



## Assessment of a cu-perovskite material in a heterogeneous electro-fenton process for the degradation of organic dyes contaminants in a wide range of ph

**Cruz del Álamo, A.; Zou, Rusen; Pariente, M. I.; Molina, R.; Martinez, F.; Zhang, Yimin**

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<b>Acrylonitrile photocatalytic degradation: An experimental and CFD study to understand the influence of media geometry on the process efficiency</b>	92
H.E. Whyte, C. Raillard, A. Subrenat, V. Héquet .....	
<b>Developments in green catalytic technologies for a circular economy of water</b>	94
M. Canle, M.I. Fernández, Z. Marín, S. Aguilar, B. Astray, D.R. Ramos, J.A. Santaballa .....	
<b>About the formation of carbon monoxide in Fenton oxidation process: influence of the operating conditions</b>	95
J. Carbajo, A. Quintanilla, J.A. Casas .....	
<b>Assessment of a Cu-perovskite material in a heterogeneous electro-Fenton process for the degradation of organic dyes contaminants in a wide range of pH</b>	97
A. Cruz del Álamo, R. Zou, M.I. Pariente, R. Molina, F. Martínez, Y. Zhang .....	
<b>Coupling of an electro-Fenton device in a domestic system for wastewater disinfection</b>	99
L. Godínez, E. Becerra, I. Robles .....	
<b>Assessment of two strategies of solar photo-Fenton at neutral pH for simultaneous disinfection and microcontaminant removal in secondary WWTP effluents</b>	101
P. Soriano-Molina, B. Esteban García, J.A. Sánchez Pérez .....	
<b>Calcium peroxide modified Fenton oxidation as treatment technology for pesticide contaminated groundwater</b>	103
L. F. Sørensen, J. Muff .....	
<b>Enhanced photocatalytic activity of novel TiO<sub>2</sub> thin films developed on a laser-microstructured Si surface</b>	105
C. Christophoridis, M. Kanidi, M. Kandyla, G. Mousdis, E. Bizani, A. Hiskia .....	
<b>Highly PEC active hematite nanoflakes grown from magnetron sputtered iron films on FTO substrate: role of substrate temperature during the plasma deposition</b>	107
H. Ahn, Š. Kment, M. Zelny, R. Ctvrtilk, H. Kmentova, Z. Hubicka, A. Naldoni, P. Schmuki, R. Zboril .....	
<b>Investigation of the effect of fluidization regimes in the degradation rates of an annular photo reactor</b>	109
L. A. Diniz, D. Matsumoto, A. C. S. C. Teixeira .....	
<b>Modeling the capacity of a continuous flow photocatalytic reactor with dynamic solar irradiation conditions for the degradation of caffeine</b>	111
C. Dezani, C. Caliot, G. Plantard, V. Goetz .....	
<b>EPR study of hybrid noble-metals/TiO<sub>2</sub> nanomaterials</b>	113
D. Dvoranová, Z. Barbieriková, V. Brezová, M. Saeli, D.M. Tobaldi .....	
<b>Effect of experimental parameters on photocatalytic activity of supported TiO<sub>2</sub>, nano-photocatalyst on degradation of amoxicillin in a pilot sunlight reactor</b>	115
F. S. Moosavi, C. Cheng, T. T. Gheinani, A. Kanaev, M. Nikravech .....	
<b>Alumina supported composite catalysts for industrial air purification by catalytic oxidation</b>	117
T. Žumbar, A. Ristić, M. Popova, N. Zabukovec Logar, N. Novak Tušar .....	
<b>Co-Mn mixed oxides supported on hierarchical macro-mesoporous silicas for Co and VOCs oxidation</b>	118
S. Todorova, J. L. Blin, A. Naydenov, B. Lebeau, H. Kolev, P. Gaudin, A. Dotzeva, D. Filkova, I. Ivanova, L. Vidal, L. Michelin, L. Josien, K. Tenchev .....	
<b>Effect of MoVTeNbO catalyst modification on the physicochemical and catalytic properties in oxidative dehydrogenation of ethane</b>	120
E. Lazareva, V. Bondareva, D. Svintsitskiy, E. Kovalev, T. Kardash .....	



