+01	1.1E+00
	RSD	<0.01	0.02	0.02	8.4E-02	6.2E-02
0.02	Mean	99.91	96.17	96.13	6.4E+01	1.2E+00
	RSD	<0.01	0.01	0.01	1.6E-01	1.2E-01
0.05	Mean	99.75	97.97	97.95	7.0E+01	1.3E+00
	RSD	<0.01	0.01	0.01	4.4E-01	8.5E-02
0.1	Mean	99.42	98.22	98.16	5.0E+01	1.4E+00
	RSD	<0.01	0.01	0.01	4.4E-01	3.1E-02
0.2	Mean	99.95	99.55	99.59	6.4E+01	1.4E+00
	RSD	<0.01	<0.01	<0.01	3.1E-01	3.3E-02
0.5	Mean	100.00	99.97	99.94	6.2E+01	1.6E+00

RSD	<0.01	<0.01	<0.01	2.5E-01	2.4E-01
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Table S9. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the DNN2 algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	100.00	93.03	93.12	9.2E+01	1.1E+00
	RSD	<0.01	0.02	0.02	8.7E-02	4.6E-02
0.02	Mean	99.73	96.24	96.17	6.5E+01	1.2E+00
	RSD	<0.01	0.01	0.01	1.2E-01	3.6E-02
0.05	Mean	99.05	97.44	97.42	5.5E+01	1.3E+00
	RSD	0.01	0.01	0.01	4.2E-01	9.2E-02
0.1	Mean	99.18	98.01	97.93	5.0E+01	1.4E+00
	RSD	0.01	0.01	0.01	3.5E-01	6.7E-02
0.2	Mean	99.95	99.60	99.63	7.5E+01	1.5E+00
	RSD	<0.01	<0.01	<0.01	3.3E-01	2.9E-02
0.5	Mean	99.99	99.95	99.93	5.5E+01	1.5E+00
	RSD	<0.01	<0.01	<0.01	2.3E-01	4.4E-02

Table S10. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the DNN3 algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	97.09	90.48	90.49	8.5E+01	1.2E+00
	RSD	0.07	0.06	0.06	1.9E-01	1.2E-01
0.02	Mean	99.73	96.12	96.06	6.1E+01	1.2E+00
	RSD	<0.01	0.01	0.01	1.2E-01	4.7E-02
0.05	Mean	98.95	97.48	97.41	5.4E+01	1.3E+00
	RSD	0.01	0.01	0.01	4.4E-01	7.3E-02
0.1	Mean	98.00	97.23	97.03	4.5E+01	1.4E+00
	RSD	0.01	0.01	0.01	3.8E-01	4.5E-02
0.2	Mean	99.90	99.50	99.50	7.0E+01	1.5E+00

	RSD	<0.01	0.01	0.01	2.9E-01	1.2E-01
0.5	Mean	100.00	99.92	99.92	5.5E+01	1.5E+00
	RSD	<0.01	<0.01	<0.01	1.7E-01	2.8E-02

Table S11. Mean and RSD of training, testing and validation sets accuracies, and training and testing times using full spectra as inputs and using the CNN1 algorithm over 10 iterations.

Train set size	Value	Training set accuracy / %	Testing set accuracy / %	Validation set accuracy / %	Training time / s	Testing time / s
0.01	Mean	82.55	77.62	77.68	1.1E+02	1.4E+00
	RSD	0.44	0.44	0.44	4.7E-01	5.5E-02
0.02	Mean	82.00	80.00	79.94	8.1E+01	1.5E+00
	RSD	0.44	0.44	0.44	5.3E-01	9.2E-02
0.05	Mean	99.31	98.07	98.18	9.4E+01	1.6E+00
	RSD	0.01	0.01	0.01	6.3E-01	5.1E-02
0.1	Mean	99.24	98.90	98.88	1.1E+02	1.7E+00
	RSD	0.01	0.01	0.01	5.8E-01	3.6E-02
0.2	Mean	100.00	99.93	99.94	1.2E+02	1.7E+00
	RSD	<0.01	<0.01	<0.01	2.4E-01	2.1E-02
0.5	Mean	100.00	100.00	99.99	7.2E+01	1.8E+00
	RSD	<0.01	<0.01	<0.01	2.3E-01	1.0E-01

