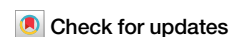


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Using a resistor network to simulate charge transport in organic semiconductors through injection layers and transport layers



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In the advancement of organic semiconductor devices, computer simulations now provide performance predictions and valuable insights into the inner mechanisms of potential device architectures, streamlining the need for time-intensive laboratory experiments. The Kinetic Monte Carlo method is a well-established approach for simulations of organic semiconductors on a mesoscopic scale, modeling charge carrier hopping by sampling discrete time steps. However, this method exhibits slow convergence for complex device architectures and systems with high charge carrier densities as typical for doped injection layers or transport layers. Here, we demonstrate an approach to compute charge transport even in samples with high charge carrier densities, utilizing a Resistor Network model with time-independent resistances. Using this approach we can reproduce current densities for a variety of device architectures while drastically reducing the computational time. Hence, this method can be used to simulate charge transport in non-emitting device layers tenfold faster than with a pure Kinetic Monte Carlo simulation.

Charge transport in amorphous organic semiconductors typically occurs through a hopping mechanism^{1–4}. In this transport mode, the charge carriers are localized on individual molecular sites. The carriers can hop between neighboring sites with a certain transition probability^{1,5}. Continuous efforts are devoted to investigating the complex interplay of material parameters with device architectures. These investigations rely on computational models and have proven to provide valuable insights into charge transport on a mesoscopic scale. Among the established computational methods, the Kinetic Monte Carlo (KMC) method has proven particularly useful because it can explicitly account for the Coulomb interaction between individual charge carriers⁶. KMC can be used to reliably simulate the charge transport in organic semiconductors (OSC)^{1,6,7}. However, KMC simulations become inefficient for systems containing a large number of charge carriers. In each simulation step, the charge distribution evolves by moving a single charge. The choice of which charge hops to a neighboring site is randomly selected from a vast option space, with the probabilities for these transitions and the associated time increment determined by the corresponding hopping rate^{1,6,7}. This necessitates frequent recalculation of transition rates for all charge carriers and their respective target sites. As the number of charge carriers increases, the most computationally demanding aspect of this process becomes the calculation of

Coulomb interactions between the carriers^{1,6}. Additionally, the occurrence of state rattling⁸, where carriers repeatedly jump back and forth, leads to an inefficient use of simulation steps, hindering the progression of the system.

During the simulation of charge transport in organic light-emitting diodes, high charge carrier concentrations are present in doped transport layers and injection layers^{9,10}. These layers foster charge transport towards an emissive layer, where the charges can recombine and radiatively decay^{9–11}. Apart from the contribution to the overall charge transport, the explicit carrier-carrier interaction of individual pairs is usually not of interest in these layers. Nevertheless, a major part of the computational effort will be spent on these layers even though only minor insight into the total device behavior is gained from state rattling steps. Explicit KMC simulations of charge transport in multilayered OLEDs are, therefore, computationally intensive and inefficient. This behavior is highlighted in Fig. 1, where a majority of charge moves is used in the injection layers due to state rattling. This work introduces an alternative and efficient approach to calculating charge transport in organic layers, circumventing the need for frequent recalculations of carrier-carrier interactions. The aim of this study is, to provide a model that can replace expensive current calculations in charge transporting layers in full device KMC simulations and focus the KMC steps

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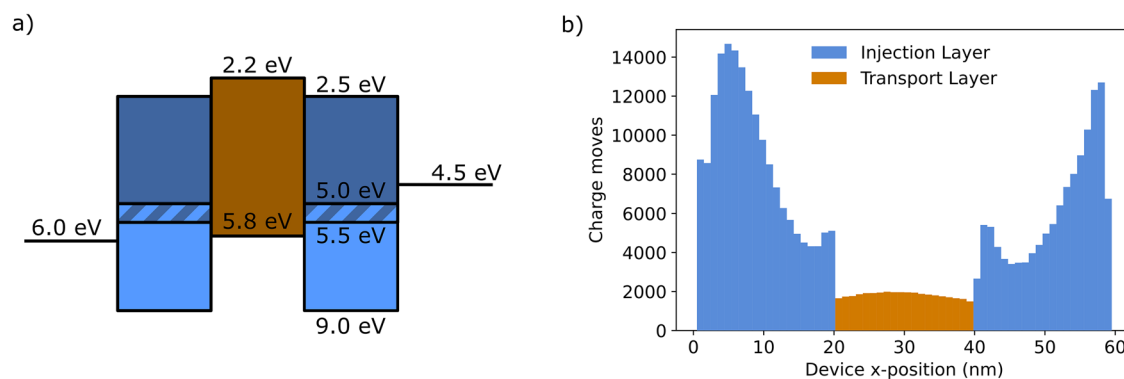