## ASSESSMENT OF A Cu-PEROVSKITE MATERIAL IN A HETEROGENEOUS ELECTRO-FENTON PROCESS FOR THE DEGRADATION OF ORGANIC DYES CONTAMINANTS IN A WIDE RANGE OF pH

A. Cruz del Álamo<sup>a</sup>, R. Zou<sup>b</sup>, M.I. Pariente<sup>a</sup>, R. Molina<sup>a</sup>, F. Martínez<sup>a</sup>, Y. Zhang<sup>b</sup>

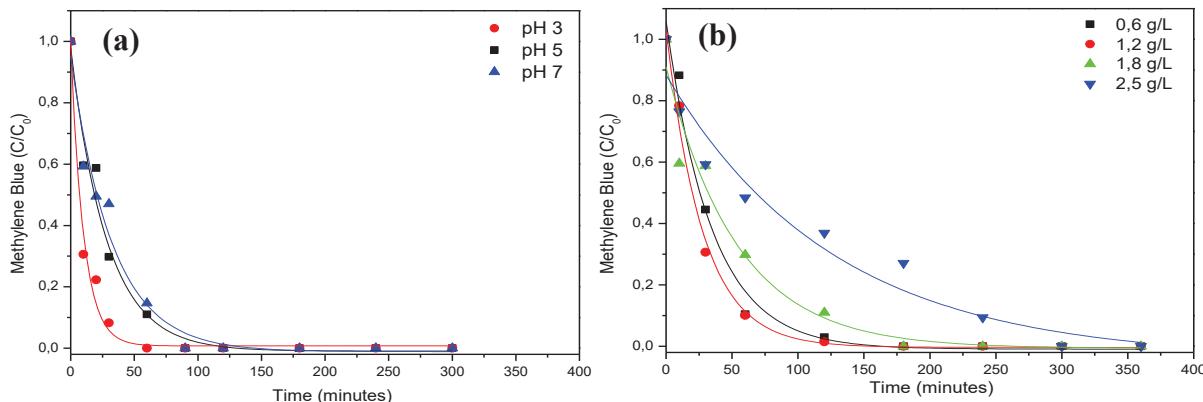
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These days, the textile industry produces large volumes of tinted wastewater containing a high amount of organic contaminants (Pazdior et al., 2018). Among them, dyes are considered particularly relevant in industrial effluents due to it is difficult to use conventional treatment technologies for their elimination (Cintra Fernandes et al., 2018; Cruz-Rizo et al., 2017). Some processes such as coagulation-flocculation, flotation, adsorption, membranes or reverse osmosis have been reported for the dyes removal. However, they are not effective for the complete decoloration treatment (Cintra Fernandes et al., 2018; Cruz-Rizo et al., 2017; Saleh and Taufik, 2019a). Alternatively, advanced oxidation processes (AOPs), mainly photochemical and electrochemical systems, have showed promising potential for removal of dyes from industrial wastewater streams (Saleh and Taufik, 2019b; Poza-Nogueriras et al., 2018). The electro-Fenton process has been received much attention in order to improve the classical Fenton system by in-situ hydrogen peroxide electrogenereration in the cathode via reduction of dissolved oxygen. Until now, most of the works reported in literature have studied homogeneous electro-Fenton systems under acid pH conditions (Poza-Nogueriras et al., 2018). Current efforts are focused on the development of active heterogeneous catalyst in a wider range of pH. Perovskite-type oxides ( $\text{ABO}_3$ ) provides remarkable red-ox properties and highly mobility of oxygen and electrons through their crystalline structures (Carrasco et al., 2016). These features makes perovskite-type oxides promising heterogeneous Fenton-like catalysts. At present, the use of perovskite materials in electro-Fenton-like reactions is scarce and limited to acid pH conditions (Ben Hammouda et al., 2019). Thus, the aim of this work is the evaluation of Cu-perovskite material ( $\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$ ) as heterogeneous electro-Fenton-like catalyst in the degradation of methylene blue (MB) as model pollutant.