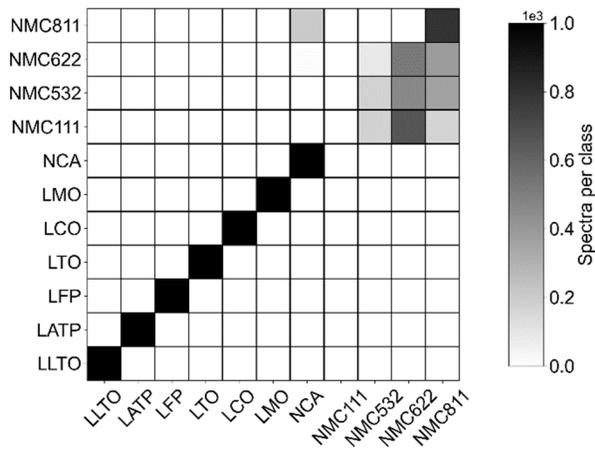


Figure S7. Confusion matrix using full spectra as inputs and using the AM algorithm with a 0.01 train set size.

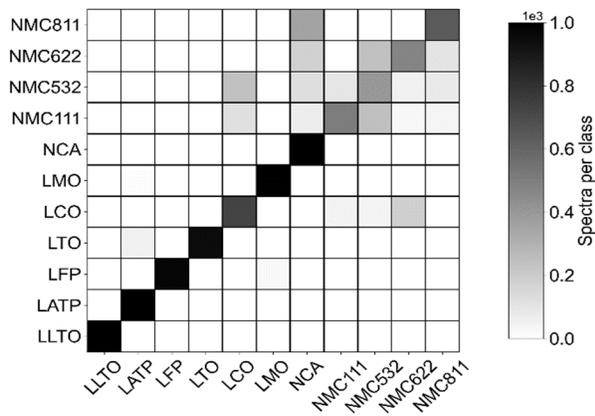


Figure S8. Confusion matrix using full spectra as inputs and using the SVM algorithm with a 0.01 train set size.

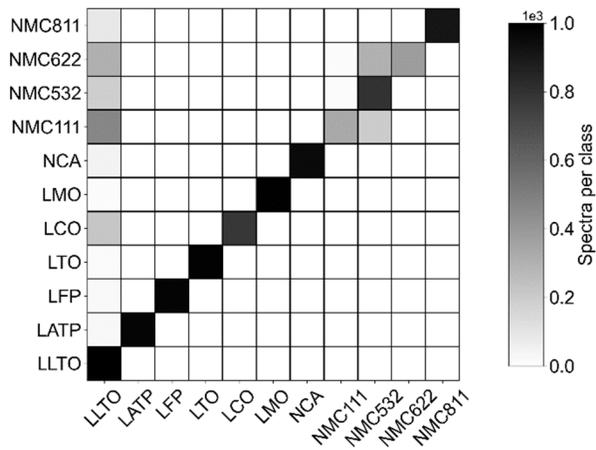


Figure S9. Confusion matrix using full spectra as inputs and using the RF algorithm with a 0.01 train set size.