Fig. 1 | Demonstration of state rattling at interfaces. State rattling in doped injection layers makes kinetic Monte Carlo (KMC) simulations computationally inefficient in full device organic light-emitting diode (OLED) simulations and hinders longer simulation times, as needed, for example, in degradation studies. A simple toy system (a) is used to highlight the computational effort that is drawn toward the injection layers. The system consists of two doped injection layers with a host material (electron affinity (EA)/ionization potential (IP) at 2.5/5.5 eV) doped with a dopant material (EA/IP at 5.0/9.0 eV) at a concentration of 15%. Between these injection layers is a simple transport material (EA/IP at 2.2/5.8 eV). All layers

are 20-nm wide, and all materials share an energetic disorder of $\sigma = 100$ meV. Investigating the spatial distribution of charge moves (b) highlights the imbalance of computational effort for such a system. The majority of moves in the KMC simulation are simulated in the outer injection layers. Only a small fraction of charge moves are simulated in the middle layer. Hence, the majority of charge moves does not contribute to the calculation of current through the system, but the computational effort is used for processes like state rattling through which only minor insight into the overall device performance is gained. The KMC simulation was performed with 20 replicas with an applied voltage of 9 V.

to the remaining layers. Thus, in future work, this model can be used in systems such as the one in Fig. 1 to reduce the overall computational cost of the simulation by replacing the expensive layers. In this study, we present a method to accurately reproduce the current flowing through such layers. In this method, the OSC is modeled as a network of Ohmic resistors, with resistances connecting the molecular sites^{3,12}. The charge transport is then described by calculating the currents flowing through this resistor network (RN). This calculation is performed using a matrix equation for the currents, building upon the potential energies on the molecular sites, the resistances between the sites as modeled from microscopic hopping rates and an incidence matrix encoding the network topology. The construction of these matrices will be shown in the Methods section. Cottaar et al. previously introduced an RN to model charge transport in OSC¹³, where they considered percolation pathways, i.e. the pathways of least resistance, for their mobility calculations. While their model was able to reproduce mobilities, the approximations made during the derivation of the underlying formulas correspond to the low carrier density limit. This made it not applicable to systems with high carrier densities. While their model improves upon pure percolation theory by including a distribution of conductances instead of a single pathway, we want to go further and use all contributing pathways. Our RN model was used to calculate current densities in OSC systems. The method is applied to a variety of toy systems and compared to current densities obtained from KMC simulations. Different degrees of energetic disorder are examined, along with various system architectures relevant to multilayer OSC devices. Looking ahead, the goal of this method is to replace costly KMC simulations in parts of OSC devices where charge transport is the primary concern, but the overall simulation is burdened by slow convergence due to high carrier density effects in these layers, like state rattling.

However, to generate the RN for individual systems, short preliminary KMC simulations were performed using the KMC software package Lightforge¹⁴ to obtain the steady-state carrier distribution in the organic systems. These steady-state distributions are then used in the RN generation, where they act as input parameters for calculating resistances, along with the hopping rates between sites, as shown in Fig. 2. A more detailed explanation of the RN generation process can be found in the Methods section.

Nevertheless, applied to the monolayer, bilayer and doped organic semiconductor systems presented in Fig. 2, the RN method reproduces the reference KMC current densities across different energetic disorders, interfacial barriers and carrier concentrations. At the same time, the RN method reduces the computational cost by roughly one order of magnitude, with the largest absolute computational time reduction for systems with

high carrier densities. In contrast to the iterative evolution of the system in the KMC simulation, the RN method computes steady-state currents using time-independent resistances. These results indicate that the approach can serve as an efficient replacement for explicit charge transport calculations in injection and transport layers of future multilayer device simulations.

Results and discussion

Charge transport

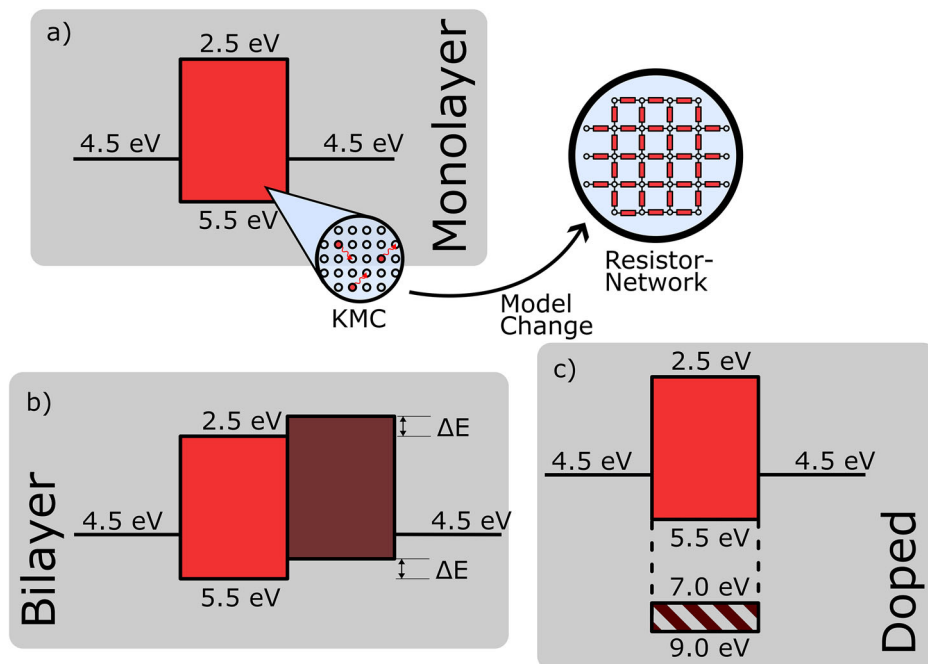
Because our approach works for one species at a time, we confined our investigation to single-carrier devices. Thus, the RN was only used to simulate hole transport in these systems. Further details will be discussed in the Methods section.