$\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$  perovkiste was synthetized following a modification of Pecini method proposed by Carrasco et al. (2016). Electro-Fenton experiments were performed in a undivided batch electrochemical chamber working at room temperature ( $22 \pm 5^\circ\text{C}$ ) under continuous stirring (200 rpm). The cathode was a graphite plate ( $9 \text{ cm}^2$ ) and the anode was titanium mesh coated Iridium mix metal oxides (Magneto special anodes B.V., The Netherlands) with a separation gap of 2 cm between them. A direct current of 2 V was supplied by a power supply (HQ PS3003, Helmholt Elektronik A/S, Denmark). Both electrodes were connected through a titanium wire of 1 mm diameter with an external resistor of  $10 \Omega$  for monitoring the current intensity in the chamber along the reaction time. The chamber was continuously bubbled with air (20 mL/min) by a peristaltic pump. In a typical run, 130 mL of a methylene blue solution (5-10 mg/L) in mixture with 50 mM of  $\text{Na}_2\text{SO}_4$  was placed in the chamber after appropriate

adjustment of the pH (3, 5 or 7). Powder Cu-perovskite was added in the desired catalytic loading (0.6-2.5 g/L). After that, the power supply was turned on, starting the reaction. Several aliquots were withdrawn along the reaction for the determination of MB removal as indicator of the catalyst activity.

Different experiments were carried out to assess the effect of two operating conditions: pH and perovskite dosage. **Figure 1** shows the evolution of methylene blue concentration for electro-Fenton experiments performed at different pH (a) and perovskite concentration (b).



**Figure 1.** Removal of MB (a) at pH 3, 5 and 7 with initial concentration of  $\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$  of 1.85 g/L and  $[\text{MB}]_0 = 5 \text{ mg/L}$ ; (b) at different perovskite concentration (0.6, 1.2, 1.85 and 2.5 g/L) with pH 5,  $[\text{MB}]_0 = 10 \text{ mg/L}$ .

A total decoloration of MB was obtained after 30 minutes at pH 3, but it was also noteworthy the activity of  $\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$  at higher pH conditions of 5 and 7. This fact is particularly remarkable as efficiency of homogeneous electro-Fenton systems is normally limited to acid pH about 2-3 (Ben Hammouda et al., 2019). The current intensity in these reactions were in the range of 5 and 10 mA (0.55 and 1.1 mA/cm<sup>2</sup>). The decrease of the catalyst concentration until 0.6 g/L allowed a better electrocatalytic performance of Cu-perovskite. These results seem to be also in agreement with the data reported in literature (Ben Hammouda et al., 2019), as an excess of electro-active metal species can promote the scavenging of hydroxyl radicals. Additionally, the presence of powder heterogeneous particles of Cu-perovskite could increase the internal resistance for the electrons transfer from anode to cathode.

As conclusion,  $\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$  showed a remarkable activity for the removal of MB at circumneutral pH. These results suggest that the perovskite would be a promising option for organic compounds removal by electro-Fenton process working with neutral water pH.

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M. Canle 94, 381  
 V. Capucci 57  
 J. Carbojo 95, 479  
 C. F. Carbuloni 134  
 C. de O. Cardoso 570  
 G. Carraro 50  
 J. Carrillo-Abad 547  
 J. A. Casas 95, 195, 298, 479, 500, 616  
 J. L. Casas Lopez 199, 427  
 S. Castilhos 326  
 M. Á. Castillo 288, 719  
 O. Castillo 306  
 S. Castro-Silva 238, 455  
 J. L. Cenis Anadon 707  
 G. Cerrato 57  
 G. Cerreta 66  
 J. I. Chairez 243, 306, 670  
 T. Chankhaniththa 284  
 A. Chatzikakis 286  
 S. I. Chávez Ruiz 77  
 P. Chelme-Ayala 201  
 J.Y. Chen 188  
 X. Chen 122  
 Y.J. Chen 697  
 C. Cheng 115  
 K. Cho 136, 528  
 S. Cho 178, 280  
 J. Choi 557  
 C. Christophoridis 105, 161, 398, 502  
 C. Cionti 184, 399  
 S. Cipagauta-Díaz 687  
 S. Cirés 298  
 R. Ciriminna 61  
 K. Cizek 72  
 S. Clerici 184  
 S. Collado 81, 159, 407  
 R. Comparelli 653  
 S. Contreras 263  
 L. Cornejo Ponce 225  
 O. Cotazo 643  
 S. Cotillas 150, 265  
 A. Cruz del Alamo 97, 508  
 A. Cruz-Alcalde 276, 445, 632, 635, 637  
 R. Ctvrtlik 107  
 M. L. Curri 653  
 B. Czech 210  
 Zs. Czékes 336  
 A. Czyzewski 396