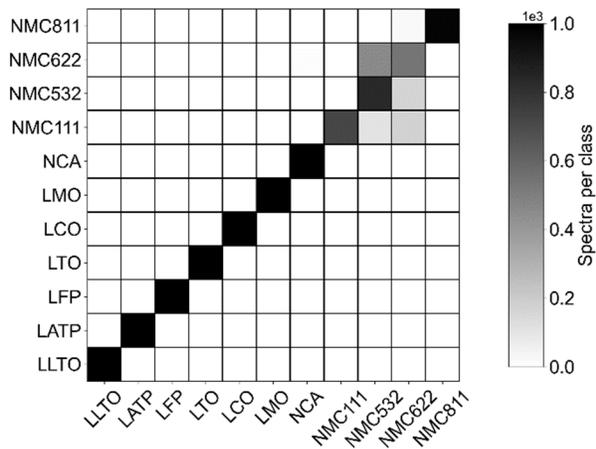


Figure S10. Confusion matrix using full spectra as inputs and using the DNN1 algorithm with a 0.01 train set size.

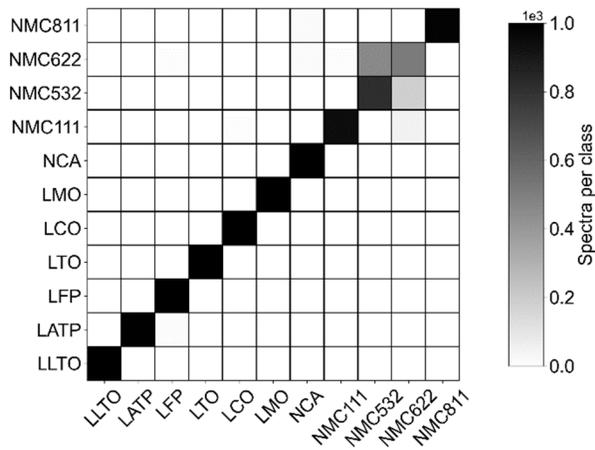


Figure S11. Confusion matrix using full spectra as inputs and using the DNN2 algorithm with a 0.01 train set size.

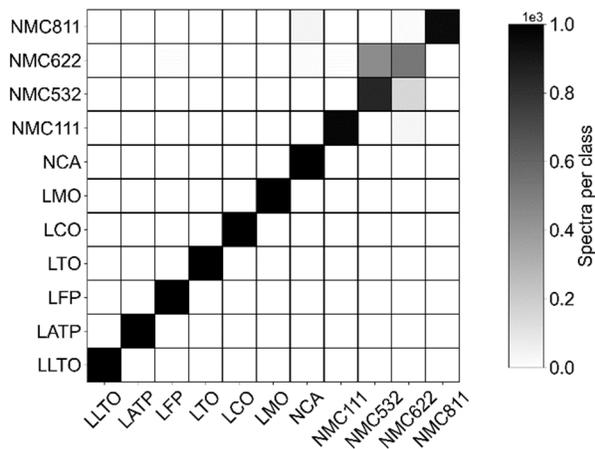


Figure S12. Confusion matrix using full spectra as inputs and using the DNN3 algorithm with a 0.01 train set size.

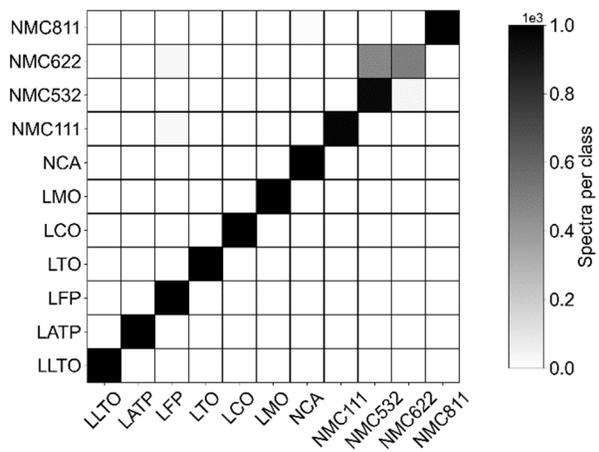


Figure S13. Confusion matrix using full spectra as inputs and using the CNN1 algorithm with a 0.01 train set size.