The results were compared with reference simulations performed using Lightforge¹⁴.

Monolayer systems. The monolayer systems consist of a single organic layer sandwiched between two electrodes. This simple system served as the initial test case to evaluate the capabilities of our RN model in reproducing the KMC current densities. To capture the structural disorder inherent in OSC materials, the energy levels of individual sites are sampled from a Gaussian distribution^{15–17}, with the mean values shown in panel a) in Fig. 2 and the standard deviation σ representing the energetic disorder. To explore a variety of organic materials, the energetic disorder is varied in different calculations. The resulting current characteristics are presented in Fig. 3, where the modeled disorder ranges from $\sigma = 15$ to $\sigma = 100$ meV. Regardless of the disorder, the results align well with the reference simulation, successfully reproducing the current densities. Additionally, the model accurately predicts the increased current density in monolayer devices with higher disorder, attributed to enhanced injection from the electrode into energetically closer tail states of the organic material, which has a broader state distribution¹⁸. Thus, the model is capable of simulating currents flowing into and through OSC materials.

Bilayer systems. Bilayer systems are the second type of systems examined, consisting of two different organic layers sandwiched between the electrodes. The interface between these layers introduces an additional energy barrier within the system. The purpose of investigating such bilayer systems is to determine whether the model can capture the behavior of carriers in the presence of barriers between organic layers. This is important for the model's potential use in simulating multilayer structures or complete multilayer devices, with the ultimate goal of replacing injection and transport layers with

Fig. 2 | Overview of the resistor network method and the tested systems. For the resistor-network approach, the material is modeled by a set of resistors, each of which represents a connection between hopping sites, forming a network and mediating the currents in the sample material. This is in contrast to the random hopping transitions of individual charge carriers used by the kinetic Monte Carlo (KMC) method. During the simulations, different systems are investigated. For the monolayer system (a), the sample layer consists of one material with LUMO and HOMO levels set to 2.5 and 5.5 eV. In addition, the layer is contacted by two electrodes with work functions set to 4.5 eV. For the Bilayer system (b), an additional layer is put between the first layer and the electrode. The energetic offset between the energy levels of these two layers is varied from 0.1 eV to 0.3 eV. For the doped system (c), a fixed percentage of sites in the Monolayer material are exchanged for dopant sites with LUMO and HOMO levels of 7.0 and 9.0 eV. The dopant molar ratio is varied from 0.1 % to 15 %.



our RN model. The energy diagram is shown in panel b) in Fig. 2. The energetic offset between the two organic layers is varied across different calculations. The resulting current characteristics are shown in Fig. 4. The energetic offset is varied to 0.1, 0.2, and 0.3 eV. The results show good agreement with the reference simulation. It is important to note that the visually large error bar at the 0.1 eV case arises from the symmetric error plotted on a logarithmic scale. Nevertheless, the calculated value is close to the reference, and the error bars are of similar magnitude. This suggests that the model can effectively handle interfaces between organic layers, which is a significant result, making our RN model suitable for simulating multilayer organic structures.

Doped systems. A third category under investigation is guest–host systems, where guest molecules improve the injection and conductivity properties of the organic layer^{9,19,20}. These systems, commonly known as doped systems, hold significant potential for acceleration if they can be accurately modeled by our RN model, due to their high concentration of free charge carriers. Since, the KMC simulation relies on the combinatorial calculation of interactions, the individual KMC step tends to become more computationally expensive in systems with a high charge carrier concentration. Hence, by introducing a method without frequent recalculations, there is a high cost-saving potential in these systems. This makes the ability to simulate charge transport in doped systems a critical foundation for integrating our RN method into full-stack OLED simulations. The energy levels of the dopant molecules are modeled using a Gaussian distribution, with considerably deeper HOMO and LUMO energies (see panel c) in Fig. 2). In this study, the guest molecules function as p-dopants, facilitating the transfer of electrons from the hosts HOMO levels to the vacant dopant LUMO levels. This process generates a free hole on the host molecule, therefore increasing the overall density of free-charge carriers. The performance of our RN model in computing current density is analyzed for three doping concentrations, ranging from low (0.1%) to high (15%). These results are benchmarked against current densities obtained from explicit KMC simulations, as depicted in Fig. 5. The RN model reproduced the KMC current densities for all three doping concentrations. However, for the 1% and 15% doping concentrations, the agreement was slightly worse at lower applied voltages compared to higher voltages. This voltage-dependent variation was not seen in the monolayer and bilayer cases (see Figs. 3 and 4). The smaller current

densities observed in the KMC simulations compared to those from our RN model can be attributed to the Coulomb interaction between dopants and host molecules. The attractive Coulomb interaction between the guest anions and the free holes leads to a form of hole-trapping close to the dopants, being reflected in an overall reduction of current density. Increasing the driving voltage mitigates this effect for two reasons: first, a higher concentration of holes is injected into the organic guest–host system, which diminishes the impact of individual trapped holes on the overall current density. Second, the stronger electric field acts as a driving force, helping to detrapp more holes from the attractive guest anions.