## Č

L. Čapek 593, 685  
 M. Čeh 649  
 M. Čehovin 170, 576

## D

R. F. Dantas 632  
 N. De Bellis 340  
 A. E. De Carvalho 370  
 I. de Godos 62  
 I. De La Obra Jimenez 199, 427  
 V. A. de la Peña O'Shea 44, 383  
 A. de Noni Junior 83, 604  
 Z. M. de Pedro 298, 500  
 M. Deggelmann 145  
 T. Delclos 634  
 C. Della Pina 399  
 D. G. Della Rocca 604  
 M. Dell'Edera 653  
 F. Demartin 57  
 K. Demeestere 206  
 H. Demir-Duz 263  
 E. Denierea 206  
 R. Dewil 253, 614, 651  
 C. Dezani 111  
 T. D'Halluin 300  
 C. di Luca 465  
 A. Di Michele 57  
 M. V. Diamanti 655  
 M. Diaz 81, 159, 407  
 J. Diaz-Angulo 443, 483  
 J. L. Diaz de Tuesta 142, 532  
 N. Diban 560  
 R. Dillert 272  
 D. Dimotikali 413  
 M. Dinca 364  
 L. A. Diniz 109, 647  
 D. D. Dionysiou 144, 360, 416, 618  
 P. Djinović 59, 308, 459, 461  
 K. Domen 36  
 A. Doménech 441  
 C. M. Dominguez 191, 471  
 N. Donate 582  
 E. Donner 701  
 H. J. Dorantes-Rosales 243, 670  
 A. J. M. dos Santos 489  
 O. A. A. dos Santos 326, 328, 370, 372,  
     374, 418  
 J. Dostanić 319  
 A. Dotzeva 118  
 B. Drigo 701  
 X. Duan 618  
 L. Dubnová 593, 685  
 S. Dudziak 126, 668  
 J. Dueñas Moreno 243, 306, 670

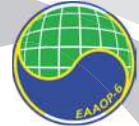
N. Dulova 242  
 D. Dusnoki 391  
 D. Dvoranová 113  
 P. Dzik 368, 612

## E

P. Ecorchard 524, 595  
 M. Edelmannova 685  
 K. Eha 242  
 A. Elaissi 451  
 N. B. El-Assy 572  
 E.A. El-Sharkawy 572  
 Á. Encinas 274  
 A.A. Esmailpour 261  
 S. Esplugas 276, 445, 635, 637  
 B. Esteban Garcia 101, 199, 427

## F

D. Fabbri 268  
 E. Falletta 399  
 M. Fanetti 267  
 J. L. Faria 70, 142, 423, 532  
 G. Farinelli 217  
 D. I. Farkas 554  
 A. Fašková 437  
 L. Fašková 556, 559  
 D. Fatta-Kassinios 457, 701  
 Á. Fazekas 552  
 E. Felis 411  
 L. Feng 229  
 J. Fenoll 705, 707  
 M. Ferentz 310  
 J. R. Fernandes 491  
 D. Fernández 407  
 M.I. Fernández 94  
 D. Fernández-Pérez 324  
 M. Fernández-Castro 383  
 P. Fernández-Ibáñez 213, 251  
 A. M. Ferrari 134, 518  
 C. Ferreiro 445  
 C. Ferronato 75  
 V. Fester 628  
 J. Fick 249  
 M. Z. Fidelis 370, 372, 418  
 J. L. Figueiredo 79  
 D. Filkova 118, 545  
 L. Fine 75  
 M. N. Fini 477  
 D. Fino 247