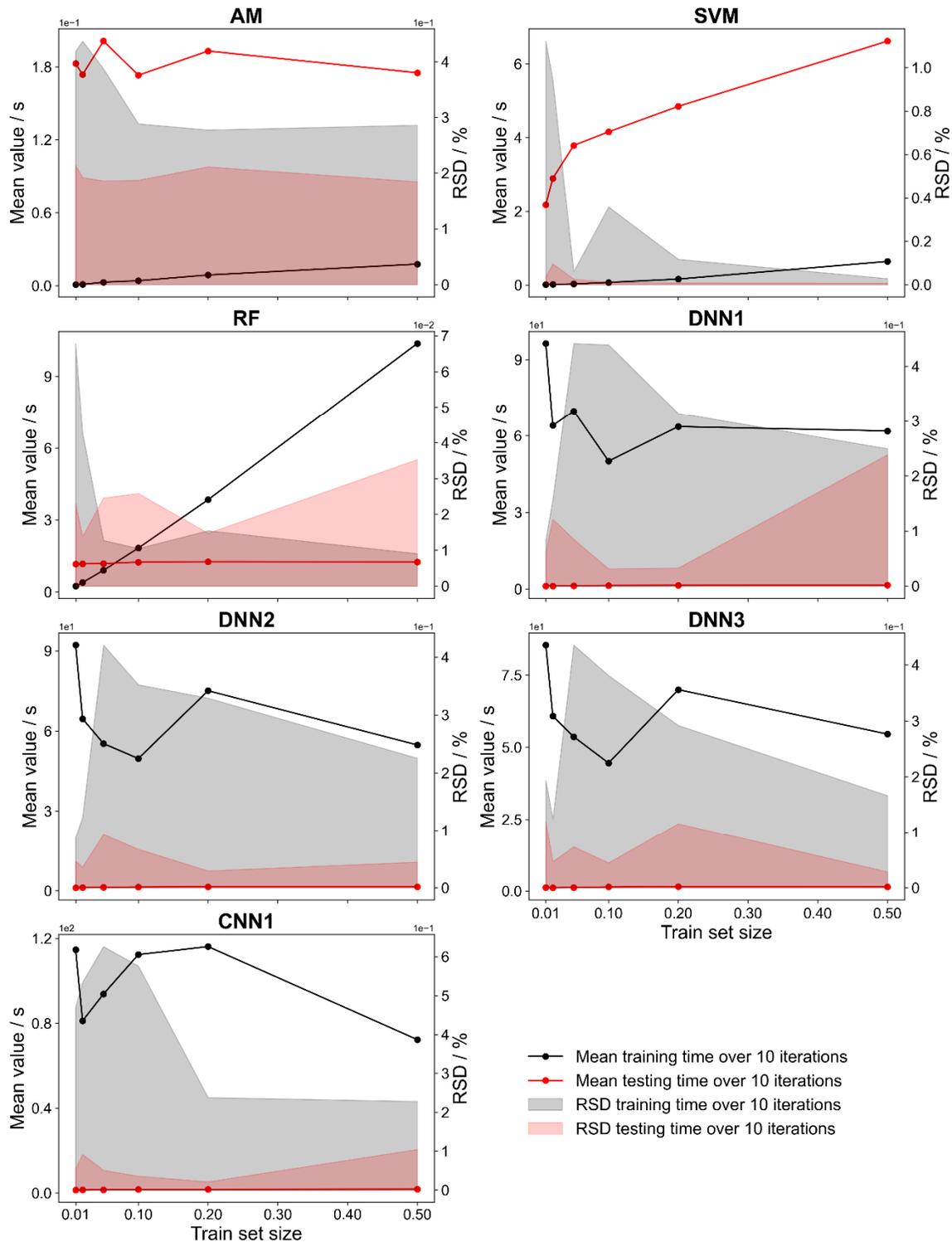


Figure S14. Training and testing times and RSD as functions of the size of the training set for different machine learning classification algorithms over 10 iterations using full spectra.