Computation time savings

We demonstrated that the RN model is capable of computing current densities in organic layers of varying complexity, ranging from monolayer systems with significant energetic disorder, to bilayer systems with inter-layer energy barriers, and finally to host-dopant systems with a wide range of dopant concentrations. During these calculations, a preliminary KMC simulation was used to obtain the carrier distribution. This will be explained further in the Methods section. We observed a potential speed-up in not running the preliminary KMC simulation until the currents have fully converged, but only to the point that the carrier distributions have sufficiently converged to be extracted for the RN. Since, the calculation of the RN currents is fast compared to the KMC simulation, this entails the potential for a speed-up. Nevertheless, the aim of our study was not to speed-up these calculations but to provide a model that can assist KMC in the simulation of full OSC devices in offering a replacement for expensive charge transporting layers after inexpensive calibration to these layers.

A runtime comparison for all the studied systems, between the pure KMC simulations and our RN model, is presented in Fig. 6. All calculations were performed on the same machine, equipped with an AMD EPYC 7702 chipset, to ensure the comparability of runtimes. For all the investigated systems, we found good convergence of the KMC simulations I-V-curves after 10^5 simulation steps. This can be seen by the cumulative charge displacement shown in the Supplementary Note 2. Regarding the occupation probabilities, we observed a convergence, measured by the distribution of interelectronic Coulomb interaction, already after 10^5 KMC simulation steps for all of the systems. These interactions are also shown and discussed in the Supplementary Note 3. It is clear that the calculation time for the RN approach depends on the size of the simulated system, but remains constant

Fig. 3 | Results for the monolayer systems.

Resulting I-V-curves from the resistor network (RN) approach with varying energetic disorder ($\sigma = 15$, $\sigma = 50$, and $\sigma = 100$ meV) as compared to a kinetic Monte Carlo (KMC) simulation performed by Lightforge. Error bars correspond to the standard deviation to the mean value, obtained from 20 replica systems. The data points taken at the same voltage are separated for clarity.

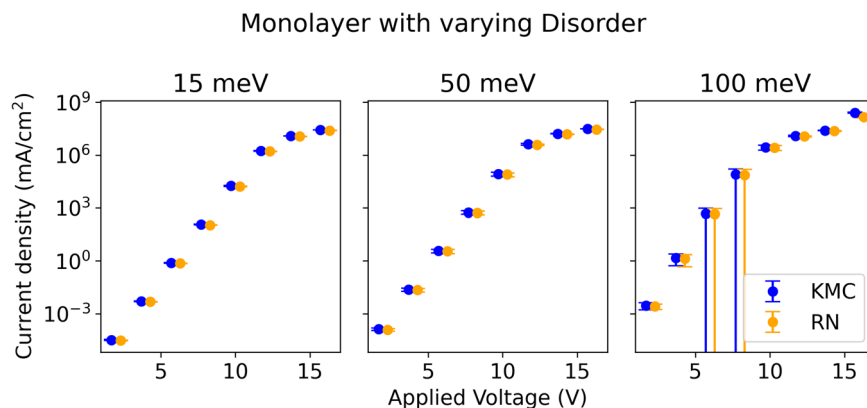
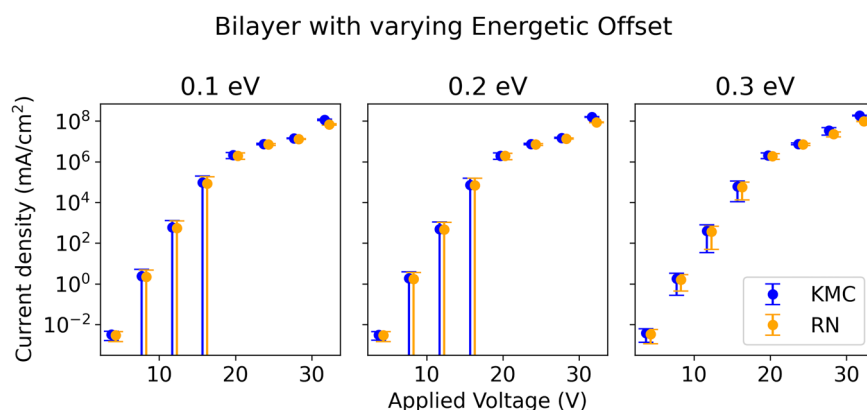


Fig. 4 | Results for the bilayer systems. Resulting I-V-curves from the resistor network (RN) approach while simulating a bilayer system with varying offset between the two layers as compared to a kinetic Monte Carlo (KMC) simulation performed by Lightforge. The energetic disorder is set to $\sigma = 100$ meV in all systems. Error bars correspond to the standard deviation to the mean value, obtained from 20 replica systems. The data points taken at the same voltage are separated for clarity.



with respect to the number of carriers since this information is encoded into the resistor values. This is expected, as larger systems result in more nodes and edges in the network. Not only do the resulting matrices become larger and more demanding to handle, but the resistor for each edge is calculated sequentially, leading to a linear dependence of the calculation time on the number of edges. In contrast, the KMC runtime is highly dependent on the number of carriers. As a result, the calculation time for the system with 15% doping using KMC is much longer than for any other tested system. This system's carrier density, in the order of 10^{20} carriers per cm^3 , results in a calculation time in the order of 10^{-2} s per KMC step. In contrast, the fastest tested system takes 10^{-4} s per KMC step at a carrier density in the order of 10^{16} carrier per cm^3 .