**K**

O.N.H. Kaabeche 381

Z. C. Kadirova 210

F. Kaissouni 451

R.J. Kaleńczuk 376

V. Kálmán 589

T. Kaloudis 161

Yu. Kalvachev 545

S.-K. Kamaraj 566

B. Kamenická 72, 645

A. Kamiyama 232

A. Kanaev 115, 625

M. Kandyla 105

M. Kanidi 105

A. J. Karabelas 405

P. Karaolia 457

A. Karapa 425

A. Karatas 163

T. Kardash 120, 603

Zs. Kása 589, 639

M. Kask 657

P. Kassai 587

C. P. Katsumata 487

B. Kaur 242

E.-Z. Kedves 304, 666

T. Kedzierski 358

N. Keller 64, 85

L. Kępiński 674

S. Kertesz 587, 589

G. Keszthelyi-Szabó 554

A. Khedr 715

C. Kim 178, 280

D. I. Kim 350, 352

H.-R. Kim 166

J. Kim 528

V. Kitsiou 393, 395, 497

P. Kluson 504

Š. Kment 90, 107, 377

H. Kmentová 107, 377

O. Koba Ucun 163, 626

K. Kočí 680, 685

J. Koc-Jurczyk 610

H. Kolev 118, 545

D. I. Kondarides 356

T. Konečná 680

M. Kong 144, 618

F.-D. Kopinke 221

I. Kornev 659, 678

M. E. Kounaris Fuziki 370

G. Kovács 336, 354, 666, 710

K. Kovács 693

Z. Kovács 304, 639, 641

J. Kovač 649

S. Kovačič 165

E. Kovalev 120

N. S. Kovalevskiy 211, 415

E. Kowalska 278

K. Kowalska 411

D. V. Kozlov 211, 415

P. Kraszkiewicz 245

M. Krbaľ 321

M. Krichevskaya 657

I. Krivtsov 245

J. Krýsa 46, 153, 317, 332, 504

H. Krýsová 46

E. Kubíňáková 437

M. Kulkarni 377

L. Kuntus 659

E. Kusiak-Nejman 396

P. Kuštrawski 593, 685

**L**

E. Lacasa 150

G. Lafaye 451

K. Lalas 580

L. Lan 566

M. Landau 310

J. A. Lara-Ramos 443, 483

L. Lartundo-Rojas 124

U. Lassi 257

A. M. Lastre-Acosta 487, 489

Zs. László 552, 554, 587, 589

E. Laurenti 155

U. Lavrenčič Štangar 50, 267, 564

E. Lazareva 120, 603

J. Le-Cunff 623

B. Lebeau 118

S. Ledakowicz 234, 431, 447, 449

K. Ledwa 674

B.-K. Lee 180

H. Lee 186

J. W. Lee 350, 352

J. Lee 265, 534, 721

S. Lee 270

W. J. Lee 534

C. Lefevre 85

M. Lelis 514

G. G. Lenzi 326, 328, 370, 372, 374, 418

C. Lepeytre 300

C. Li 566

G. Li Puma 40, 282, 586

T.-T. Lim 534

C.-C. Lin 551

X. Lin 566

Y.R. Lin 697

R. Lindberg 249

D. Liu 566

L. Lo Presti 184

A. Lobnik 208

V. Loddo 334

K. Loganathan 205, 438

J. I. Lombraña 445

M. A. Lominchar 219

D. Lončarević 319

M. F. Lopes 372, 374

F. López 473

N. Lopez -Saavedra 483

M. J. López-Muñoz 324, 407, 493, 634

N. López-Vinent 632, 635, 637

D. Lorenzo 471, 481

C.Y. Lu 697

M.S. Lucas 322, 491

M. Luna 383

**M**

J.M. Macák 46

C. Maccato 50

E. Macedo 506

B. F. Machado 142

A. Machálková 524, 595

F. Machuca-Martinez 443, 483

L. M. Madeira 157, 506, 699

P. J. Mafa 292

G. Magnacca 672

K. Magyari 695

G. Mailhot 268

I. Major Barbosa 489

J. L. Makuda 372

S. Malato 48, 66, 77, 213, 225, 520

M. I. Maldonado 213

F.J. Maldonado-Hódar 506

M. Malefane 294

I. Malinowska 148, 401

R. Mallada 591

J. A. Malvestiti 632

M. Manfredi 296

R.V. Mangalaraja 314

M. Manrique-Gallardo 687

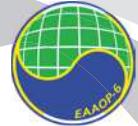
H. D. Mansilla 314

A. Mantilla 124

- H. Mantilla 510  
 D. Mantzavinos 159, 356, 578, 580, 608  
 F. Manzotti de Souza 326  
 M. Marcé 445  
 C. Marchal 64  
 P. Marco 635, 637  
 Z. Marín 94  