Regarding the number of charge carriers, the runtime of the resistor network calculation depends on two contributions: One contribution from the short KMC simulation and a contribution from the resistor network calculation itself. The first contribution is, as all KMC simulations, highly dependent on the number of charge carriers. The second contribution, on the other hand, is constant with regard to the number of charge carriers. Hence, the overall performance gain starts if the network calculation is faster than ~90% of the KMC runtime and from then onwards, the gain gets larger the more carriers are in the system, as the time for the network calculation stays constant, but the KMC calculation will take longer.

Model restrictions

Even though the RN model proved accurate in all tested systems, there are systems that can not be simulated by this approach.

Emissive layers, for example, can not be simulated as the emission of photons relies on the pairwise interaction of holes and electrons. As the RN models always only one type of carrier (assuming that this will be the dominant type - which is usually known for injection or transport layers) this pairwise interaction is difficult to reproduce. Shifting the focus from the individual emission of single photons to a time-averaged radiative flux

would result in a current sink in the network, where carriers can leave the system. Such current sinks are unaccounted for in the RN model.

Further, degradation studies model the destruction of sites under load over time²¹. The aim is to model the evolution of the device performance over time and capture shifts in carrier and exciton distributions due to the altered availability of pathways through the system. Since degradation happens through quenching events between particle pairs²², this leads to a similar problem to the one for emissive layers, since only one type of carrier is considered at a time. But further, during these simulations, sites are expected to be deactivated over time. Such time-dependent changes are difficult to capture in the RN as the underlying resistances and their topology are only constructed once.

Conclusion

In conclusion, the resistor network approach can be seen as a good approximation to the full KMC simulation in a broad range of organic layers, relevant for OLED technology. Even though a short preliminary KMC simulation remains a requirement for the parametrization of the resistor network, this model reduces the computational cost of current calculations drastically. Charge transport through organic materials in different device geometries were reproduced successfully, including geometries with internal energy barriers and doped materials. While the uncertainty is usually higher than with the KMC simulation used as a reference, the calculated values are generally in good agreement with the reference.

In addition to this good agreement, the calculation time using this approach can be reduced drastically, with regard to the full KMC simulation, in all cases presented previously. Those improvements are especially apparent in systems with high charge carrier densities. This combination of good results and a shorter calculation time makes this approach a promising extension to the pure KMC simulation. For the simulations regarding transport layers, or in general layers, where only the current throughput is needed, this approach can be used to speed up the simulation drastically.

Fig. 5 | Results for the doped systems. Resulting I-V curves from the resistor network (RN) approach while simulating a doped system with varying doping concentration as compared to a kinetic Monte Carlo (KMC) simulation performed by Lightforge. The energetic disorder is set to $\sigma = 100$ meV in all systems. Error bars correspond to the standard deviation to the mean value, obtained from 20 replica systems. The data points taken at the same voltage are separated for clarity.

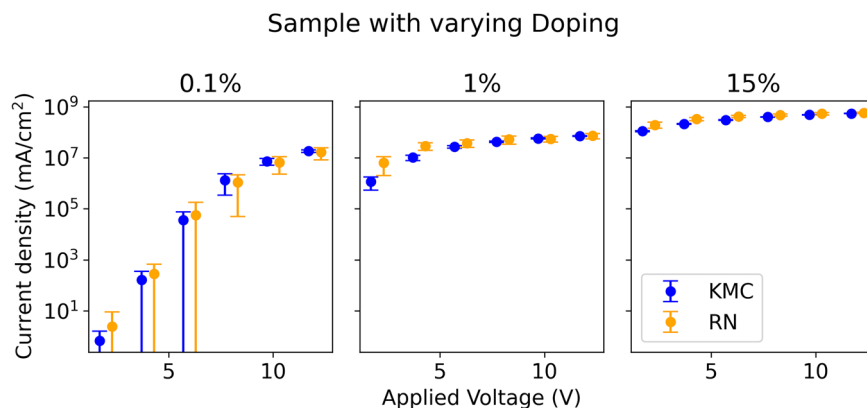
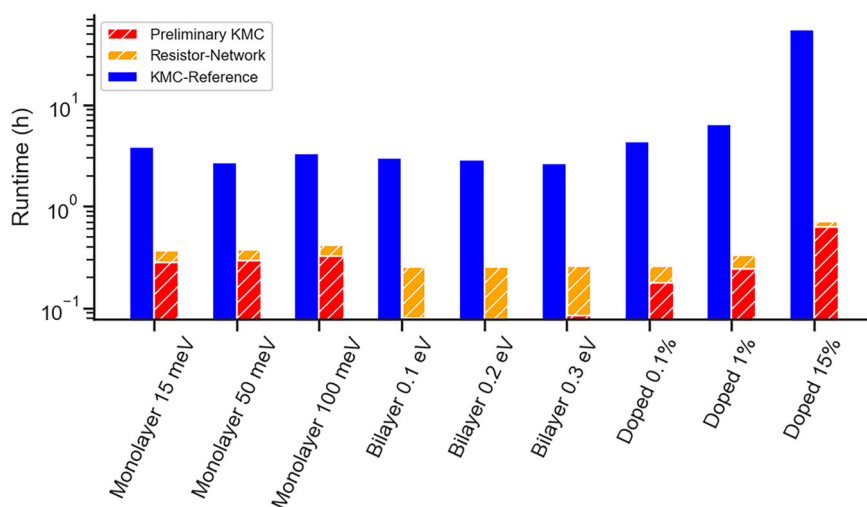


Fig. 6 | Comparison of runtimes for different methods. Runtime comparison of the combined run of a short kinetic Monte Carlo (KMC) simulation for parametrization, combined with a resistor network calculation against a full KMC simulation.