 M. J. Marin Figueredo 247  
 Ž. Marinko 649  
 A. Marinković 387  
 R. G. Marques 134  
 C. Márquez-Álvarez 634  
 J. Marszałek 447  
 V. Márta 710  
 M. Martín-Sómer 274, 682  
 T. Martinez 364  
 F. Martínez 97, 508  
 L. Martínez 383  
 A. Martínez García 251  
 D. Martínez-Pachón 172  
 J. Marugán 274, 682  
 N. Masri 385  
 L. Matějová 593, 685  
 L. Matoh 50, 564  
 L. M. Matos Jorge 326, 328  
 D. Matsumoto 109, 647  
 V. Maurino 340, 342  
 K. Maver 50  
 L. Maya-Treviño 348  
 M. Mazúr 59  
 S. Mei 584  
 F. Meneguzzo 61  
 N. Merayo 62  
 D. Meroni 184, 399  
 S. A. Messele 201, 606  
 S. Mestre 547  
 S. G. Michael 457, 701  
 I. Michael-Kordatou 457, 701  
 L. Michelin 118  
 J. C. Mierzwa 489  
 N. Miguel 608  
 D. Mijin 387, 467  
 E. Mijowska 358  
 H. Milh 253, 651  
 D. Min 280  
 M. Minella 217, 268  
 C. Minero 217  
 S. Miralles-Cuevas 225, 427, 520, 522  
 M. A. Miranda 713  
 S. M. Miranda 721  
 E. Mitsika 502  
 E. Mocchio 342  
 M. Mokhtarifar 655  
 C. B. Molina 562  
 R. Molina 97, 508  
 P. K. Molnar 477  
 A. Moncayo-Lasso 172  
 V.M. Monsalvo 274  
 B. Montazeri 626  
 J. P. Monteiro 721  
 N. Montesinos 312  
 F. S. Moosavi 115, 625  
 M. Mora 441, 574  
 V. C. Mora 197  
 J. Mora-Gómez 547  
 A. W. Morawski 396  
 A. J. Moreira 328  
 F. C. Moreira 265  
 N.F.F. Moreira 586  
 I. S. Morelli 197  
 J. Moreno-Andrés 193, 403  
 R. Mosteo 703  
 M. Motola 321  
 A. Moura Bernardes 138, 151  
 G. Mousdis 105  
 G. Moussavi 435  
 E. Mrotek 148  
 M. Mueses 443, 483  
 J. Muff 103, 477  
 M. Munoz 298, 500  
 R. Muñoz 62  
 M. Muñoz-Morales 147  
 M. Mustafa 249
- N**
- L. S. Nahim-Granados 203  
 A. Naldoni 90, 107, 377  
 S.-H. Nam 350, 352  
 S. Nanan 284, 385, 689, 709  
 E. Nascimben Santos 589  
 C. O. C. Nascimento 512  
 K. Nasr Esfahani 495  
 D. Nasuhoglu 176  
 S. Navarro García 705, 707  
 J. A. Navío Santos 703  
 A. Naydenov 118  
 E. Nebot 193, 403  
 J. Nedeljković 319  
 M. Neumann Spallart 46, 317, 332  
 S. Neumayer 429
- N. T. Nguyen 377  
 V. Nguyen 221  
 M. V. S. Nicolini 372  
 J. Nieto-Sandoval 298, 500  
 M. Nikravech 115, 625  
 A. G. Nogueira 238, 455  
 T. Norby 286  
 N. Novak Tušar 50, 89, 117, 621  
 P. Nunes 238, 455  
 R. Núñez-Salas 348
- O**
- O. Ogunbiyi 438  
 W. D. Oh 122  
 T. Ohno 54, 153  
 B. Ohtani 278  
 A.N. Ökte 619, 676  
 M. Olak-Kucharczyk 447, 449  
 J. R.P. Oliveira 532  
 I. Oller Alberola 48, 66, 213, 225, 251, 288, 296, 520  
 T. Olmez-Hancı 163, 626  
 S. Omanovic 176  
 A. Onjia 467  
 O. Oputu 628  
 E. Orbán 639  
 J. J. M. Órfão 79  
 C. A. Orge 421, 423, 439  
 M. P. Ormad 608, 703  
 F.E. Oropeza 601  
 E. M. Ortega 151, 547, 549  
 I. Ortiz 344, 538  
 P. Oulego 81, 159, 407  
 G. Ovejero 75, 453
- P**
- C. Pablos 274  
 L. Pacileo 317  
 M. C. Paganini 672  
 M. Pagliaro 61  
 J. L. Paiva 647  
 S. M. Palácio 512, 570  
 J. Palarčík 723  
 G. Palazzo 312  
 P. H. Palharim 485, 489  
 V. Palma 516  
 G. Palmisano 61, 74, 334, 389, 463, 634, 684  
 M. Panagiotopoulou 425  
 T. Pandiyarajan 314  
 D. Paneva 545