This opens the possibility for future work to replace some layers in KMC simulations of multilayer devices with cheaper Resistor Network calculations. Further, the resistor network proved able to simulate multiple layers at once, making it even more useful in this scenario.

Methods

In contrast to the hopping transport model used in KMC simulations, the resistor network approach employed here represents molecular sites as nodes connected by Ohmic resistors, with charge transfer treated as a continuous process rather than discrete hopping events. In our method, we calculate resistances between neighboring site-pairs from intra- and intermolecular properties, such as electronic couplings and molecular energy differences. A graph network is then generated, with molecular sites as nodes and intermolecular resistances as edges. The electrical current is computed using Kirchhoff's laws, which are implemented in matrix equations that are well-established in electrical circuit theory.

In this section, we will discuss the underlying charge transport model and introduce the network architecture, as well as the important matrices. After giving such an overview, we will elaborate on the computation of these building blocks. In the end, this will be brought together for the calculation of a current density in the system.

Charge transport

The premise of the resistor network model is, that it is sufficient for transporting layers to know the current flowing in and out of a certain layer. Over longer periods of time, it is more instructive to characterize these currents by time-averaged constant currents instead of sets of individual carriers arriving at distinct time steps. Thus, it is not necessary to track individual carriers

moving through these layers. This opens the question of how to calculate such constant currents even though the microscopic motion of carriers in OSC is well described by a hopping motion in distinct steps^{1–4}. Similar to the model by Cottaar et al.¹³, which uses conductances, the Resistor Network model utilizes resistances between hopping sites to mediate the currents through the system. But in contrast to our RN model, the approximations they made during the calculation of their conductances correspond to the low carrier-density limit. The RN model does not rely on these approximations and can thus also be applied to injection layers and transport layers with high carrier densities. Further, Cottaar et al. relied on the use of percolation pathways, i.e. paths of least resistance, to model the current through the system. This is well-suited for systems with high energetic disorder³. To extend the resistor network analogy also to low-disordered systems, the RN model considers not only the most dominant connections but all connections.

This resistor network model is tailored towards use on injection layers and transport layers, where the dominant charge carrier type is usually known before the simulation, i.e. it is known whether charge transport in these layers will be dominated by either hole transport or electron transport. So, the carriers of different types do not interact explicitly during the matrix construction or calculation in the resistor network. Thus, the implementation used is restricted to calculating only the transport of one kind of carrier. The calculation of currents due to transport of the opposing carrier type could be calculated in parallel by starting the matrix construction from an energetic landscape and occupation probabilities from the same preliminary KMC simulation, but corresponding to the opposing carrier type. Results presented in this paper are obtained by using only the dominant carrier types' energy landscape. This distinct treatment of holes and electrons also makes the RN not applicable to layers where excitonic processes

play an important role (i.e. emissive layers [ref. 23, p. 322ff]). The formation, movement and decay of excitons can not be represented by the use of currents mediated by a set of resistors.

Note further that the high carrier density, which is emphasized throughout this work, is not used explicitly during the calculation or to justify some approximation. The high charge carrier density is important to this study not as a conceptual limitation but as it is the regime where KMC gets especially computationally expensive. Hence, the cost-saving potential is high there, while on the one hand, these regimes are important for general OSC simulations, and at the same time, other surrogate models are not applicable.

Network topology

For the topology of the network, a square lattice with a lattice constant of $a = 1$ nm is used to represent sites^{13,24}. The lattice points correspond to sites where carriers can hop onto. The dimensions of the simulated network is set to $20 \text{ nm} \times 20 \text{ nm} \times 20 \text{ nm}$ per organic layer. For set HOMO or LUMO levels, the energy landscape of on-site potentials in such a lattice is generated by drawing from a Gaussian distribution centered around these HOMO or LUMO levels^{6,15}. The width of this distribution models static disorder in the material [ref. 23 p. 224].

Since the electronic connection between sites will be modeled by resistors depending exponentially on the hopping distance in (6), only connections up to the third nearest neighbor are considered (connections in cardinal direction, in-plane diagonal connections and three-dimensional diagonal connections)²⁵.

Starting from a list of N_{Nodes} nodes, the vector $\vec{X}$ with the on-site potentials E_i on the corresponding positions is constructed.

$$\vec{X} = \begin{bmatrix} E_1 \\ E_2 \\ \vdots \\ E_{N_{\text{Nodes}}} \end{bmatrix} \quad (1)$$

Using the coordinates from the list of nodes, an incidence matrix $\hat{A}$ is constructed connecting all considered neighbors. This matrix is of shape $N_{\text{Edges}} \times N_{\text{Nodes}}$ and contains a row for every edge and a column for every node [ref. 26 p. 88]. To encode the topological information, the entries of $\hat{A}$ are 0 if the corresponding nodes are not connected and ± 1 if they are connected as shown in Table 1. Since each row contains the information for one edge, there are two non-zero entries in each row corresponding to the start and endpoint of that edge. Which node is the start and which is the end determines in which direction current is measured (not to be confused with the direction in which a charge carrier is moving). This is implemented by choosing the measurements to be from the node with a lower index to the node with a higher index. Following the indexation of edges used for the incidence matrix, a list of conductances $\vec{C}$ is constructed. The conductance value between sites i and j is

$$C_{ij} = \frac{1}{R_{ij}}, \quad (2)$$

building upon the resistor definition that will be presented in the next section (and shown in (4)). These conductances are computed and put into the position of the corresponding edge in $\vec{C}$. This list is then transformed into main diagonal entries of the conductance matrix $\hat{C}$, which is a diagonal matrix of shape $N_{\text{Edges}} \times N_{\text{Edges}}$.

$$\hat{C} = \begin{bmatrix} \frac{1}{R_{12}} & 0 & 0 & \dots & 0 \\ 0 & \frac{1}{R_{13}} & 0 & \dots & 0 \\ 0 & 0 & \frac{1}{R_{14}} & \dots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \dots & \end{bmatrix} \quad (3)$$

Table 1 | Row-wise (edge-wise) construction of the entries in an adjacency matrix of a system with four nodes and five edges

	1	2	3	4
I				
II				
III	-1	0	1	0
IV				
V				

Here, the edge with the Roman numeral III is constructed starting at node 1 and ending at node 3.

Resistances

The resistance between molecular sites i and j is defined by:

$$R_{ij} = \frac{\Delta E_{ij}}{q^2 \cdot \left(k_{ij} \cdot p_i \cdot (1 - p_j) - k_{ji} \cdot p_j \cdot (1 - p_i) \right)}, \quad (4)$$

where ΔE_{ij} is the energy difference between the sites, q is the elementary charge, k_{ij} is the charge hopping rate (also used in the KMC approach) and p_i is the occupation probability of site i . This use of occupation probabilities is similar to the work of ref. 27. A hop between sites is more probable if the start-site is occupied and if the target-site is not already occupied. This behavior reflects conservation of charges and Coulomb repulsion, effectively forbidding double occupation of sites.

In this work, the hopping rate was computed using the Miller-Abrahams formula^{6,9,12,28}:

$$k_{ij} = \frac{\pi}{2 \cdot \hbar \cdot k_B T} |J_{ij}|^2 \begin{cases} \exp\left(-\frac{\Delta E_{ij}}{k_B T}\right) & , \text{ if } \Delta E_{ij} > 0 \\ 1 & , \text{ otherwise.} \end{cases} \quad (5)$$

For the electronic coupling, a parametric model is used⁹:

$$J_{ij} = j_0 \cdot \exp\left(-2 \frac{\Delta r_{ij}}{\delta_0}\right), \quad (6)$$

where Δr_{ij} is the distance between site i and j , δ_0 is the overlap decay length and j_0 being a coupling constant. The chosen values for these parameters can be found in the supporting information.

The energy difference in the Miller-Abrahams formalism consists of three terms¹:

$$\Delta E_{ij} = \Delta E_{ij}^0 + q \Delta x_{ij} \cdot F + \Delta \Phi_{ij}, \quad (7)$$

where the first term represents the difference in HOMO or LUMO levels for hole or electron transport¹, it highly depends on the structural and energetic disorder present in the system. The second term introduces the energy shift, induced by an external electric field in the x -direction. The last term contains all electrostatic contributions to the energy difference. This is the energy difference between the states before and after a potential hop, resulting from the change in distance between carriers as every charge carrier is interacting with every other charge carrier via Coulomb interaction^{29,30}.

Since, the Coulomb interaction between carriers is the key mechanism in the formation of a stable distribution of occupation probabilities, the influence of the Coulomb interaction energy contributions and the occupation probabilities are closely linked. To avoid double-counting, the Coulomb interaction in ΔE_{ij} was omitted for its use in (4) and (5).

Further, the currents in the system are calculated once during the simulation. Hence, the resistors in this network are only constructed once

and do not change during the simulation. Therefore, all resistors must be defined in a time-independent manner. This proves challenging for the occupation probabilities p_i and the underlying Coulomb contribution to the energy differences $\Delta\Phi_{ij}$, as both depend on the current charge distribution and therefore change with every hop. Thus, instead of using the fluctuating time-dependent quantities, the time-averaged occupation probabilities per site are used. To calculate those averages, a short preliminary KMC simulation is performed to find a steady state. Here, the system has found a state with stable percolation pathways. Hence, the average occupation probabilities and Coulomb interactions are equilibrated. Plots showing the equilibration of the Coulomb interaction can be found in the Supplementary Note 3. A stable distribution of Coulomb interactions was taken as an indication of a system in a steady state. Thus, after investigating such equilibrations, the length of the preliminary simulation was set to 10% of a full KMC simulation. This is in line with literature suggesting, that KMC has to run for 10% of the full runtime to become independent from the starting configuration⁶.

With the steady state at hand, the occupation probabilities are extracted from the Lightforge output for each molecular site and used in (4) for the corresponding site in the Resistor Network. In the KMC, the occupation probability is calculated as a time average by using the number of steps in the Markov chain for which a given site was occupied during the simulation. At this point, the resistor network is represented as a graph object with attributes on its nodes and edges. Here, the information of occupation probability is stored as a node attribute at each node and accessed later during the construction of the resistors. Also, the distribution of Coulomb interaction contributions to the energies in the system has converged at this point.