- Zs. Pap 304, 330, 331, 336, 338, 354, 391, 589, 639, 641, 660, 662, 664, 666, 693, 695, 710  
 I. Papailias 128, 379, 413  
 M. I. Pariente 97, 508  
 M. P. S. Parizi 487  
 J.-W. Park 166, 186  
 S. Park 352  
 Š. Paušová 153, 317  
 D. Paustian 145  
 A. Pavanello 713  
 J. Pavlović 621  
 D. Pavúková 556, 559  
 Y. Paz 56, 231  
 MP. Pedefterri 655  
 N. L. Pedersen 477  
 J. A. Pedraza-Avella 687  
 Ö. Pehlivan 163  
 C. Peillex-Delphe 163  
 F. Pellegrino 340  
 M. Peñas-Garzón 416, 562, 717  
 R. Peralta Muniz Moreira 282  
 M. F. R. Pereira 79, 423  
 J. A. Peres 322, 491  
 V. Pérez Herranz 151, 547, 549  
 L. A. Pérez-Estrada 77  
 M. Pérez-Moya 227, 495  
 S. Periyasamy 566  
 J.r. Pessemier 253  
 A. Petala 159, 356, 578, 580  
 F. Petronella 653  
 M. Petrovic 366  
 P. N. Petzi 405  
 C. Pham 64  
 A. Pintar 59, 89, 308, 310, 459, 463, 465  
 L. Pirault-Roy 451  
 J. Piriyanon 689  
 M. Piumetti 247  
 K. V. Plakas 405  
 G. Plantard 111  
 J. Plaza 493  
 P. Plaza-Bolaños 66, 203  
 O. Pliekhova 267  
 E. Pližingrová 000,  
 A. Podgornik 420  
 M. Podlogar 420  
 D. Pogocki 362  
 L. Pola 81  
 M. I. Pólo-López 203, 213, 251, 296  
 K. Polychronopoulou 255  
 G. Ponnusamy 205, 438  
 M. Popova 117  
 S. Portilla 259  
 I. Poulios 393, 395, 497, 499  
 T. Poznyak 243, 306, 670  
 P. Praça 532  
 M. Pradénas 314  
 S. Preis 659, 678  
 J. Prikryl 46  
 N. Prlainović 387  
 T. Prostějovský 680  
 K. Pstrowska 601  
 N. Pueyo 608  
 C. Pulgarín 229, 435, 469  
 R. Pupo Nogueira 711
- Q**
- L. Qian 221  
 Z.M. Qiang 132, 526  
 A. Quesada 298  
 N. Quici 312  
 A. Quintanilla 95, 479
- R**
- T. Radošević 420  
 J. M. Raez 324  
 S. S. Rai 660  
 C. Raillard 92, 315  
 N. Rajić 621  
 G. Rákely 331  
 A. Ramirez 591  
 M. Ramirez 711  
 D.R. Ramos 94, 381  
 L. Rancaño 344, 538  
 S. Ratchahat 68, 232  
 S. Rath 542  
 S. Raufeisen 145  
 A. Ravasz 304  
 G. Reis 433  
 M. Reli 680  
 P. Renones 383  
 B. Réti 695  
 D. M. Revelo 536  
 A.R.L. Ribeiro 586  
 R.S. Ribeiro 70  
 M. Rico-Santacruz 64  
 A. Ristić 89, 117  
 G. Rivas Ibañez 199  
 M. J. Rivero 344, 538  
 L. Rizzo 66, 168
- D. Robert 64, 85  
 I. Robles 87, 99  
 E. Robotti 691  
 M. A. Roccamante 66, 520  
 R. P. Rocha 79  
 M. A. Rodrigo 147, 150  
 C. S. D. Rodrigues 157, 699  
 C. Rodrigues-Silva 542  
 J. Rodríguez 74, 306  
 S. Rodríguez 481  
 J. J. Rodríguez 416, 562, 717  
 J. L. Rodríguez 000, 670  
 J. Rodríguez-Chueca 348, 469  
 A. Romero 191, 471, 481  
 L. Romero-Martinez 193, 403  
 J. A. Rosso 197  
 J. Ruiz Desales 243  
 A. Ruiz-Delgado 48  
 C. Ruiz-Palomar 62  
 N. Russo 247  
 J. Rynkowski 568
- S**
- R. Saab 255  
 C. Sabater 288, 719  
 M. Saeli 113  
 C. Sáez 147, 150  
 E. Salinas 306  
 I. Salmerón 213  
 E. Samaniego-Benítez 124  
 Z. Samardžija 649  
 E. F. S. Sampaio 699  
 K. Sánchez Gómez 443  
 I. A. Sanchez Ortiz 536  
 J. A. Sánchez Pérez 101, 199, 203, 427, 493, 522  
 S. Sanchis 364, 366  
 C. Sans 215, 276, 599, 632  
 T. Sansenya 385  
 J.A. Santaballa 94, 381  
 J. Santamaría 189, 591  
 P. Santander 314  
 C. Santillí 510  
 A. Santos 191, 219, 471, 481  
 A. S. G. G. Santos 421  
 M.S.F. Santos 506  
 J.S.P. Santos 134  
 T. D. Santos 518  
 V. A. Santos Ribeiro 346, 518  
 L. Santos-Juanes 475