Before deciding on the use of occupation probabilities instead of the Coulomb interaction energy contributions, testing the use of both in conjunction highlighted that only one of them should be used to avoid potential double-counting errors. A discussion of these tests is presented in the Supplementary Note 3. Nevertheless, Coulomb interactions are implicitly accounted for through the use of occupation probabilities in (4) from the preliminary KMC simulation. The preliminary KMC produces a self-consistent description of the local carrier distribution under explicit consideration of the Coulomb interaction between these carriers. In particular, the prohibition of double occupancy is enforced in (4) by suppressing current toward highly occupied sites through the terms $(1 - p_x)$.

To use the Coulomb interaction instead of the occupation probabilities, future work could track the pairwise energy difference during the equilibration phase, then generate distributions for multiple system slices perpendicular to the applied field and for different resistor orientations. This would allow sampling energy differences per resistor from corresponding distributions, enabling investigation of deviations, particularly in regions with permanently high carrier densities like near interfaces, due to the explicit Coulomb interaction.

We expect neglecting $\Delta\Phi_{ij}$ may affect the IV curve if it alters the resistance of bottleneck resistors. This might, for example, happen if a large charge buildup would push charges over a barrier or if fluctuations in $\Delta\Phi_{ij}$ consistently help to detrapp charges. While the p_i in (4) model these effects trendwise, there might be nominal differences, which may become particularly important in bottleneck resistors. As our test systems showed very good agreement between the RN model and KMC, we deduce that these kinds of effects are not yet relevant for the tested device architectures.

Calculation of the current density

After construction of the network with the corresponding vectors and matrices, the currents over all connections can be obtained by matrix multiplication [ref. 26 p. 110ff]

$$\vec{I} = -\hat{C} \cdot \hat{A} \cdot \vec{X} \quad (8)$$

with a vector $\vec{I}$ containing the currents flowing over each connection, the vector $\vec{X}$ containing the on-site potentials for each site, an incidence matrix $\hat{A}$ containing the topology of the network and a diagonal conductance

matrix $\hat{C}$ with the conductances of the connections on the main diagonal. In an equilibrated system, currents obtained from this equation will fulfill Kirchhoff's circuit laws [ref. 26 p. 110ff]. The indexation of currents in $\vec{I}$ is the same as used previously for the respective edges in the incidence matrix and the ordering of the entries in the conductance matrix.

$$\vec{I} = \begin{bmatrix} I_{12} \\ I_{13} \\ I_{14} \\ \vdots \end{bmatrix} \quad (9)$$

An illustration of the construction and use of these matrices is provided in the Supplementary Note 4.

Current densities. To make the results from our method comparable with other methods, current densities are used instead of currents flowing over individual connections. Thus, the partial currents flowing through an end layer of the sample are used to calculate the current density. Thus, the edges connecting nodes from the second to last and last layer are selected, and the corresponding entries in $\vec{I}$ are added up. Then this sum is divided by the surface of the observation cross-section. This includes explicitly not only connections in cardinal direction, but also diagonal connections through the surface of observation.

Error estimation. The KMC software package Lightforge is used to perform the reference and the preliminary simulation. Here, 20 replicas of the system, started from different random seeds, are used. Lightforge uses the results of these 20 replicas for an error estimation. As the preliminary KMC simulation results are also in 20 replicas, the results from these different replicas are used to run 20 resistor network calculations. These results are then also used for an error estimation for the resistor network method.

Resulting plots presented in this paper feature the mean of 20 calculations, corresponding to 20 replicas from the preliminary KMC simulation, and error bars representing the standard deviation to this mean.

KMC

The kinetic Monte Carlo (KMC) method is used as a reference and for the preliminary simulation of the Resistor Network method. The preliminary simulation is used to obtain occupation probabilities for the sites in the Resistor Network. Both methods share the same topology, presented before for the resistor network. While the resistor network uses constant currents in a steady state, KMC uses the motion of individual carriers to model the time evolution of a sample and can thereby also be used to find steady states⁷.

During the simulation with KMC, the system evolves between snapshots by individual events involving charge carriers and other particles situated on the sites^{1,6}. In each step, the transition probabilities to all possible states have to be calculated. This includes the costly calculation of interaction terms with all carriers. Hence, this is a bottleneck to the simulation of systems with high charge-carrier densities. Nevertheless, this method is able to accurately model the processes relevant to charge transport in OSC as the carriers are localized to individual sites and can hop from one site to another by tunneling between the sites^{1,6,7}.

Data availability

Data is archived and available from the authors upon request to the corresponding author (W.W.). The input files for the KMC simulation of the individual systems, presented in this study, can be found in the supplementary material.

Code availability

All code used to implement the methods and reproduce the findings presented in this paper is publicly available at <https://gitlab.kit.edu/utxxi/>

ResistorNet/-/tree/main/Final. The version used for all computations in this work is archived on Zenodo³¹.

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Author contributions

A.D.Ö. and W.W. conceived the idea; F.S. contributed his knowledge on the KMC software used; O.P. wrote the resistor network code and carried out the simulations; and O.P. prepared the manuscript with the inputs from all the co-authors.

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Competing interests

Author F.S. was employed by Nanomatch GmbH. W.W. holds shares of Nanomatch GmbH. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Additional information

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