- G. Visser 628  
 J. Volmajer Valh 208  
 M. Vorokhta 245  
 A. V. Vorontsov 182  
 M. Voumard 229  
 A. Zielińska-Jurek 126, 148, 278, 401, 668  
 D. Zingaretti 219  
 A. Zylan-Yavaş 130  
 J. Zoppas Ferreira 540  
 R. Zou 97  
 R. Zouaghi 381

## W

- B. Wabende 584  
 A. Wanag 396  
 H. Wang 249  
 Y. Wang 249  
 Y.-L. Wei 551  
 T. Weidlich 72, 645  
 M. Weiße 145  
 D. Wen 526  
 H. E. Whyte 92, 315  
 S. Wohlmuth da Silva 138, 151  
 M.C. Wu 697  
 I. Wysocka 126

## Ž

- R. Żyłka 447, 449  
 E. Žagar 165  
 R. Žebrák 680  
 G. Žerjav 59, 308, 310, 459, 463, 465  
 A. Žgajnar Gotvajn 170, 576  
 T. Žumbar 117

## X

- Y. Xiangwei 227  
 K. Xu 286

## Y

- M. J. Yañez-Gascón 707  
 J. W. Yang 352  
 J. Yang 584  
 M. Yang 584  
 V. Yargeau 176  
 L. Yıldız Ozer 61  
 J.-H. Yu 350, 352  
 A. Yusuf 334

## Z

- N. Zubukovec Logar 117  
 M. Zakar 554  
 N. Zambrano 508  
 M. Zampieri Fidelis 370, 372, 418  
 M. D. Zandi 495  
 J. Žavašnik 459, 461  
 J. A. Zazo 195, 616  
 R. Zazpe 46, 321  
 M. Zbair 451  
 R. Zbořil 90, 107, 377  
 M. Zelny 107  
 S. Zhang 358  
 Y. Zhang 97  
 E. S. Zhuravlev 415  
 B. Zielińska 376

